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Publication Editors

Eleanor M. O'Neal

Peter W. Murphy

**Laser Program
Annual Report
1987**

**Lawrence Livermore
National Laboratory**

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Laser Program Annual Report 1987

**Lawrence Livermore
National Laboratory**

Preface

This report presents the unclassified activities and accomplishments of the Inertial Confinement Fusion (ICF) and the Advanced Laser Development (ALD) elements of the Laser Program at the Lawrence Livermore National Laboratory (LLNL) for the calendar year 1987. The classified aspects of our work are reported elsewhere. The LLNL Laser Program is supported by the United States Department of Energy. Some of the activities in Advanced Laser Development are supported by agencies of the United States Department of Defense.

Our objective in preparing these Laser Program Annual Reports is to disseminate the results of our work for the benefit of the inertial confinement fusion, plasma physics, and laser communities. We attempt to provide, in one volume, sufficient detail to allow full understanding, to permit critical technical evaluation, and to make it possible for others to reproduce or extend our work. Therefore, we present here new concepts, theoretical analyses, computational results, the design and configuration of experiments, and analytical data—the results of our year's research.

We have organized this report into major sections that correspond to our principal technical activities. Section 1 notes the highlights of the year's technical accomplishments and briefly summarizes Program resources and facilities. Section 2 relates our work in target theory, design, and computer code development. In Section 3, we discuss the Nova laser program, including experiments, diagnostics, and data acquisition and management, as well as the laser system and its management. Our effort in the area of ICF target technologies and materials is described in Section 4, and in Section 5 we discuss our work on basic laser research and development. Section 6 presents our research on solid state lasers for high-average-power applications. Finally, Section 7 reviews the ICF applications we have studied this year.

L. W. Coleman

Acknowledgments

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This volume of the LLNL Laser Program Annual Report summarizes the technical work during 1987 of some three hundred scientists and engineers and the many technicians who assisted them. Their impressive scientific and engineering contributions speak for themselves in the ensuing pages, but we also wish to acknowledge their accomplishments and to thank them for their cooperation in writing the articles. The assistance of the scientists who served as section editors, Steve Weber, Mike Campbell, Blanca Haendler, Howard Powell, Mike Summers, and Mike Tobin, was especially valuable, and we thank them for it. We also gratefully acknowledge the Program's secretarial staff for doing the demanding technical typing and other clerical work required to produce the manuscripts.

After the scientists and engineers had written their drafts, there remained the onerous task of preparing them for publication. As in the past, this was ably accomplished by members of the Laboratory's Technical Information Department. Publication Editors Eleanor O'Neal and Peter Murphy solicited the scores of manuscripts and coordinated the efforts of the many technical editors, artists, composers, and printers engaged in the project. Editors Bob Kirvel, Elaine Price, Judy Prono, Tim Peck, Sondra Reid, Lisa Wallerstein, Bill Wright, and June Canada polished the language of the manuscripts. Compositor/proofreader Jill Sprinkle coordinated the difficult typesetting and proofreading with the help of Jane Winter and Maria Fogle. Penny Keenan designed the cover and divider pages. Artists Ellen Baldwin, Ken Ball, Ron Collins, Janet Crampton-Pipes, Henry Kidd, Duane Hoepker, George Kitrinos, Kirk Hadley, Ron Strawser, Regina Nagel, Merry Oman, and Mark McDaniel and his co-workers prepared the artwork under the vigilant eye of Art Coordinator Wilma Leon. Layout artists Mary Louise Alviso and Kim Lanto helped prepare the printer's boards, and Bill Fulmer arranged for commercial printing. Without the special talents, dedication, and good will of such an outstanding publication staff, this Report could not have appeared in its present form.

All the people named above and others who participated in this effort deserve and have our sincere thanks.

L. W. Coleman

MASTER

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Laser Program Overview

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Section Editor: L. W. Coleman

Laser Program Overview

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Section Editor: L. W. Coleman

Laser Program Overview

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Laser Program Overview

Introduction

Erik Storm and L. W. Coleman

In 1987, the Laser Program at Lawrence Livermore National Laboratory (LLNL) made outstanding progress in its research on inertial confinement fusion and high-average-power lasers. The continuing objectives of the Inertial Confinement Fusion (ICF) element of the Laser Program are to understand and develop the science and technology of ICF so that its full spectrum of capabilities and benefits can be realized. This potential will be fulfilled with the achievement of well-diagnosed, high-yield (100- to 1000-MJ) fuel-capsule implosions in a laboratory. Microfusion in this range will make it possible to fully utilize ICF in research supporting U.S. defense programs: the study of weapons physics and vulnerability, lethality, and effects experiments. At the same time, this achievement of high-yield microfusion with an ~ 10 -MJ driver will demonstrate the scientific feasibility of ICF as a candidate technology for electrical power production and for other civilian applications.

This year we continued to conduct experiments and develop diagnostics in the laboratory with the Nova laser and in underground tests at the Nevada Test Site. (The Nevada tests were performed as part of the Halite/Centurion program, a collaborative effort between LLNL and Los Alamos National Laboratory.) Our Program continued to make advances in the theory and design of targets; in target fabrication and materials science; in laser and optics research and development; and in ICF applications studies.

We utilized the Nova laser very productively to add to our data base in several technical areas. Nova's ten beams have routinely and reliably provided as much as 20 to 25 kJ of energy in (nominally) 1-ns pulses of $0.35\text{-}\mu\text{m}$ -wavelength light, a capability that enabled us to add substantially to our knowledge of the physics of indirectly driven targets. We conducted experiments in the physics and scaling of hohlraums, accumulated a large body of data on x-ray conversion with $0.35\text{-}\mu\text{m}$ laser light, and greatly added to our understanding of the drive symmetry and capsule-convergence ratios accessible with the indirect-drive approach to ICF. The fusion: fuel-confinement conditions we achieved continued to improve, and we obtained record-setting neutron yield (a measure of fusion efficiency) with relatively large, directly driven targets. In addition to the excellent experimental conditions and high-quality data provided by Nova, we have at our disposal important target-design tools and models now developed to the point that we can accurately calculate almost all the main features of the experiments.

This year, by emphasizing new experimental and diagnostic techniques on Nova, we significantly enhanced our diagnostic capabilities. We began to exploit the capabilities of a high-resolution, time-resolved x-ray microscope for implosion and hydrodynamics studies. Two new, four-frame, x-ray framing cameras with better than 200-ps resolution were used for a variety of imaging and spectroscopic measurements. We also developed new diagnostics for characterizing the imploded capsule core and implosion times.

We completed a set of laboratory x-ray laser experiments that made progress toward the biologically interesting "water window" between 22 and 44 Å by demonstrating gain, with nickel-like ytterbium, at a lasing wavelength of 50 Å. In a separate experiment, using the 206- and 209-Å selenium x-ray laser lines, we demonstrated the potential for building an x-ray laser cavity and also used the selenium laser to produce the world's first hologram made with an x-ray laser.

In our work on target fabrication and materials science, we continued to develop foam capsule materials for cryogenic, directly driven ICF applications. The foams must meet stringent requirements for such properties as density, cell size, mechanical properties and radiation exposure. We improved the characteristics of polystyrene foams and, in an effort to obtain even smaller cell size, pursued the development of resorcinol-formaldehyde foams. We also initiated study of a promising new foam material, cellulose acetate.

Our research and development of ICF lasers and optics included new technology for laser glass and edge cladding, which would be applicable to replacement disks in Nova's 31.5- and 46-cm amplifiers. In conjunction with commercial laser-glass manufacturers, we have developed new manufacturing techniques, reducing the number of damaging platinum inclusions in the new glass to an inconsequential level (more than a thousand-fold improvement over Nova's original glass). A new, less expensive edge cladding for the glass disks has been developed and thoroughly tested. Our laser and optics research and development also addressed the key technologies with the greatest potential for reducing the cost of a multimegajoule, solid state laser concept developed last year. The areas on which we concentrated were amplifier efficiency, improved optical materials and coatings, improved nonlinear optical materials, and crystalline laser materials. In addition, we conducted basic studies of fundamental but longer term importance for laser development.

The High-Average-Power Solid State Lasers component of the LLNL Laser Program has continued a broad-based effort to develop technologies to make average-power solid state lasers usable in new and demanding applications. We have concentrated on developing laser and nonlinear optical materials, developing computational design tools, validating them on experimental test beds, and projecting this technology to specific missions. Our work has produced strong, low-loss laser glass and significant advances in the understanding and technology of crystalline optical materials and their growth. In the experimental areas, we have learned most of the physics and engineering issues germane to average-power, zig-zag lasers in slab geometries. Capitalizing on the program's state of technological advancement, we are also developing a solid state laser for a specific, near-term application: submarine laser communication.

Our ICF applications studies focused on near-term design issues for a Laboratory Microfusion Facility (LMF), in particular, the design of the experimental area in an LMF. We concentrated on analyzing the technical design requirements for a successful LMF and on evaluating ways to meet these requirements cost effectively. In addition to the LMF studies, we examined in more detail the tritium safety issues associated with

the Cascade ICF reactor concept and radiological issues concerning targets in heavy-ion driven reactors. We have also updated the economic requirements for a heavy-ion accelerator designed to drive a reactor.

The successes we have achieved in this year's research have significantly strengthened our conviction that we can meet the goals of ICF. Our experimental and calculational advances support this confidence that high-gain ICF can be achieved in the laboratory with a 10-MJ, short wavelength laser and that the technologies to realize that driver are within reach.

Progress and Accomplishments

Target Design and Experiments

In the LLNL Laser Program, 1987 was a very productive year for target design, modeling, experiments, and diagnostics work. Our near term activities included experiments directed toward completing the ICF technical data base and theoretical studies to develop models and calculational techniques that adequately explain and predict experimental results. Data obtained both from experiments with the Nova laser and from the Halite/Centurion program have confirmed the accuracy of our models.

During this year, Nova operated as a productive experiment facility, reliably providing 20 to 25 kJ of 0.35- μm light in 1-ns pulses from its 10 beams. This shorter wavelength light has virtually eliminated the issue of hot electrons, which were a major detriment at wavelengths of 1 μm and larger, and allowed us to concentrate on issues of radiation transport and hydrodynamics. Our emphasis this year was on developing new experimental and diagnostic techniques for a better understanding of target performance and of the laser itself. In addition to performing various target and hohlraum experiments, we developed techniques for quantitatively measuring mixing between dissimilar accelerated materials. We also conducted an extensive series of experiments to understand the physics of x-ray production by high-Z plasmas.

We advanced our diagnostic capabilities in several areas. The 22 $\times$ Wölter

microscope was activated and now supplies high-resolution, time-resolved images for implosion and hydrodynamics studies. We further added to our x-ray diagnostic capability with the installation of two new four-frame, microchannel-plate, x-ray framing cameras with <200-ps resolution. These will be used for both imaging and spectroscopy. In the area of neutron diagnostics, we began time-of-flight measurements of secondary deuterium-tritium neutrons to obtain fuel *pr* in compressed targets. We also began using GaAs and scintillator microchannel-plate detectors to measure target implosion times.

Target Materials Development

We investigated three foam materials for possible application to cryogenic, directly driven, ICF targets: polystyrene (PS), resorcinol-formaldehyde (RF), and cellulose acetate (CA). Our goal is to develop a material with a density <50 mg/cm³, a cell size <4 μm , and a pore size <1 μm , that is easily fabricated into shells and that is compatible with liquid deuterium-tritium.

As a result of a series of statistically designed experiments on the formulation, polymerization, and drying processes of the foam, we can now repeatedly produce foams of 85 mg/cm³ density and with cell sizes of 2 to 3 μm . With RF foams, we gained a better understanding of gel formulation and processing, which enabled us to reduce foam density from 150 to 20 mg/cm³. To further improve RF mechanical properties, we investigated carburization in an inert atmosphere at 1100°C. This produced an all-carbon foam but also caused some shrinkage, increasing the density to 41 mg/cm³.

Cellulose acetate foams produced by supercritical extraction resulted in densities of 30 to 150 gm/cm³. Thus we now have three excellent candidate materials for use in future cryogenic targets.

Laboratory X-Ray Lasers

Using the two-beam target chamber, we made significant progress toward the "water window" between 22 and 44 Å, the wavelength range required for biological holography. We demonstrated laser gain at a 50-Å wavelength using the 4d → 4p transition in nickel-like ytterbium. We believe that in the future it will be possible to obtain significant gain at wavelengths less than 44 Å using neon-like tungsten or rhenium. We also believe that similar success can be achieved with hydrogen-like aluminum ($n = 2$ to 3 recombination schemes driven by a 10-ps laser pulse), and we are now in the process of building such a short-pulse oscillator for Nova.

In separate experiments in the two-beam target chamber, we demonstrated laser gain at 206- and 209-Å wavelengths using selenium. With this laser and x-ray beam splitters, we produced the world's first x-ray hologram.

Laser Science and Technology

Our primary goal for 1987 was to address the issues that led us to limit the output of our large amplifiers on Nova to less than original design levels. Working with the commercial suppliers of glass for the Nova optics, we developed new manufacturing techniques that reduce the platinum inclusions by over three orders of magnitude. We also developed an on-line quality assurance process that will ensure that all glass procured in the future will meet design specifications for particulate inclusions. This has enabled us to commit ourselves to replacing all of the glass in the 31.5- and 46-cm amplifiers by the end of calendar year 1988. When this has been done, Nova will be able to operate reliably at its full design energy. In addition, a new edge-cladding technique was developed for suppressing amplified spontaneous emissions in large disks. This technique, which is cheaper

and more convenient, has been tested in a prototype amplifier for over 2000 shots. The new cladding will be used on the inclusion-free glass to be installed next year.

In support of studies initiated this year on the feasibility of a high-gain ICF machine, known as the Laboratory Microfusion Facility (LMF), we concentrated on identifying ways of reducing the cost of the driver system. We looked at four areas where significant gains could be made:

- High-efficiency solid state amplifiers.
- High-damage-threshold optical materials and coatings.
- Advanced nonlinear optical materials.
- Advanced crystalline laser materials.

The results of our studies in these areas led us to project that a 10-MJ LMF using Nd:glass lasers could be operational within 10 years at a cost of approximately \$750 million.

We continued our research in the area of advanced crystalline materials for use in solid state laser applications, including ICF power plants, where high pulse-repetition rates are required. We identified a number of new materials and developed advanced methods for growing large, homogeneous crystals. We also worked on a variety of topics with laser materials, including nonlinear indexes of refraction, spectroscopic properties of impurities, and damage-inducing interactions of light with optical surfaces.

High-Average-Power Solid State Lasers

We continue to pursue a broad range of topics aimed at developing high-average-power lasers for a variety of advanced applications. Part of this effort supported the development of glass free of platinum inclusions, as noted above. Working with glass suppliers, we also developed practical new surface-finishing techniques and surface-ion treatments that increase the tensile strength of laser-glass surfaces by factors of 5 to 10. In our crystal material development program, we gained a better understanding of the Czochralski process, which has enabled us to grow large Nd:GSGG crystals, and we are now working on ways of

reducing energy losses caused by light absorption in this material. We also made progress in growing flat Nd:YAG, although much remains to be done with this material.

Our laser experiments explored a number of concepts. Our research on zig-zag slab lasers furthered our understanding of thermally induced wavefront aberrations. We worked on gas-cooled slab lasers to understand hydrodynamics, heat transfer, and optical interactions. Finally, we carried out a number of critical tests on electro-optic switches. We now understand the physics and engineering issues well enough to integrate these components into an operating laser during the coming year. Work is also proceeding on rare-earth-doped crystalline and glass lasers for use in submarine laser communication.

ICF Applications Studies

Stimulated by the increased national interest in the Laboratory Microfusion Facility (the next generation of ICF machine), the ICF Applications Group shifted its attention from long-term studies to near-term design. Collaborating with other members of the ICF community on a committee created by DOE's Inertial Fusion Division, we helped develop the design requirements for such a facility. Our specific responsibility in this cooperative effort was the experiment area of the LMF. We developed vaporization and condensation models for target chambers exposed to 1000- to 1500-MJ energy releases. We studied beam quality degradation during transport through protective gas clouds and x-ray damage thresholds for fused silica lenses and windows. We also looked at neutron activation of impurities in aluminum. Finally, because cost is of such concern in the LMF project, we evaluated the possibility of using the Nova target chamber as part of an LMF. In the process, we developed a better understanding of the behavior of hot gases released during a laser shot; these gases are of interest because they may help protect the final optics.

We also conducted several studies related to ICF reactor plants for producing

electricity. One study on the tritium cycle concluded that it will be possible to maintain an inherently safe operation of a combined, collocated reactor and target fabrication facility. The other studies were directed toward the use of lead as a tamper in heavy-ion-driver systems. These studies concluded that lead would not pose a significant health or environmental hazard, and that using lead depleted of ^{204}Pb would significantly reduce what hazards from activation do exist. Finally, we updated our 1985 study of the economics of heavy-ion drivers and concluded that a 10-MJ driver with target gains of 50 could be competitive for a 100-to 2000-MW_e power plant.

Author: L. W. Coleman

Program Resources

Financial resources for the LLNL ICF Program in fiscal year 1987 totaled \$67.6 million, of which \$64 million was in operating funds and \$3.6 million was available for capital equipment expenditures. Figure 1-1 shows the resources available to the ICF program over the last ten years. The figure shows the operating and equipment funds expressed in constant FY-87 dollars in addition to the as-spent dollars to show the effects of inflation. The adjustment for inflation was made using after-the-fact estimates of the average decrease in purchasing power (including both manpower and procurement). These curves show that since 1981 our budgets have decreased in purchasing power: by 23% in operating funds, by 22% for manpower, and by 47% in capital equipment funds.

While the impact of any one year's decrease in purchasing power is difficult to assess, the cumulative effects are apparent. In the late '70s and early '80s LLNL conducted a balanced program divided equally among the three critical elements necessary for long-term ICF success: driver research, laboratory beam/target interaction studies, and the classified Halite program. In the latter half of the '80s, we have not been able to afford such a balanced program. Despite this, we have

Program Facilities

Fig. 1-1. Actual funds (solid colored lines) expended are compared to constant 1987 dollars (dashed colored lines) for the past 10 years, showing the effects of inflation on the purchasing power of the 1987 dollar.

made progress each year by putting one of the three critical elements, in effect, on hold while concentrating on the other two elements. However, it will be difficult to keep up this juggling act indefinitely.

The impact of the decrease in capital equipment funds has been even more drastic. The ratio of equipment funds to operating funds has fallen to 5.6%. This has severely hampered our ability to outfit new laboratories with the equipment necessary to evaluate or develop new materials for laser or target design. In addition, though the Nova laser itself is a unique research tool, it contains hundreds of pieces of standard equipment that are commercially available and must be purchased with equipment dollars. The equipment resources available in FY 87 were insufficient to purchase the number of spares we needed to assure smooth operation of Nova in the event of many types of failure. As failures occur, we can expect to see disruptions in some Nova operations. It is critical that this trend toward decreasing capital equipment resources be reversed.

Author: T. M. Barlow

Program Facilities

In the Inertial Confinement Fusion Program, uniform and controlled facilities growth was maintained in 1987, supporting a work force of about 500 full-time employees and contractors. The space needed for this scientific effort requires more than 450,000 ft² of office and technical space in ten permanent buildings and 50,000 ft² in 20 temporary structures.

The volume of ICF construction and maintenance work on facilities continued to grow in 1987. With the completion of many major building projects in prior years, new emphasis evolved in 1987 toward a larger number of tasks of lesser magnitude. This new emphasis was highlighted by additions to several existing permanent structures, comprehensive remodeling and renovation of existing laboratory and office spaces, and a continuing increase in temporary work areas.

Among the facilities projects completed were the addition of new laboratory facilities and the major mechanical equipment overhaul in Building 391, and the activation of laboratories in the recently completed Building 162 (see Fig. 1-2).

Fig. 1-2. Map of Laser Program area at LLNL.

Building construction and maintenance, including remodeling of permanent and temporary buildings, is managed by the Laser Facilities Group. Group services for the ICF Program include project management and supervision, design and specification, planning and estimating, and material procurement. These services required the initiation and completion of construction projects valued at almost \$3 million.

Major Laser Facilities Group projects completed in 1987 include:

1. Direction and coordination of the Laser Facilities Team Crafts Group that included 60 skilled multi-craft people.
2. Management of procurement activities that included approximately 800 transactions, bills of materials, and purchase orders, with a total value of more than \$500,000.
3. Initiation of 120 job orders for various programmatic projects, with a total value of approximately \$1.1 million.
4. Completion of a document control computer network between Buildings 381, 481, and 482.

5. Design and related construction support for the following ICF projects:

- a. Completed the Kilohertz Damage Testing Laboratory in Building 162 to provide additional technical work area.
- b. Completed a \$123,000 overhaul of mechanical systems at Building 391.
- c. Installed optical viewing windows in the new Free-Electron Laser (FEL) Laboratories.
- d. Completed the preliminary facilities planning for the Large Neutron Scintillator Array Diagnostic Chamber.
- e. Made mechanical and electrical modifications to Buildings 162, 381, and 391.
- f. Completed the Moving Melt-Zone Furnace Facility in Building 162.
- g. Installed an exterior canopy at Building 391.
- h. Renovated offices in Building 481.
- i. Removed two 12,000-gallon underground fuel tanks from Buildings 381 and 391.

Author: R. A. McCardle

Target Design

2

Section Editor: S. V. Weber

Target Design

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Section Editor: S. V. Weber

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Target Design

Introduction

J. D. Lindl

The year 1987 was a period of outstanding progress and success in design and modeling for both the Nova and Halite Programs.

Our models developed over the last several years have proved capable of accurately calculating almost all of the main features of Nova experiments including laser deposition, hohlraum temperatures, temperature gradients, and shock pressures. They also accurately calculate capsule yields, fuel temperatures, fuel pr , neutron and x-ray pinhole images, and symmetry. These experiments culminated in our ability to design capsules with radial convergence ratios in excess of 30 that achieved near one-dimensional (1-D) yields.

These experiments and the very close correspondence with the modeling take us a major step toward demonstrating that lasers can produce the hohlraum conditions required by high gain capsules.

This rapid progress has been made possible, to a significant extent, by the fact that hot electrons, which were a dominant focus of our research with 1- μm and longer wavelength lasers, have been almost a nonissue at 1/3 μm . This has allowed us to concentrate on precision radiation transport and hydrodynamics experiments.

Most of the hohlraum experiments have used 1-ns nominally constant power pulses, with no pulse shaping. Pulse shaping is essential for high gain targets, and it will be the focus of Nova experiments for the next 2 to 3 years. Pulse shaped hohlraums, because of the longer pulse length, require more detailed understanding of the hohlraum plasma conditions.

The combination of results from our Nova hohlraum experiments and the Halite/Centurion Program gives us confidence that high gain can be achieved with a 10-MJ laboratory driver.

One of the attractive features of x-ray-driven capsules is that they are driver independent if the driver can produce x rays with the required temporal, spectral, and spatial characteristics. We accumulated a large body of data in 1987 on x-ray conversion using 1/3- μm laser light. Using XSN, our current standard atomic physics code, we can quite accurately model the magnitude and spectral features of the conversion process as a function of laser intensity and material for nanosecond pulses. Our current model differs from our older models in that it invokes bound-bound and bound-free rate multipliers of 0.5 and 0.25 respectively. The primary deficiency of this model is its current inability to reproduce the laser pulse length dependence of the x-ray conversion efficiency. The experimentally observed conversion efficiency increases more rapidly than predicted for multnanosecond pulses and decreases more rapidly than predicted as the pulse length is reduced to 100 ps. Our efforts to improve the atomic physics models in XSN are now being incorporated into LASNEX. A local thermodynamic equilibrium (LTE) version of statistical configuration accounting (SCA) is now operational and a non-LTE version is in the testing phase.

We continue to view heavy-ion drivers as the primary long-term alternative to lasers for driving high gain targets. During 1987, we made significant progress in quantitatively assessing the issue of target charging

during irradiation. We showed that at least two mechanisms will keep the target potential low. If the target is covered with as little as a monolayer of hydrogen-containing material, the protons expand to a sufficiently large radius that the potential will be kept at a small fraction of the beam energy. Also, even with low densities of residual gas in a chamber, ionization by photons emitted by the target as it heats up will produce enough electrons to assure a low target potential. Photoionization will also affect the charge state of the incoming ions and adversely affect the beam focusability. We are still assessing this effect, but preliminary results indicate a relatively small impact.

Various beam-target interaction collective instabilities have been postulated as potential sources of preheat for ion beam drivers. Order-of-magnitude estimates done in previous years indicated that the effect of such instabilities would be small. Particle simulations carried out in 1987 confirm that both two-stream and Weibel velocity anisotropy instabilities saturate at negligible levels in the target corona.

In the laboratory x-ray laser work during 1987, we continued to make progress toward the water window between 22 and 44 Å needed for biological holography. We demonstrated gain at a wavelength of 50 Å. This was achieved with a 4d → 4p transition in nickel-like ytterbium. During 1988, it may be possible to get below 44 Å for nickel-like tungsten or rhenium. We believe that similar success can be achieved in a hydrogen-like aluminum $n = 3$ to 2 recombination scheme driven by a 10-ps laser pulse. Such a short pulse oscillator is being built for Nova. Since at present Nova's shortest pulse is in the 100-ps range, we are experimenting with recombination systems at that pulse length to test our modeling capabilities, but we do not expect large gain.

In summary, we are now able to do inertial confinement fusion (ICF) experiments and model the results with a precision that has been the goal of the past decade of development. During the next two years, we will concentrate on demonstrating precision pulse shaping on Nova and on applying what we have learned to develop detailed designs for a high gain facility, the Laser Microfusion Facility (LMF), to follow Nova.

ICF and X-Ray Laser Target Design

Introduction

The bulk of our 1987 ICF laser target work on Nova consisted of extremely successful indirect-drive implosions. In addition, a few direct-drive targets, such as gold disks, were shot and are described here. Exciting advances were made in soft x-ray lasing using the nickel-like 4d → 4p scheme. This work, along with other developments in soft x-ray laser design, is also reported here.

The first two articles describe a new calculational model for predicting x-ray

conversion efficiency. It differs from older models in that it invokes bound-bound and bound-free multipliers of order 0.5 to 0.25. The new model correctly calculates the conversion efficiency vs intensity and element, as well as spectral features such as relative amounts of M- to N-band radiation. Discrepancies between data and the model's predicted conversion efficiency at short pulse lengths are explored in a variety of creative ways. Other conversion efficiency vs detailed pulse-shape dependencies are dealt with in the third article.

Understanding the Rayleigh-Taylor and Richtmyer-Meshkov instabilities remains a crucial milestone in the path towards high-gain target performance. Hydrodynamic

shock tube experiments, with rapidly extracted membranes separating fluids of different densities, have been performed at the California Institute of Technology (Caltech). Modeling that simulates those experiments far into the nonlinear regime is described in the fourth article, "Two-Dimensional Simulations of the Richtmyer-Meshkov Instability in Shock-Tube Experiments."

Our modeling of soft x-ray laser experiments has steadily increased in sophistication. The fifth article, "Analysis of Data for Neon-Like Soft X-Ray Lasers," makes the point that all the sophistication we can muster must be brought to bear to explain the rich harvest of detailed observations. Even our simple models are becoming more sophisticated, as shown in the sixth article, "Multidimensional Similarity Models for Exploding Foils," in which solutions for exploding foils are extended from one to two and three dimensions.

Our greatest soft x-ray laser success in 1987 was to achieve gain at wavelengths as short as 50 Å, at the virtual doorstep of the 22-to-44-Å "water window" needed to produce x-ray holograms of biological structures. This was achieved with the 4d — 4p ytterbium transition and is described in the seventh article, "Analysis of Nickel-Like X-Ray Lasers with Wavelengths Approaching the Water Window." We believe that similar success can be achieved in a $n = 3$ to $n = 2$ recombination scheme in nickel-like aluminum driven by a 10-ps pulse. Because at present Nova's shortest pulse is in the 100-ps range, we are experimenting at that pulse length to test our modeling capabilities. This work is described in the eighth article, "Analysis of Data on Recombination-Pumped Soft X-Ray Lasers."

Author: M. D. Rosen

Gold Disk Modeling

Last year we found a physically reasonable, one-dimensional LASNEX model for gold disks that does a good job matching most of our time-integrated, gold disk data for 1-ns laser pulse length

illumination. In particular, it seems to properly model:

- 3ω conversion efficiency vs laser intensity.
- Fraction of radiated energy greater than 1.5 and 2.0 keV vs laser intensity.
- The weak intensity dependence of the high-frequency edge of the gold N band.
- 3ω absorption vs Z .
- Approximate equivalence of the measured conversion efficiencies of uranium and gold at a laser intensity of 2×10^{15} W/cm².
- 2ω Novette data.

The Model. Our LASNEX model uses mostly default LASNEX parameter settings. We run Cray LASNEX with 128 photon energy groups (pbins), Pn radiation transport, hot electrons, and the old two-dimensional (2-D) ray-trace laser. The flux limit is 0.05, 20% laser energy deposition at turning point (dumpall), and default inverse bremsstrahlung. We apply small corrections to the XSN (opacity code) radiative bound-bound and bound-free rates, multipliers of 0.5 and 0.25, respectively.

We believe there are two significant differences between this modeling and work we have reported earlier.¹ One is using lots of pbins. The other, more important change, is to tune the radiative rates with the multipliers, especially the bound-bound rate. The fraction of radiated energy XSN puts into the gold M lines turns out to be quite sensitive to the bound-bound rate; see Fig. 2-1, which plots the fraction of radiated energy greater than 2 keV vs the XSN bound-bound multiplier. Uncertainties in the theoretical rates make both such changes plausible.

Fig. 2-1. In simulating the x-ray emission from gold disks, we find that the fraction of radiated energy at photon energies greater than 2 keV is quite sensitive to the XSN bound-bound rate multiplier.

This modeling does not have a multiplier on the emission opacity. We do 1-D disk calculations following Meade's prescription²: a spherical cap whose radius is twice the diameter of the disk we are trying to model. A test calculation using

much finer zoning gave results virtually identical to those of our standard production zoning.

Comparing the Modeling with the Data. Figure 2-2(a) is a plot of radiated x-ray energy less than 1.5 keV divided by

Fig. 2-2. (a) Radiated x-ray energy <1.5 keV divided by incident laser energy vs laser intensity. (b) Calculated total conversion efficiency of the preferred model (which includes energy > 1.5 keV) vs laser intensity. The data points, which are the ones in part (a), show how at high intensity there is significant radiated energy above 1.5 keV.

Fig. 2-3. (a) Fraction of radiated energy > 2 keV vs laser intensity. Various models differing in bound-bound and bound-free multipliers are compared with the data. (b) Fraction of radiated energy > 1.5 keV vs laser intensity. Various models are compared with the data. For all modes, $f = 0.05$.

incident laser energy vs laser intensity. This quantity is plotted for several models. The curve of our preferred model, described above, is identified in this figure. The models differ in flux limiter f , and bound-bound and bound-free multipliers. All the models shown agree with these data.

Figure 2-2(a) also shows the experimentally observed conversion efficiency for 1-ns gold targets.³ Our preferred model agrees well with the data (as do the other models). Note that the experimental definition of conversion efficiency, while not identical to ours, essentially neglects any energy above 1.5 keV. The differences between our definition and the experimental one are small.

Figure 2-2(b) plots calculated total conversion efficiency vs laser intensity. The plot includes all the radiation above 1.5 keV; consequently it does not drop as precipitously at high intensity as in Fig. 2-2(a) [the data points from Fig. 2-2(a) are reproduced to underscore the point]. Note how the total conversion efficiency has a peak at about 6×10^{13} W/cm² and then drops. This is discussed further in the next article, "Design Studies Using the Recently Developed Gold Disk Model."

The principal shortcoming of previous modeling, which did not have any tuning of the XSN radiative rates, was our inability to reproduce various aspects of the x-ray preheat spectrum.¹ Our current model gets these details about right. For example, Fig. 2-3(a) plots the fraction of total radiated energy above 2 keV vs laser intensity. The data came from Dante measurements done at LLNL and transmission grating spectrometer experiments done at Rochester.⁴ Figure 2-3(a) shows how, without the bound-bound multiplier, we greatly underestimated the x-ray preheat at intensities in the mid- 10^{14} W/cm² range, while the new modeling matches the data well.

Figure 2-3(b) shows the same things for x rays of energies greater than 1.5 keV. Various models are compared with the data. Again, our new modeling is far better.

The Henway spectrometer has also been used to estimate M-band emission from Nova disks.³ A comparison between these observations and our modeling is

▲ Fig. 2-4. Fraction of incident laser energy between 2 to 4 keV. Two LASNEX models are compared with Henway spectrometer data.

Fig. 2-5. Overlays of Dante spectra at three intensities show how the N band's upper bound does not advance much with intensity.

Fig. 2-6. Energy of the N band's upper bound vs laser intensity for different models and for the data.

made in Fig. 2-4, which plots the fraction of total laser energy emitted between 2 and 4 keV vs laser intensity (this differs from Figs. 2-2 and 2-3(a), which plotted the fractions of total radiated energy that were >1.5 and >2 keV). The point at 2×10^{15} W/cm² is the only 1-ns data point we are aware of for which our preferred model seriously disagrees with the data.⁵

Fig. 2-7. The model says that 3ω absorption is high and relatively insensitive to Z . Laser intensity is 2×10^{15} W/cm², 1900 J/1 ns.

Fig. 2-8. (a) LASNEX estimate of <1.5 keV conversion efficiency vs laser intensity for several Z s. (b) Total conversion efficiency vs laser intensity for several Z s. ▼

Much of the discrepancy comes from the fact that this particular experiment gave 23% <1.5 keV conversion efficiency, while our model gives 30% <1.5 keV conversion efficiency [see Fig. 2-2(a)].

The final area of clear disagreement between our previous modeling and the data was in the variation of the upper bound of the N band as a function of laser intensity. This is illustrated in Fig. 2-5 where we overlay Dante spectra at a few laser intensities. The data suggest that as the laser intensity is increased, the half-intensity λ of the upper bound N band does not increase when the laser intensity is greater than 1×10^{15} W/cm². Figure 2-6 plots N band upper bound energy vs intensity for a variety of experiments⁴ and for our old and our new modeling. The new modeling shows a much slower increase in the N band upper bound. This improvement results from our reducing the bound-free radiative rate, and is the reason this adjustment to XSN is included in the modeling.

One perplexing result that has been coming out of Nova experiments is consistently high absorption that is independent of Z . Figure 2-7 shows the laser absorption fraction vs Z predicted by our preferred model at a laser intensity of 2×10^{15} W/cm². Again, it (surprisingly) agrees with the observations, saying that the absorption should be very high, close to 100%, except at very low and very high Z values where it drops off slightly.

Two uranium disks have also been shot at laser intensities of about 2×10^{15} W/cm². The conversion efficiency has been found to be about equal to that of gold.³ This is also consistent with our modeling, as is shown in Fig. 2-8(a), which is a plot of <1.5 keV conversion efficiency vs laser intensity for several materials including gold and uranium. The uranium curve in Fig. 2-8(a) shows a band of conversion efficiency at intensities above 1×10^{15} W/cm² because a 2-keV cutoff is a better approximation to the way the uranium Dante data were interpreted. The upper part of the curve represents a 2-keV cutoff, the lower, a 1.5-keV cutoff. Figure 2-8(b) shows the LASNEX estimate, under this model, of

the total conversion efficiency. The total conversion efficiency of uranium is somewhat higher than that of gold.

We have also run this model with a 2ω laser. The conversion efficiency vs laser intensity generally agrees well with the Novette data⁶ (see Fig. 2-9). However, we still cannot explain the one "rogue" data point at 3×10^{13} W/cm² from Argus, even when we take into consideration the relatively low energy and, consequently, the small spot size.

Conclusion. We have developed a physically reasonable, 1-D LASNEX model for gold disks that does well matching most of our time-integrated, 1-ns gold disk data. This model can be used as a design tool, which we shall do in the following article.

Authors: L. J. Suter and S. H. Langer

Design Studies Using the Recently Developed Gold Disk Model

We have used our recently developed 1-D model for gold disks, described in the previous article, to make a variety of predictions and investigations. They include low-intensity conversion efficiency (CE); short-pulse CE; Z-scaling of CE and x-ray preheat; intensity scaling of CE for various Z values and the accompanying spectral changes; attempts to optimize CE with mixtures of materials; and a quick attempt to simulate the effects of induced spatial incoherence (ISI) on CE.

Conversion Efficiency at Low Intensity. LASNEX predicts that there will be a peak in the 3ω CE of gold for 1-ns illumination at a laser intensity of $\sim 6 \times 10^{13}$ W/cm². This was shown in Fig. 2-2(b) of the previous article. A similar peak has been calculationaly observed by the Garching group⁷ and experimentally observed by the Osaka group,⁸ although their measurements of absolute CE differ from ours.

The decrease in CE at lower intensity can be readily explained with an old and simple model of laser-heated disks in which the disk is viewed as a conversion layer that stands off from a reflector. In

Fig. 2-9. Our current model's estimate of 2ω , 1-ns conversion efficiency, compared with Novette and Argus data.

Fig. 2-10. Simple view of a disk where an x-ray conversion layer stands off from a reflecting surface. η_c is the total emission.

this model, the cause for low-intensity drop-off of CE turns out to be a decrease in the albedo of the reflector.

This simple model is shown in Fig. 2-10. The disk's physics is divided into an x-ray conversion layer with intrinsic CE η_i which stands off from a reflecting surface with albedo α . Half of the x rays produced in the conversion layer directly escape. The other half head towards the reflection layer where some are absorbed and most, typically, are reemitted. Thus, the quantity we call CE is

$$\eta_c = \frac{\eta_i}{2} (1 + \alpha). \quad (1)$$

Figure 2-11 indicates how we find the instantaneous, intrinsic CE from our LASNEX simulations. The figure plots the net radiation generation rate per cubic

Fig. 2-11. Net radiation production per cubic centimeter in a simulation vs position. Laser light is incident from the right.

Fig. 2-12. Intrinsic CE (η_i) of the conversion layer at the end of a 1-ns pulse; albedo α of the reflector and resulting instantaneous CE η_c vs laser intensity.

centimeter (PHOV) vs Z for a typical simulation. The laser light is incident from the right. On the right side of the plot is a region where there is net production of x rays. This is the conversion layer. On the left is the reflecting region, where there is net absorption of radiation. The x-ray power per square centimeter generated in the conversion layer is simply the integral of the product of the radiation generation rate per cubic centimeter over the region where it is positive (i.e., over the conversion layer)

and the area. The instantaneous, intrinsic CE is this power divided by the absorbed laser power.

Figure 2-12 shows how intrinsic CE of gold at the end of a 1-ns pulse scales with incident laser intensity. There is a broad peak at about 80% centered around $2 \times 10^{13} \text{ W/cm}^2$. At around $6 \times 10^{13} \text{ W/cm}^2$ the intrinsic CE begins to drop quickly, and it has fallen to about 45% at $2 \times 10^{15} \text{ W/cm}^2$.

The scaling of the albedo of the reflecting layer can be estimated with separate LASNEX simulations in which we apply a radiation flux $= (1/2)n_e I_i$ to a gold slab and find the fraction that is re-radiated. This produces a scaling of albedo (at 1 ns) with incident laser intensity shown in Fig. 2-12.

Using Eq. (1) to combine the albedo and intrinsic CE into an instantaneous, observed CE gives the curve shown in Fig. 2-12. It is qualitatively and quantitatively similar to the time-integrated CE shown in Fig. 2-2(b) of the previous article.

Discussion. This simple model seems to explain, or at least isolate, the physics of the scaling of CE with laser intensity. The low-intensity drop-off of CE is due to the dropping albedo of the gold reflector, a natural consequence of low intensity. The high-intensity drop-off is due to a fall-off in the intrinsic CE.

At $\sim 6 \times 10^{13} \text{ W/cm}^2$ the scaling of intrinsic CE with laser intensity makes a transition from being essentially independent of intensity to decreasing noticeably with intensity. We cannot yet explain why this is so. However, examination of the simulations shows that this change in behavior occurs at an intensity where the laser-plasma interaction is making a transition between two qualitatively different regimes.

At high intensities, LASNEX produces very familiar conditions. The laser penetrates to critical density n_c ; there is a clearly defined, nearly isothermal blowoff corona that starts a few tens of microns away from critical density. Radiation is mostly generated in a narrow—a few microns full width at half maximum (FWHM)—spike located around n_c , in a

region characterized by exceptionally steep temperature and density gradients that are hard to resolve with the code.

By contrast, simulations in which the intrinsic CE has plateaued show a very different situation. There is no clear isothermal blowoff. The laser never penetrates to critical density; its termination at, say, $1 \times 10^{13} \text{ W/cm}^2$ begins at $\sim 0.25\eta_c$. The radiation is emitted from a very broad region several tens of microns wide, most of which is at lower density than the laser's termination. The density and temperature gradients over the deposition/conversion layer are quite shallow and easy to resolve in a simulation.

A laser intensity of $6 \times 10^{13} \text{ W/cm}^2$ appears to be a transition point in the plasma profiles as well as in the intrinsic CE. Although the laser only penetrates to about half critical density, the blowoff is close to isothermal. The electron density and temperature gradients near the laser end points are getting steep, but can still be easily resolved. Also, while the net radiation generation is more spiked than it is at lower intensities, it still comes from a reasonably broad region.

Short-Pulse Conversion Efficiency. A quick check to see if we correctly model the 100-ps CE experiments⁹ gave the predictions that are compared with the data in Fig. 2-13. Quantitatively, we overestimate the 100-ps CE by a lot. Qualitatively, however, the model shows some of the right spirit. With increasing laser intensity, the 100-ps CE is about constant until it intersects the 1-ns CE. Beyond that point the 100-ps CE tracks the 1-ns CE.

We have followed up this quick check with a number of simulations that scrutinized the way LASNEX models early-time x-ray CE. We found that CE can be a rapidly increasing function of pulse width in the 100-ps region when using nearly square pulse shapes. However, we also found LASNEX short-pulse CE to be quite sensitive to pulse shape. Given two shapes of the same FWHM, the one with the longer "foot" can have a substantially higher CE. We suspect that in LASNEX, to a far greater degree than in nature, gold's early-time intrinsic CE can be preconditioned to be high by slowly rising laser energy. The source of this excessive preconditioning might be incorrect estimates of the laser-plasma coupling at early times, when the

Fig. 2-13. Estimate from current LASNEX model of 100-ps, $< 1.5 \text{ keV}$ CE vs laser intensity and corresponding experimental data. For comparison, we also show predicted 1-ns CE.

Fig. 2-14. (a) Net x-ray generation rate in the conversion layer vs time for two intensities. (b) and (c) Conversion efficiency at two intensities vs pulse FWHM for nearly square pulses (shown). Also shown in (c) are CEs for two other pulse shapes.

interaction scalelengths are comparable to the 3ω wavelength.

The findings that led to the two speculations are as follows:

- The instantaneous, intrinsic CE has a rise time that is of order 100 ps or longer, depending on the intensity. Lower intensities have longer rise times. Figure 2-14(a) plots the x-ray generation power in the conversion layer vs time for two intensities (the incident power in the simulations was 1.2×10^{12} W). The laser pulses used in these simulations rose to a flat top in 25 ps.

- Shorter pulses can lead to extremely low conversion efficiencies (CEs). Figures 2-14(b) and (c) plot CE as a function of the pulse's FWHM for two intensities. The nearly square pulse shape used in these simulations (except for the special cases discussed below) is shown in the inset of Fig. 2-14(b). It rises to a flat top in 25 ps

and falls instantly when we shut it off. With this pulse shape we calculate short-pulse CEs that are much lower than the 1-ns CEs. In particular, we can get CEs at ~ 60 ps that are close to what we measured during the Nova 100-ps experiments [compare Figs. 2-14(b) and (c) with Fig. 2-13].

If this result held for other pulse shapes with the same FWHM, then we could argue that the physics in the code is close to explaining the 100-ps experiments; only we are somewhat off in estimating the rise time. Unfortunately, LASNEX shows a clear dependence on pulse shape. For example, points on Fig. 2-14(c) also show calculated 60- and 100-ps FWHM CEs for two different pulse shapes of $\sim 6 \times 10^{14}$ W/cm² peak intensity. The actual pulse shapes are called out in the figure; the 60-ps shape being the shortest reasonable pulse length the 3 ω Nova pulse could have had during the 100 ps series. The CEs we calculate with these other shapes are much closer to the 1-ns value than they are to that of the nearly square pulse. Additionally, Lasinski¹⁰ calculates the CE with short-pulse Gaussians to be much higher than the same FWHM in a nearly square pulse.

At this point, it appears that in LASNEX (but not in nature) the intrinsic CE can be preconditioned to be high by a relatively long, low-intensity precursor to the short pulse. The intrinsic CE at the peak of the 60-ps FWHM Nova pulse (which peaks at 80 ps) is close to what it is at the end of an 80-ps FWHM nearly square pulse.

It has been suggested that LASNEX may be incorrectly modeling the physics of the exceptionally short scalelength plasmas that exist at early times. Our simulations have the early time laser-plasma interaction occurring in a distance comparable to the $1/3\omega$ wavelength. For example, Fig. 2-15 shows the laser deposition and density scalelengths 60 ps into the 60-ps FWHM Nova picket. We speculate that when the laser-plasma interaction is occurring over distances less than several wavelengths, there are effects not included in the modeling that result in less energy deposition and, consequently, a slower rise in the internal CE than we model.

Fig. 2-15. Laser deposition and electron density vs position at 60 ps into the 60-ps FWHM Nova picket. All the action occurs on a scalelength comparable to the $1/3\omega$ wavelength of the laser.

Fig. 2-16. The prediction of our model of how CE will scale with Z at two intensities.

Conversion Efficiency and Spectral Properties at Other Values of Z . Figure 2-16 shows our model's prediction of how CE will scale with Z at 2×10^{14} W/cm² and 2×10^{15} W/cm². The highest CEs occur at the highest Z s.

Figure 2-17 shows the fraction of radiated energy at photon energies greater than 2 keV vs Z at the same two intensities. It oscillates. Note that the >2 keV fraction, which is the important fraction for preheat for Nova-scale capsules, has a peak around gold. Uranium is close to a minimum, as is uranium alloyed with 7% atomic niobium ($Z = 41$). Figure 2-18 shows our model's XSN spectra for disks of different Z s, irradiated at 2×10^{15} W/cm².

We have also done a few laser intensity scalings with materials other than gold. Figure 2-8 of the previous article shows the <1.5 keV and total CE vs laser intensity for a few, selected elements. For some materials, such as erbium, we predict very strong spectral changes with laser intensity; see Fig. 2-19.

Attempts to Optimize Conversion Efficiency with Mixtures. We have taken a cursory look at mixtures as a way of increasing CE. The basic idea, which is rather old, is that different materials radiate strongly in different spectral

regions. See, for example, Fig. 2-18. A mixture is likely to have a fuller spectrum and, presumably, would radiate more efficiently.

A mixture of tungsten and lanthanum might be considered promising. Figure 2-20(a) overlays simulated 2×10^{14} W/cm² disk spectra for the two materials. Lanthanum has a nice peak just where tungsten is weak.

A LASNEX simulation of a 50:50 tungsten-lanthanum atomic mixture indicates that while the resulting spectrum

Fig. 2-17. Fraction of radiated energy >2 keV vs Z at two intensities.

Fig. 2-18. Predicted spectra for disks of various Z values.

Fig. 2-19. For some materials, such as erbium, we predict strong spectral changes with laser intensity.

Fig. 2-20. (a) At $2 \times 10^{14} \text{ W/cm}^2$, lanthanum has a nice peak just where tungsten is weak. (b) Resulting spectrum of a 50:50 tungsten-lanthanum mixture. It is fuller than either of the individual spectra, but has a lower peak.

Table 2-1. Summary of CEs for materials and mixtures studied.

Laser intensity = $2 \times 10^{14} \text{ W/cm}^2$		Laser intensity = $2 \times 10^{15} \text{ W/cm}^2$	
Material	η_c	Material	η_c
^{74}W	0.57	U	0.55
^{57}La	0.52	W	0.47
W + La (50:50)	0.57	U-W	0.48
W + La (75:25)	0.54		
W + La (25:75)	0.53		
^{92}U	0.69		
U-W	0.63		
^{79}Au	0.60		
^{64}Gd	0.55		
Au + Gd	0.58		

is fuller, the CE is not changed. Figure 2-20(b) shows the spectrum of the mixture, which has a lower peak than that of either of the individual materials. The CE of the mixture is 57%. The CEs of the individual materials are tungsten 57% and lanthanum 52%.

While our investigations are far from exhaustive, we have looked at a number of other mixtures that, by the same criteria, looked promising. None of them produced a significant increase in CE over the constituents, and none of them exceeded the calculated CE of uranium. Our findings are summarized in Table 2-1.

We are trying to extract from the code a simple physical reason why simulations of these mixtures did not result in higher CEs.

Induced Spatial Incoherence and Conversion Efficiency. We tried to reproduce the effects of ISI (Ref. 11) with some 1-D simulations. We replaced our usual 1-ns flattop laser source with a rapidly oscillating source having 10-ps triangular oscillations with a maximum to minimum intensity of 40:1. This simulation, then, tries to isolate the effects of an ISI beam's bandwidth on the laser plasma interaction. The time step of the simulation was made small so we could resolve the intensity oscillations.

The result was no change in CE at 2×10^{15} W/cm². The plasma density, temperature, and other profiles in the ISI simulation are extremely close to what they are in the flattop simulations.

Emory¹² has reported 2-D laser-plasma interaction simulations in which he saw a churning-up of the material by ISI. He attributes this convection-like motion to the random poking effects of ISI. This churning could affect CE. It is an effect that cannot be included in these 1-D simulations.

Author: L. J. Suter

Modeling Conversion Efficiency in Gold-Beryllium Disks

Introduction. Conversion efficiency experiments measure the fraction of the energy in a laser beam illuminating a gold disk that is converted into x rays. Experiments of this type have been done for many years because good performance by indirect drive ICF targets requires the efficient conversion of laser light into soft x rays. In the summer of 1987 a new series of CE experiments were run on the Nova laser. These experiments used several new and improved detectors to obtain a much more complete set of data than has ever before been available (see articles in Section 3).

This set of experiments is also the first to study a new type of target, referred to as a dilution target. Our dilution targets are disks made of a mixture of gold and beryllium. Current LASNEX models do a good job of matching the CE for gold, but the set of LASNEX parameters used to achieve this agreement is not unique. In a dilution target, the gold concentration can be lowered to the point that all of the gold lines become optically thin. This is a different situation from an ordinary gold disk, where the optical depth in the N and M bands is large enough to prevent a significant fraction of the emitted radiation from escaping. If a set of LASNEX parameters can be found that matches both the pure gold and the dilution target results, it will greatly increase our confidence in the predictive powers of LASNEX.

The CE is the most important quantity that is measured, and current LASNEX models do a good job of matching it over a large range of laser intensities and gold concentrations. This is true both for the soft x rays (below 1.5 keV), and the M band x rays (roughly 2 to 3 keV). A careful check of CE versus pulse length has not yet been made, but the modeling appears to work for pulses of 1 ns and longer.

The good results on CE modeling encouraged us to take a look at the data on time dependence that several detectors obtained in various x-ray bands. At energies above roughly 500 eV, the x rays take a few hundred picoseconds to come up to their full intensity when driven by a flat-topped laser pulse. After that time they track the laser light quite closely. At energies below 500 eV, the x-ray intensity is still increasing at the end of a 3-ns flat-topped pulse. The dilution targets may prove helpful in understanding the rise time of the x rays, because they show an even greater delay relative to the turning on of the laser. Simple LASNEX models predict that the x-ray time profile should match the laser profile with a delay of 100 ps or less. The experimental results are so recent that no serious attempt at explaining differences between the data and LASNEX models has been made. Thus this article shows the measured x-ray time profiles for several laser pulse lengths, x-ray energies, and gold fractions but does not attempt to model the time dependence.

A preliminary analysis of the spectrally resolved data taken during this series has also been made. The spectra predicted by the XSN atomic physics model in LASNEX (with the use of rate multipliers) are in reasonable agreement with the data, given the simplicity of the XSN model, but comparisons with more sophisticated atomic physics models have not yet been made.

All of the data considered in this article were obtained from single-beam, 3ω Nova shots. A typical shot had roughly 1.6 kJ of blue light incident on the disk. Detectors used to obtain the data include Dante (a broad band, time integrated soft x-ray spectrometer), the Henway crystal spectrograph (which is time integrated, and has roughly 1 eV resolution over the spectral band from 2 to 4 keV), TGSS

(a transmission grating spectrograph that is streaked in time), SDSS (a streaked version of Dante), and several other detectors that will not be discussed here. All of the LASNEX results presented in this

Fig. 2-21. Conversion efficiency for x rays below 1.5 keV vs laser intensity.

Fig. 2-22. Conversion efficiency for 2 to 4 keV x rays vs laser intensity.

Fig. 2-23. Conversion efficiency for x rays below 1.5 keV vs gold concentration (by number of atoms) at laser intensities near 6×10^{14} W/cm 2 .

article were obtained with 1-D spherical cap models, but future work will consider 2-D effects.

Conversion Efficiency. Figure 2-21 shows the Dante CE ($E < 1.5$ keV) for gold disks as a function of laser intensity. Also plotted are several CEs calculated by LASNEX. The agreement is quite good, even at high intensities. Figure 2-22 shows the Henway and LASNEX (2 to 4 keV) conversion efficiencies as a function of the intensity for gold disks. The agreement is good at low intensities, but there are differences at high intensity.

The LASNEX models used in this article all had the bound-bound rates reduced by between 30 and 70% from their nominal values (as well as a few other non-default parameter settings). This choice of parameters was made to achieve a rough match with the measured CE (as of fall 1987) at high intensities and low gold concentrations. Since the time that Fig. 2-22 was prepared, a revised calibration for the Henway data has been developed. The new calibration gives a higher intensity in the M band, and thus disagrees with the data from the earlier reduction procedure shown in these figures and with the LASNEX models that were designed to match the earlier data. The new reduction procedure should remove part of the disagreement at high intensities seen in Fig. 2-22, when there has been time to apply it to all the shots. When the data have been reprocessed, a search for LASNEX parameters that match the Henway, Dante, and time-dependent data will be made. The agreement between the data and the models should be at least as good as that shown in Fig. 2-22, which has been included as an indicator of the kind of results we expect to obtain.

Figure 2-23 shows the Dante CE as a function of the gold concentration (by number of atoms) for the gold-beryllium disks. The intensity for all of these shots was near 6×10^{14} W/cm 2 . With the exception of the pure beryllium shot, the agreement is excellent. The pure beryllium disk is plotted at a concentration of $10^{-3}\%$ because the samples have at least this concentration of various heavier

metals. The conversion for pure beryllium does not have a significant contribution from lines, so it provides a good way to check the modeling of bound-free transitions. Further consideration of these data is necessary, but the agreement is already good.

Figure 2-24 is the same as Fig. 2-23, except that it covers the 2- to 4-keV band. Once again the agreement is quite good, with any disagreements being comparable to those caused by the uncertainty in the laser energy and the response of the Henway detector (this figure was also prepared using the old calibration).

Figure 2-25 compares two LASNEX spectra for a 1:100 Au:Be dilution target (all dilutions are measured by the number of atoms of each species in the mixture). The first model (black line) used a radiative bound-bound rate multiplier of 0.7 and a collisional bound-bound rate multiplier of 0.2. The second model (colored line) used a radiative bound-bound multiplier of 0.3 and a collisional bound-bound multiplier of 1. The spectra are obviously different, especially in the ratio of the N band (0.4 to 1.2 keV) to the M band (2 to 4 keV). This sensitivity to the LASNEX parameters shows that measured x-ray spectra can be used to select one set of LASNEX parameters over another, and this is one of the goals of the experiments. The spectra are usually more sensitive to the LASNEX parameters than is the CE, so spectra are a good way to set parameters in the code.

Figure 2-26 shows the ratio of the LASNEX prediction for each Dante channel to the observed Dante value for a pure gold disk irradiated by a 1-ns pulse at an intensity of 7×10^{14} W/cm². This prediction is made using DIRT, a version of the ALMA program that replaces the old TDG radiation transport code. This program takes the emissivity and opacity structure from a LASNEX run and solves the radiation transport equations for the escaping radiation intensity. DIRT can fold the intensity through a detector response and produce time- and space-integrated results, or streaked images. The black line is from the LASNEX model with a bound-bound opacity multiplier of

0.7 and a collisional bound-bound multiplier of 0.2. The colored line shows the 2 to 4 keV contribution to each channel

Fig. 2-24. The conversion efficiency for 2- to 4-keV x rays gold concentration (by number of atoms) at laser intensities near 6×10^{14} W/cm².

Fig. 2-25. LASNEX spectra for a 1:100 gold:beryllium dilution target illuminated by a 1-ns laser pulse at an intensity of 6.5×10^{14} W/cm² with the radiative and collisional bound-bound multipliers shown.

Fig. 2-26. Pure gold disk target, laser pulse 1 ns, 7×10^{14} W/cm² (shot 17080507): black curve, ratio of LASNEX prediction for each Dante channel to corresponding Dante value; colored curve, M-band contribution to each Dante channel signal.

Fig. 2-27. 1:100 gold:beryllium dilution target, laser pulse 1 ns, 5.6×10^{14} W/cm² (shot 17072105): definition of curves as in Fig. 2-26.

Fig. 2-28. LASNEX spectra for pure gold disk and 1:100 gold:beryllium dilution disk targets. Laser intensity 6.5×10^{14} W/cm², bound-bound opacity multiplier 0.7, collisional bound-bound multiplier 0.2.

(obtained by folding the revised Henway spectrum through the Dante response function). The channel energy increases with increasing channel number, with channels 11 and 12 responding only to x rays above 2 keV. The colored line has a value of nearly 1 for channels 11 and 12. This curve is a ratio of Henway data to Dante data, so the value of 1 means that the two detectors agree on the total M-band energy. The colored curve also shows that some of the lower energy channels, especially 5 and 6, have a significant contribution from 2-keV x rays. Each Dante channel also responds to x rays over a fairly broad range of energies around its nominal energy ($\delta E/E \approx 0.15$ to 0.2). Given these features of the Dante, it is better to fold the LASNEX spectrum through the response of the Dante, as has been done here, than to try to compare an unfolded Dante spectrum to the LASNEX spectrum. The Dante signal is integrated over time and space, so

DIRT is used to calculate a corresponding time-integrated LASNEX prediction. The agreement between LASNEX and Dante is good, except for channel 8, which has a central energy of 920 eV. The large disagreement comes about because LASNEX thinks that there is still gold N-band radiation at this energy, while the Dante data indicates that the N band actually ends at somewhat lower energies. The LASNEX atomic physics model should be corrected to fix this, but it is not a major concern because it can be fixed by a small shift in the energy of the N band.

Figure 2-27 is the same as Fig. 2-26, except that it is for a disk where the gold-to-beryllium concentration ratio is 1:100. The black curve is quite close to the Dante values. Channel 8 for LASNEX is high, just as it was for the gold disk. This LASNEX model overestimates the M band emission, but that can be fixed by a modest change in some of the LASNEX parameters. The colored curve shows that the contribution of the 2- to 4-keV x rays to the low energy channels is still significant. The agreement in both shape and absolute level between Dante and DIRT is close enough that a slightly different set of LASNEX parameters will produce a satisfactory match.

Figure 2-28 compares DIRT spectra for a pure gold disk and a 1:100 dilution disk. In addition to the decrease in overall level, the 1:100 disk has gold M lines (2 to 3 keV) that are stronger relative to the continuum (~ 1.2 to 1.8 keV) than is the case for pure gold. For both cases, the radiative losses are dominated by gold line emission in the N band (0.6 to 1.2 keV). The losses clearly do not drop by as much as the factor of 100 decrease in concentration of gold or the factor of 10 decrease in solid density. This reflects the fact that the conversion layer in pure gold has an optical depth of order unity in the N band, and therefore reducing the concentration allows some of the radiation that was trapped in the pure gold disk to escape.

Figure 2-29 compares the DIRT and revised Henway spectra for the pure gold disk of Fig. 2-26 and for the 1:100 dilution target of Fig. 2-27. The 1:100 spectra have been multiplied by 0.5 to separate

Fig. 2-29. LASNEX/DIRT and revised Henway spectra for pure gold disk of Fig. 2-26, and for 1:100 dilution target of Fig. 2-27 (multiplied by 6.5 to separate curves).

the curves. The LASNEX and Henway spectra are fairly similar, but LASNEX does not resolve the individual lines seen by the Henway. The spectral distribution is also different. In particular, LASNEX produces less emission in the $n = 5$ to $n = 3$ band (3 to 4 keV). The Henway spectrum for the 1:100 disk has narrower line clusters than for the pure gold disk, showing that a smaller range of ionization stages is found in the emitting region. The LASNEX models have parameters chosen to match the revised Henway data. In general, LASNEX/DIRT does remarkably well, especially considering its primitive model of the frequency distribution of the line emission.

The next group of figures shows the experimental time dependence for the SDSS and TGSS x-ray detectors and the 1ω laser intensity (the 3ω intensity looks much like the 1ω intensity for a flat-topped pulse). No LASNEX time dependence

curves are shown, but they would track the laser intensity closely (with a delay of no more than 100 ps) while the laser was on and then continue to radiate for a while after the laser was turned off. Thus any failure of the x-ray intensity to track the laser (during the flat-topped part of the pulse) indicates a problem for current LASNEX modeling. As the figures show, there are such differences in the time dependence, so one of the goals for the coming year will be to come up with LASNEX models that follow the features seen in the experimental time dependence.

Figure 2-30 shows the 1ω laser intensity, the 500- to 1000-eV x rays as measured by TGSS, and SDSS channel 5 (~ 1 keV) as functions of time. All curves in this and the remaining figures have been normalized to the same peak value, so only differences in shape should be considered. This shot was nominally a 1-ns flat-topped pulse at an intensity of

ICF and X-Ray Laser Target Design

Fig. 2-30. SDSS channel 5 (1 keV), TGSS (0.5 to 1 keV), and 1ω laser intensity for gold disk target. Laser pulse 1 ns, 7×10^{14} W/cm².

Fig. 2-31. Same as Fig. 2-30, but laser pulse 3 ns, 4.4×10^{14} W/cm².

7×10^{14} W/cm² onto a pure gold disk. The laser intensity comes up to its full value in roughly 100 ps. The x-ray flux initially increases rapidly, then somewhat more slowly, and does not reach its full value until roughly 200 ps after the laser.

Figure 2-31 shows the same curves for a 3-ns pulse at a laser intensity of 4.4×10^{14} W/cm². In this case, the laser pulse and the x-ray intensity are all basically flat topped, at least after the first couple of hundred picoseconds. The data indicate that the x-ray conversion process, at energies above 500 eV, has settled down to a steady state within 300 ps.

Figures 2-32 and 2-33 show the laser intensity, SDSS channel 1 (~ 190 eV), and TGSS intensity for the 300- to 500-eV band as functions of time for the same two shots. The data show that the low

Fig. 2-32. SDSS channel 1 (290 eV), TGSS (300-500 eV), and 1ω laser intensity for gold disk target. Laser pulse 1 ns, 7×10^{14} W/cm².

energy emission is still rising steadily, even after 3 ns. The increasing emission below 500 eV will lead to a somewhat higher CE for longer pulses and may be of interest for multimegajoule-scale targets. A series of shots with the same intensity and different pulse lengths could be made to study this effect, but it is currently hard to get pulse lengths longer than 4 ns on Nova. These results are still fairly preliminary, so their implications for LASNEX modeling are not clear, but they are interesting and suggest a number of things that must be carefully studied in the coming year.

Figure 2-34 is the same as Fig. 2-30, except that it is for a 1:100 dilution target shot with a 1-ns flat-topped pulse at an intensity of 5.6×10^{14} W/cm². The

Fig. 2-33. Same as Fig. 2-32, but laser pulse 3 ns, 4.4×10^{14} W/cm².

Fig. 2-34. SDSS channel 5 (1 keV), TGSS (0.5–1.1 keV), and 1ω laser intensity for 1:100 gold:beryllium dilution target. Laser pulse 1 ns, 5.6×10^{14} W/cm².

observed 500- to 1000-eV x-ray flux increases throughout the shot, while the laser intensity comes up to its peak value in about 100 ps. This disagreement persists much longer than that seen in the pure gold disk, so it offers a chance to check the time dependence predicted by LASNEX for N-band radiation in a situation where the x rays clearly do not track the laser intensity.

In conclusion, early models of the CE experiments have been quite successful at explaining the CE for 1-ns pulses. The dilution targets provide a new type of information that helps to constrain the set of "knobs" that can be used with LASNEX. A single set of LASNEX parameters that fits the CE of both the dilution targets and the gold disks can be found.

Preliminary studies of time-resolved data indicate that LASNEX thinks the x-ray brightness above 500 eV turns on faster than is observed, and the data indicate a steady brightening below 500 eV throughout a 3-ns flat-topped laser pulse. Further examination of the data and the modeling is required, but the time dependence is very interesting. Agreement between the experimental and LASNEX x-ray spectra is good, considering the simple atomic physics model in XSN. Future work should try some of the more sophisticated atomic physics models in LASNEX. It would also be very helpful to obtain an accurate energy calibration for TGSS, because this would give us our first clear measurement of the spectrum in the N band (0.6 to 1.2 keV), which

contains most of the x-ray flux. The experiments performed in 1987, and additional shots since then, are proving to be quite useful in checking the accuracy of the LASNEX models, and in developing a reliable predictive modeling capability for CE.

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Two-Dimensional Simulations of the Richtmyer-Meshkov Instability in Shock-Tube Experiments

Introduction. Strong shocks are generated during the implosion of ICF capsules. As the outer surface of a shell is ablated by x rays or by laser light, a shock forms ahead of the ablation front and moves toward the center of the capsule containing deuterium-tritium (DT) fuel. Small perturbations that may be present on the inner surface of the shell grow as the shock crosses the shell/fuel interface. The shock-induced amplification of perturbations at a density discontinuity or contact surface is known as the Richtmyer-Meshkov (RM) instability.^{13,14}

A few years ago, we reported our analytic work in the linear regime of the RM instability in which we neglected the effect of compression.^{15,16} In that work, we treated the shock as an instantaneous acceleration. This year we carried out two-dimensional simulations that are fully compressible and allow us to study the linear regimes as well as the nonlinear regime in which the amplitude of a perturbation becomes comparable to its wavelength.

Our calculations simulate actual and potential experiments, which may be carried out with the California Institute of Technology shock tube. The Laboratory has contracted with Caltech to carry out such experiments with B. Sturtevant as the principal investigator. Some results are already available, and our simulations agree well with experimental data. However, many of our simulations are for future experiments at the Caltech facility,

and we expect that within a few years new results will become available to compare with our code calculations.

Figure 2-35(a) shows the Caltech shock tube and the type of calculations we performed. A shock of Mach number $M_s = 1.2$ to 2.0 is generated in a long tube filled with air. The shock travels down the tube and into a test section containing a gas that is either lighter than air (e.g., helium) or heavier than air (e.g., freon or SF_6). (One characteristic of the RM instability is that perturbations grow in either case, as predicted by linear theory.) At the interface between air and the test gas, perturbations of various shapes can be set up by using a thin membrane, examples of which are shown in Fig. 2-35(b). By trapping a gas, such as freon, between two thin membranes, we can carry out double-interface experiments that are also shown in Fig. 2-35(b). The perturbations can be single-scale, double-scale, or multiscale (random).

A few experiments with single-scale perturbations have already been carried out at Caltech; double-scale, multiscale, and double-interface experiments are now in the planning stages. Our simulations include an experiment carried out in 1987 in which several factors, such as density gradients, large-scale perturbations, and small-scale random perturbations, contributed to the evolution of turbulent mix between two gases. In this article, we discuss only a few of our

Fig. 2-35. (a) Schematic of the Caltech shock tube and (b) examples of initial conditions for our 2-D simulations.

calculations using an arbitrary Lagrangian-Eulerian (ALE) code.¹⁷ A longer report on our calculations will soon be available.¹⁸

Perturbations at a Single Interface.

Figure 2-36 shows the evolution of a single, cosine-shaped perturbation at the air/helium interface. The shock has $M_s = 1.2$, and the helium test section is 60 cm long. After the passage of the shock, the perturbation changes phase, becomes exactly zero in Fig. 2-36(b), and grows subsequently. A reflected shock crosses the interface approximately 1 ms after the first shock. Because this reflected shock is moving from the lighter gas (helium) into the heavier gas (air), there is no phase reversal. This behavior is

consistent with the linear-incompressible result for the RM instability. Writing the perturbation as

$$\eta(x, t) = \eta(t) \cos kx, \quad (2)$$

where $k = 2\pi/\lambda$ and λ = wavelength, the amplitude $\eta(t)$ is given by

$$\eta(t) = \eta(0)(1 + Ak\Delta v t), \quad (3)$$

where A = the Atwood number, and Δv is the jump velocity induced by the shock. We found that Eq. (3) describes the evolution fairly well (within 10 to 20%) if we include the compressed initial amplitude $\eta(0)$ in Eq. (3), which was 2 mm in our simulation.

Fig. 2-36. Evolution of a single-scale perturbation across the 10-cm-wide shock tube. The initial amplitude is 0.2 cm, and the helium test gas is initially 60 cm long.

When we replace the test gas by freon, we see the opposite behavior: the first shock from air into the heavier gas (freon) induces no phase change but monotonic growth; the reflected shock, seeing a negative Atwood number, changes the phase of the perturbation. Experiments of this type were carried out in an earlier shock tube at Caltech, and those results are in agreement with our observations.

We can study the evolution of shorter wavelengths by using thin wires across the membrane and by slightly increasing or decreasing the pressure in the test gas. In Ref. 18 we discuss our simulations of two and four wavelengths across

the shock tube. By placing the wires asymmetrically, we can set up a large wavelength next to a smaller one. Our simulations show that at early times, i.e., in the linear regime, Eq. (3) is valid because it predicts that the longer wavelength perturbation grows more slowly (the growth rate is proportional to $k \approx 1/\lambda$). In the nonlinear regime, however, the slower moving, wider perturbation catches up with the shorter wavelength perturbation, and they advance at the same rate. Code simulations are necessary to predict such behavior because our analytic results are no longer valid in this regime.

Fig. 2-37. A 4-cm-thick freon layer in air with perturbations at its lower interface.

Two-Interface Experiments. We have carried out simulations for two-interface experiments involving, in general, three gases. For example, a middle layer could be used to effectively reduce the Atwood number between two fluids having a large density contrast (density gradient stabilization). Another issue in three-fluid experiments is the coupling between the two interfaces of the middle layer. A linear analysis of this issue of feedthrough is contained in Refs. 15 and 16. We carried out numerical simulations for the specific case of a 4-cm-thick freon

layer surrounded by air on both sides. Single- and double-wavelength perturbations were set up initially at the upper, lower, or both interfaces of the freon layer. Here we describe two cases: a two-wavelength perturbation set initially at the lower interface and random perturbations at both interfaces.

Figure 2-37 shows the first case. The 4-cm-thick freon layer is located 60 cm from the end of the shock tube. The phase reversal occurs, as expected, after the incident shock breaks through the lower freon/air interface, after which there is uniform

Fig. 2-38. Evolution of random perturbations, shown as iso-density contours, on the two sides of the freon layer shown in Fig. 2-37.

growth. The reflected shock takes approximately 3 ms to return to the layer, after which spikes in the form of mushrooms develop on the lower interface. The upper interface develops what we refer to as "anti-spikes," but such structures are small. When we put a single wavelength across the layer, mushrooms did not develop within the same time frame, but more feedthrough of perturbations occurred from one interface to another. This result is consistent with the expectation, again based on linear theory for the hydrodynamics of incompressible fluids (and hence approximate), that longer wavelengths feed through more efficiently, although they grow more slowly than shorter wavelength perturbations.

Figure 2-38 shows the second case, in which random perturbations were set up on both sides of the freon layer. Much less feedthrough takes place here compared with the first case because the wavelength of the random perturbations is very short. Every zone, which was approximately 1 mm wide, had an initial

random perturbation with a maximum amplitude of 1 mm. Longer, more coherent wavelengths evolve from such small perturbations, but they never become wide enough to significantly affect the other interface in this calculation. In Ref. 18 we compare this case with cases where random perturbations were set only on the upper or only at the lower interface; within the same time frame (4 ms), perturbations at each interface match closely those shown in Fig. 2-38.

Comparison with Experiment. In a recent experiment at Caltech, a Mach 1.3 shock was sent into a test section filled with SF_6 , which is a gas about 5 times heavier than air. The test gas was separated from the air with a metal plate that was pulled out just before the shock arrived at the air/ SF_6 interface. As the plate was removed, the two gases mixed by diffusion, forming what we call a "premix" layer prior to the arrival of the shock. In this layer, air and SF_6 are mixed, setting up an approximately linear density

Fig. 2-39. Agreement between recent shock-tube experiments at Caltech [(a) through (c)] and the isodensity contours from our simulations [(d) through (f)] is good.

gradient across the transition from air to SF_6 . The pulling action of the plate also sets up relatively large ripples or gravity waves across the premix layer. Figures 2-39(a) through (c) show the initial conditions and two later snapshots; Figs. 2-39(d) through (f) show isodensity contours comparing our simulation with the experimental results provided by Sturtevant.¹⁹

In our simulation, we included the two gravity waves in addition to random, small-amplitude perturbations throughout the premix layer. Because the test section was relatively short (10 cm), several shocks and rarefactions cross the air/ SF_6 interface within the 5.7-ms period. There is a complex interaction between the long-wavelength, large-amplitude gravity waves and the short-wavelength, small-amplitude random perturbations, but the code appears to reproduce the experimental results quite well.

Our code calculations predict that the density gradient set up by the diffusive premix layer acts to suppress, to some extent, the subsequent growth of the mixing layer. Future experiments will be designed to address this issue. Some suggestions for future work are given in Ref. 18.

Conclusions. By using a fully compressible hydrodynamics code, we have checked the predictions of simple, incompressible analytic theories in the linear regime and extended the calculations into the nonlinear regime where theory fails. We have used the code to study the evolution of both single and random perturbations at one or two interfaces between various gases. By focusing on experiments that have been or soon will be carried out at the Caltech shock tube facility, we can check our simulations against experimental results. Our comparisons to date have been positive. The simulations and experiments will give us a better understanding of the RM instability and are expected to suggest ways of suppressing this instability and the mixing at the fuel/tamper interface in ICF capsules.

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Analysis of Data for Neon-Like Soft X-Ray Lasers

Introduction. The production of a plasma medium that can serve as a suitable soft x-ray laser amplifier is a difficult task. Besides achieving the proper ionization state and population inversion, the medium must be uniform enough to support proper beam propagation. At LLNL, such media are created by the interaction of a high-power glass laser with a target. The optical laser heats the target, creating a plasma of free electrons and ions stripped to the intended charge state. We must correctly calculate the hydrodynamics of this interaction as well as the atomic physics of ionization and ultimately of population inversion.

In this article, we show many instances of the need to perform this calculation in as complete a manner as possible, which is necessarily complex. Failure to do so can lead, at times, to erroneous conclusions. By no means do we belittle modeling efforts that are less complex; indeed, we appeal to simple models whenever we can safely do so. Here we point out cases for which we could not use simple models.

After providing an overview of our modeling methodology, we review our state of knowledge as to the hydrodynamic aspects of our neon-like selenium exploding-foil targets,²⁰ which first demonstrated gain in 1984 (Ref. 21). Next, we show evidence from selenium dot spectroscopy that we are correctly modeling the ionization balance. We discuss the measured gains of many of the 3p-3s transitions in selenium and compare them to our calculations. We then discuss the timing of the 3p-3s line emissions, including the sodium-like lines, analogous data on molybdenum, and the need for detailed modeling to account for the angular divergence of the beam pattern in selenium. Finally, we touch on issues for schemes designed to produce gain at a wavelength in the sub-44-Å water window needed for biological holography.

Methodology. We use the LASNEX (Ref. 22) code to simulate the 2-D hydrodynamics of a target heated by the

optical laser. First, the laser and target parameters are fed into LASNEX. Laser absorption, electron and radiation transport, average-atom atomic physics, and hydrodynamics are then calculated, yielding density, temperature, velocity, and radiation-field data in both time and two spatial dimensions. Along with a detailed file of atomic data, these quantities are fed into the XRASER (Ref. 23) atomic kinetics code. The data file typically contains hundreds of detailed atomic energy levels²⁴ along with rates for radiative and collisional excitation and ionization, inner-shell ionization, and dielectronic recombination. XRASER combines these atomic data with the LASNEX-produced hydrodynamic quantities and calculates ionization state abundances, level populations, and gains. The effects of line transfer are also calculated. To directly compare calculation to data, we use a postprocessor, SPECTRE, which creates synthetic spectra with a ray-tracing method. Along a given line of sight with a given temporal, spatial, and spectral resolution, SPECTRE convolves the time- and space-dependent XRASER information to form a synthetic spectrum.

Neon-Like Selenium Hydrodynamics.

Our most successful target in 1984 was a 750-Å-thick selenium foil on a 1500-Å plastic backing for support.^{20,21} This target was illuminated on both sides by a 0.53- μm laser for 0.5 ns in a line focus 200 μm high by 1 cm long and with an

irradiance of about $4 \times 10^{13} \text{ W/cm}^2$ per side. To summarize our recent experimental milestones, we have

- Increased the output at 206 Å from 100 W to nearly 1 MW by increasing the length of the line focus past 4 cm (Ref. 25).
- Spatially (in 1-D) and spectrally resolved the near-field beam patterns (the analysis of which is beyond the scope of this paper).²⁶
- Produced significant double-pass and even triple-pass amplification²⁷ by use of multilayer mirrors.
- Produced Gabor holograms of C fibers using the 206-Å selenium line.²⁸
- Produced measurable gain at a wavelength as low as 50 Å in nickel-like ytterbium.²⁹

The long, theoretical chain of calculations that is finally convolved to compare with the data begins with LASNEX hydrodynamic modeling. Our usual method for determining how well we model the hydrodynamic expansion of the target is via holographic interferometry. Reference 30 describes our continued success in this area.

Another way to deduce both density and temperatures is from the stimulated Raman scattering (SRS) spectrum. The short-wavelength cutoff is due to Landau damping and is thus temperature sensitive. The long-wavelength cutoff leads to estimates of peak density. Figure 2-40 compares LASNEX predictions for temporal evolution of density n_e and temperature T_e at foil center with values derived

Fig. 2-40. (a) Electron density and (b) temperature of a selenium exploding foil vs time. Data inferred from SRS short-wavelength cutoff; LASNEX prediction at center of foil.

from the time history of the SRS spectral cutoff.³¹ The agreement between predictions and data is quite reasonable. The temperature inferred from this method is an upper limit. Because the inference leaves us with a fair amount of uncertainty in temperature, and because

ionization balance is so sensitive to that uncertainty, we would like much more temperature data derived in various ways, for example, from spectroscopy.³²
Ionization Balance in Neon-Like Selenium. We typically deduce ionization balance in neon-like selenium systems by

Fig. 2-41. (a) ▲ LASNEX mesh, (b) electron temperature, and (c) density iso-contours for a selenium dot in CH foil experiment. Snapshot is at a time of peak 3-2 x-ray emission; the selenium region is shaded.

Fig. 2-42. Snapshots of 3-2 x-ray spectra at peak emission time and 200 ps later for selenium dot of Fig. 2-41.

comparing the fluorine-like vs neon-like 3-2 x rays. In actual amplifier geometries, issues of optical depth and 2-D gradients complicate the interpretation.²⁰ To alleviate such problems, we illuminated a 40- μm -diam, 750- $\text{\AA}$ -thick selenium dot on a 3000- $\text{\AA}$ CH foil with 11 J in 700 ps of 0.53- μm light at an irradiance of $2 \times 10^{14} \text{ W/cm}^2$. Figure 2-41 shows the calculated 2-D isocontours of temperature and density at the time of peak x-ray emission. Note the remaining complicating feature of nonuniformity across the exploded dot. Figure 2-42 shows the 3-2 x-ray spectra at two different times compared with the theoretical spectra produced in a properly convolved fashion. The general similarity and relative heights of the step pattern of lines between 7.0 and 7.6 $\text{\AA}$ give us some confidence in the modeling. Satellite lines between 7.6 and 8.0 $\text{\AA}$ appearing in the data are not included in this calculation. The strong line at 7.8 $\text{\AA}$ visible in theory is suppressed in the data due to film damage.

Gains in Neon-Like Selenium. Figure 2-43 compares the measured to theoretically predicted gains³³ in neon-like selenium for the nominal target described earlier in our discussion of hydrodynamics. The predicted gains are obtained by convolving the time- and space-varying gain "data" from XRASER, by means of ray tracing, to produce a synthetic signal at a given amplifier length. We repeat the procedure at various lengths to produce a synthetic "length study" to obtain a gain coefficient in a manner directly analogous to the experimental procedure.

We obtained rather excellent agreement for all lines, except the $J = 0$ to 1 line at 182.4 $\text{\AA}$. Other lines predicted to have measurable gain are not observed, including the fluorine-like line at 203 $\text{\AA}$ and a 2s hole, 3d-3p line near 182 $\text{\AA}$. At the time of peak $J = 2$ to 1 on-axis emission, we actually calculate the $J = 0$ to 1 line to have only about 3/4 of the gain of the $J = 2$ to 1 lines. However, at earlier times the gain on the $J = 0$ to 1 line is predicted to be much larger and, despite refraction, a large signal is expected. Because this early time signal plays a major role in the predicted gain, which is high versus the data, it points to the importance of correctly modeling both the

Fig. 2-43. Comparison of predicted to measured gain on six 3p-3s lines in neon-like selenium.

time-dependent gain and the refraction in detail, as discussed below. The excellent agreement between theory and data of gain values for most of the lines could not have been achieved without the large and complex modeling procedure. In the realm of atomic data alone, we note the important effect on the kinetics of including detailed dielectronic recombination into excited states^{34,35} as opposed to simply connecting only ground states.

Timing of the Gain. In a recent paper, we presented absolutely timed data that showed the $J = 2$ to 1 emission peaking off axis early in the driving pulse.³⁶ The beam then sweeps inward and lands on axis near the peak of the drive pulse. The close agreement demonstrated in that paper between those data and the prediction of the on-axis signal was made possible by detailed modeling, convolving the predicted time and spatial history of the gain and density profiles, which affect both gain and propagation through refraction.

The timing data ruled out a scenario of lasing caused by recombination due to rapid cooling after the drive laser turned off.³⁷ One piece of data²¹ that could be interpreted along those lines shows the $J = 2$ emission increasing while the sodium-like line increases, implying recombination. Figure 2-44 is a reproduction

Fig. 2-44. Line emission vs time for a $J = 2$ to 1 neon-like line and a sodium-like resonance line.

of those data with absolute timing and theoretical predictions superimposed. Note the close agreement. We might ask how the sodium-like line can increase while the pump is still on and is heating the plasma. The answer, once again, lies in detailed modeling. Even though the system is only a few percent sodium-like, the sodium-like line is optically thick along the axis, with a mean free path of about 0.5 mm. Therefore, as the foil expands during this time, the source area increases as does the signal. Thus the sodium-like emission rises even though the plasma has not started to recombine significantly.

This example points out the unique spectroscopic qualities of looking down a long tube of plasma. For example, even the simple notion of an optically thin free-bound continuum is inaccurate. Consider a 1-cm-long exploding foil. Our standard diagnostic, a 0.5-ns, time-gated instrument with 1200 resolving power at 60 eV, typically collects 4.5×10^{-6} J/sr. In the standard analysis of such data, we usually assume a fully exploded amplifier "end-cap" area of 0.02×0.02 cm² radiating for 500 ps, leading to an inferred brightness of 2.5×10^7 W·cm⁻²·sr⁻¹, which is brightness equivalent to a 50-eV blackbody. However, in reality much of the signal arises from a 500-eV blackbody radiating at 3×10^8 W·cm⁻²·sr⁻¹ for 250 ps from an area 0.02×0.003 cm², which occurs when the foil is hotter,

denser, and optically thicker. The free-bound emission first increases in time as the optically thick foil heats up and expands its surface-radiating area. When the density drops low enough for the foil to be optically thin to the continuum, then the emission drops because thin emission is proportional to density squared, which is decreasing. The total free-bound emission thus may not scale linearly with foil length because the signal is not solely due to the foil emitting only in the optically thin limit.

Neon-Like Molybdenum Analogs. One of our observations in neon-like molybdenum³⁸ is relevant to our previous discussions of on-axis spectroscopy. The sodium-like molybdenum, nonlasing resonant line at 104 Å has a width of 120 mÅ, whereas the neon-like line with gain at 106 Å has an instrument-resolution-limited width of 60 mÅ. Thermal Doppler widths should be about 10 mÅ. With sodium-like line optical depths of about 30, a $\sqrt{\ln \tau}$ optical depth broadening brings us only to 20 mÅ. Why then is it 120 mÅ wide?

The solution to this paradox may lie in 3-D effects. The end tips of the line-focus-illuminated region of the foil blow off, not only perpendicular to our on-axis line of sight, but also into our line of sight because they are at the free edge of the foil. Thus the bulk Doppler broadening of about 100 mÅ, convolved with the 60-mÅ instrument resolution, could account for the observed 120-mÅ width. Moreover, quantitative estimates of the brightness of the region within 0.02 cm of the end of the foil, which is expected to have significant 3-D expansion, is about equal to the brightness of the rest of the tube (which is optically thick after another 0.03 cm down the axis).

Table 2-2 compares our gain measurements in molybdenum to theory.³⁸ The lines are identified by their numbers in the data file, upper $J(J_u)$, wavelength (λ_{theory}), and selenium equivalent. Their gain coefficients, derived in the properly convolved way, are presented as calculated using various data files. Table 2-2 shows that detailed models with dielectronic recombination and inner-shell ionization³⁹ change the relative gain of the $J_u = 0$ and $J_u = 2$ lines. (Compare model N2 with A1, both without trapping.)

Table 2-2. Ionization balance and neon-like gain in molybdenum.

Number in data file	f_u	Type of line		Gain coefficient from models (cm^{-1})			Gain coefficient from data (cm^{-1})
		$\lambda_{\text{theory}} (\text{\AA})$	$\lambda_{\text{Se equivalent}} (\text{\AA})$	Model A1 (dielectronic to ground state only, without trapping)	Model N2 (dielectronic detailed F to Ne, Ne to Na, without trapping)	Model N2 (with trapping)	
7-3	1	139.6	221	1.3	2.5	2.3	2.9
26-10	0	138.5	182	3.7	2.8	2.6	—
23-10	2	132.9	209	1.9	4.0	3.7	4.2
8-3	2	131.3	206	1.7	4.1	3.8	4.1
11-3	0	105.4	167	2.8	2.2	2.0	2.2
Ionization state				Percent of ions from $t = -200$ to $t = 0$ ps			
Sodium				30-32	35-27	36-30	
Neon				45-40	43-46	40-45	
Fluorine				12-15	6-13	6-12

Fig. 2-45. Comparison of beam patterns from a selenium exploding foil: (a) gain profile follows density profile; (b) gain peaks off-axis.

Indeed, because of the relatively large fraction of sodium-like ions, inner-shell ionization from those sodium-like ions contributes greatly to $J = 2$ gains. We also note that our best model, N2 with line transfer, shows the same relationship of its predictions for gain to the measured gain in molybdenum as it did in selenium. In particular, the 182- $\text{\AA}$, $J = 0$ equivalent is still misbehaving in molybdenum, while another $J = 0$ (at 106 $\text{\AA}$) has a measured gain quite close to predictions. Once again, we have used our most complex modeling procedures to obtain the generally good agreement with data.

Neon-Like Selenium Beam Pattern. As described earlier in this article, the beam refracts at early times when density gradients in the foil are relatively large. At later times as the density and its gradient are reduced, the beam can propagate straight down the foil axis. The important

point, however, is that refraction alone is not enough to quantitatively explain the observation that the time-integrated beam pattern peaks off-axis. Figure 2-45(a) shows the problem. With a reasonable assumption that the gain profile follows the density profile, the signal gains on-axis for all times because the rays that skim along the top of the parabolic density profile have both maximum gain and length and exit near 0° from the axial direction.⁴⁰

Figure 2-45(b) shows the key to solving the problem of the off-axis peak. When the gain peaks off-axis, the rainbow-shaped ray trajectories have a maximum gain-length product (despite not having maximum length) and exit the plasma at an off-axis angle. Figure 2-46(a) shows detailed, calculated profiles of density and temperature; Fig. 2-46(b) shows ionization-state fraction and gain. Indeed, the gain does peak

Fig. 2-46. (a) Density and temperature profiles for a selenium foil 120 ps before the peak of the driving pulse. (b) Gain and ionization-state-fraction profiles at the same time.

Fig. 2-47. Calculated beam patterns at three different times. Peak power of the pump laser pulse is at $t = 0$.

off-axis because the lower density there leads to less multistep collisional excitation (which causes ionization), thus lowering the fluorine-like fraction and increasing the neon-like ion fraction.

Figure 2-47 shows the subsequent calculated beam patterns at various times using the semi-analytical method described in Ref. 39 in which gain profiles are fit to quartic functions to show off-axis peaks at the appropriate time. Integrating these patterns in time produces a beam pattern with a peak off-axis at 10 mrad that is 1.3 times the on-axis signal.

Shorter-Wavelength X-Ray Laser Schemes.

The nickel-like 4d-4p lasing scheme is an obvious analog to the neon-like 3p-3s scheme. Nickel-like tungsten can lead to gain at 43 Å. However, our recent success²⁹ with $J = 0$ lines at 66 Å in europium and 50 Å in ytterbium have left us with several mysteries. In europium we also predicted⁴¹ the $J = 2$ lines to have measurable gain, but none was observed. We note however that $J = 2$ mysteries are, in principle, easier to explain than those for $J = 0$. With 2-1 x rays having lower fractional inversion to begin with than 0-1, consider a small change in the lower-level population n_1 . We calculate that a 28% increase in n_1 can destroy an inversion on 2-1 x rays while only lowering the gain on 0-1 by 20%. Thus, the discrepancy may be resolved by improved calculations of trapping, plasma parameters, and/or the coupling of the 4s state to the lower 4p laser level, a channel for feeding the lower x-ray-laser state not present in neon-like systems.

The second mystery is the relatively low gain³² of 1 cm^{-1} in ytterbium versus an expected gain of 5 cm^{-1} . Because nickel-like targets are of higher Z and denser than their neon-like selenium cousins, radiative cooling may be more important, and we may be miscalculating

the effect and, thus, the temperature. We are now investigating the latter possibility.

We are pursuing rapid-cooling recombination targets. Short-pulse, hydrogen-like aluminum, 3-2 schemes have been designed with predicted gains of about 4 cm^{-1} . We are also pursuing a line of investigation involving lithium-like ions. We have been unable to correctly predict the high gains seen in 5-3 lithium-like aluminum. We are nonetheless designing calcium (lithium-like, 5-3) and chromium (lithium-like, 4-3) targets, which might show gain near 39 Å .

Conclusions. We have discussed many examples in the analysis of our results for laboratory x-ray lasers at LLNL wherein we see a need for the most detailed, complex modeling and subsequent synthesis of convoluted theoretical data. Many mysteries still remain. We require further theoretical tools, and the finite speed of light needs to be included in SPECTRE. After all, transit time down a 3-cm-long foil is 100 ps, a time scale over which hydrodynamic variables and gains change. For holographic applications, we need to model the coherence and phase properties of lasers. The effects of turbulence on propagation and on line shapes also need to be assessed. Atomic data, including possible absorber lines in nearby ionization states, might enable us to understand missing lasing lines. Finally, the ability to make synthetic spectra that include satellite lines, along with more spectroscopic data, would go far in determinations of temperature.

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Multidimensional Similarity Models for Exploding Foils

Similarity methods have been helpful recently for modeling the hydrodynamics of laser-heated exploding foils and wires.⁴³⁻⁴⁹ They have the virtue of

reducing the partial differential equations of hydrodynamics to a set of easily solved ordinary differential equations. For many cases the similarity solutions give quite accurate representations of the "true" behavior, as determined from numerical solutions of the complete, time-dependent hydrodynamic equations.⁴⁵ For planar foils that are initially very thin, the one-dimensional analysis of Ref. 45 is appropriate for early times. However, at later times, the dynamics may become two- or even three-dimensional as the scalelengths of the plasma perpendicular to the foil's surface become comparable to the transverse scalelength, set by the dimensions of either the foil or the laser irradiation pattern.

The main subject of this article is the evolution of the plasma while two-dimensional effects are important. Multidimensional similarity solutions for exploding objects have been discussed before.⁵⁰⁻⁵² We extend this previous work by presenting solutions for laser-driven foils, by giving new analytical results for adiabatic flows, and by comparing the similarity model with 2-D hydrodynamic simulations.

The Similarity Equations. Consider an exploding foil irradiated by one or several beams of laser light. Let the x_1 axis be perpendicular to the foil surface and parallel to the laser beam, and let the x_2 and x_3 axes be perpendicular to the beam. We generally assume $L_3 > L_2 > L_1$, where the L_i are density scalelengths precisely defined below.

In Cartesian variables, the Eulerian equations describing the free expansion of the foil are

$$\frac{d \ln p}{dt} + \sum_i \frac{\partial V_i}{\partial x_i} = 0, \quad (4)$$

$$\frac{dV_i}{dt} = - \frac{T(t)}{K_1} \frac{\partial \ln p}{\partial x_i}, \quad (5)$$

and

$$\begin{aligned} \frac{d \ln T(t)}{dt} = -(\gamma - 1) \sum_i \frac{\partial V_i}{\partial x_i} \\ + (\gamma - 1) \frac{K_1 H}{T(t)}, \end{aligned} \quad (6)$$

where the convective derivative $d/dt \equiv \partial/\partial t + \sum V_i (\partial/\partial x_i)$. The subscript i runs over the appropriate number of dimensions (1, 2, or 3). In these equations, ρ and T are the mass density and temperature, respectively, V is velocity, and H is the heating rate per unit mass. The temperature is constrained to be constant in space, thus $T = T(t)$. In practice this is often a good approximation, owing to the efficiency of thermal conduction. The constant $K_i \equiv A m_a (Z + 1)^{-1} k^{-1}$, where A is the atomic number, m_a is the unit of atomic mass, Z is the effective ion charge, and k is Boltzmann's constant. In our calculations, γ , the ratio of specific heats, has been assumed to be 5/3.

We seek solutions in which the expansion is homologous, i.e., velocity is linear in position. Thus, we define

$$x_i = \frac{L_i}{L_{i0}} a_i, \quad (7)$$

where a_i is a Lagrangian coordinate in the x_i direction, and the corresponding, time-dependent scale factor is $L_i = L_i(t)$. At time $t = 0$, we have $L_i(0) \equiv L_{i0}$ and $x_i(a_i, 0) = a_i$. Consequently, the velocity components are given by

$$V_i \equiv \left. \frac{\partial x_i}{\partial t} \right|_{a_i} = \frac{\dot{L}_i}{L_{i0}} a_i = \frac{\dot{L}_i}{L_i} x_i, \quad (8)$$

where the dot denotes differentiation with respect to time.

With these assumptions, the momentum equations separate in space and time. The spatial solutions at a fixed time result in Gaussian density profiles in each direction.

The time dependencies of the momentum equations have the symmetric forms

$$L_i \ddot{L}_i = \frac{T(t)}{K_i} = V_s^2(t), \quad (9)$$

where $V_s(t)$ is the isothermal sound speed. Finally, the equations of continuity and thermal energy may be combined to give

$$\frac{d \ln T(t)}{dt} = -\frac{2}{3} \sum_i \frac{d \ln L_i}{dt} + \frac{2}{3} \frac{K_i H}{T(t)}, \quad (10)$$

Equations (9) and (10) are the primary equations discussed throughout the remainder of this article.

Analytic Solutions for Special Cases.

Constant Heating. We first consider the case of a constant heating rate. This may occur during the early period of a constant intensity irradiation, when all of the laser light is absorbed. If constant heating persists long enough, the influences of the initial conditions are lost and asymptotic solutions are approached by the scalelengths and temperature. These solutions are identical for all of the L_i s. Thus 2-D systems become cylindrical, and 3-D systems become spherical.

For 2-D expansions, the asymptotic formulae are:

$$L_1(t) = L_2(t) = (2/3)^{3/2} H^{1/2} t^{3/2}, \quad (11)$$

and

$$T(t) = (2/9) K_i H t. \quad (12)$$

The results have the same functional form as in both the 1-D and 3-D cases. The only differences are the constants, which are listed for the three cases in Table 2-3. Figure 2-48 illustrates the rate of convergence of L_1 and L_2 to the power-law solutions in representative cases when $H = 10^{25} \text{ erg g}^{-1} \text{ s}^{-1}$. If $L_{20}/L_{10} > 10^3$, less than 1 ns is required before $L_2/L_1 \rightarrow 1$.

The temperature history is well described by the power-law expressions in both the 1-D and 2-D regimes, as shown in Fig. 2-49. For the curves in Fig. 2-49, the initial conditions are $L_{10} = 10^{-5} \text{ cm}$, $L_{20} = 10^{-2} \text{ cm}$, $\rho_0 = 3.2 \text{ g cm}^{-3}$, and $T_0 = 10^{-10} \text{ keV}$ for the heated cases and 1 keV for the adiabatic case. The constant heating rate is $10^{25} \text{ erg g}^{-1} \text{ s}^{-1}$.

Laser Heating. We include the effect of laser heating by extending the simple model of Ref. 45. We assume that absorption is due to inverse bremsstrahlung and ignore the reflection of light that may occur when electron densities are above the critical value. The heat input to the whole foil is therefore taken as the surface integral of the absorbed flux.

Table 2.3. Formulae and constants for asymptotic, multidimensional similarity flows.

	Constant heating		Laser heating	Adiabatic
	$T = A_1 K_1 H t$	$L = A_2 H^{1/2} t^{1/2}$	$T \propto t^{-n}$	$T \propto t^{-m}$
Constant	A_1	A_2	n	m
Constant values				
1-D (planar)	1/3	2/3	0	2/3
2-D (cylindrical)	2/9	$(2/3)^{3/2}$	4/5	4/3
3-D (spherical)	1/6	$2^{1/2}/3$	8/5	2

We further assume that the laser beams are incident perpendicular to the foil's surface.

For the 2-D case in which the laser focal spot overfills the foil in the long (x_3) direction, we have

$$H(t) = \frac{4 Y_0 Z_0 F(t)}{M} \times \left\{ 1 - Y_0^{-1} \int_0^{Y_0} \exp[-\tau(x_2, t)] dx_2 \right\}, \quad (13)$$

where $F(t)$ is the incident laser flux, Y_0 and Z_0 are the half-widths of the laser spot in the x_2 and x_3 directions, respectively, and M is the total mass of the foil. The optical depth due to inverse bremsstrahlung is now a function of the transverse (x_2) coordinate, and is

$$\tau(x_2, t) = \frac{3.375 \times 10^6}{T t^{3/2}} \alpha \left(\frac{M^2}{16 \pi^{3/2} Z_0^2 L_2^2} \right) \times \exp \left[-\frac{x_2^2}{(2 L_2^2)} \right], \quad (14)$$

where $\alpha = (\lambda/5300 \text{ \AA})^2 (A/80)^{-2} (Z/25)^2 \Lambda/5$, where λ is the laser wavelength in angstroms, Λ is the Coulomb logarithm, and Z is the average ion charge in the plasma. Lengths in Eq. (14) are in centimeters, and temperatures are in kiloelectron volts.

For the general case, Eq. (13) is integrated numerically. For the common case in which the foil becomes optically thin only when the transverse scalelength is larger than the focal spot width ($L_2 > Y_0$), an approximate evaluation of Eq. (13) is quite accurate. At early times, when $\tau(x_2, t) \gg 1$, the exponential term in Eq. (13) is negligible; at later times, when the term is significant, $x_2^2/L_2^2 \ll 1$, and τ is

Fig. 2-48. Approach of the scalelengths to the power-law solution [Eq. (11)] for two-dimensional flows with constant heating. The runs have $L_{10} = 10^{-5}$ cm and various values of L_{20} . The foil material parameters are $A = 79$ and $Z = 24$.

Fig. 2-49. Temperature histories for constant heating, laser heating, and adiabatic flows.

independent of x_2 . Consequently, for all time, $H(t)$ may be approximated as

$$H(t) = \frac{4Y_0 Z_0 F(t)}{M} \times \left(1 - \exp \left\{ - \left[\frac{\tau_0 L_{10} L_{20}^2}{L_1 L_2^2} \right] \left[\frac{T(t)}{T_0} \right]^{-3/2} \right\} \right), \quad (15)$$

where T_0 is the initial temperature, and

$$\tau_0 = \frac{3.375 \times 10^6}{T_0^{3/2}} \alpha \left(\frac{M^2}{16\pi^{3/2} Z_0^2 L_{10} L_{20}^2} \right). \quad (16)$$

We have developed a similar formula for the 3-D case.

The temperature evolution in a typical case of laser heating is also shown in Fig. 2-49. The parameters for the laser heating are: $Y_0 = 10^{-2}$ cm, $Z_0 = 1$ cm, $M = 4 \times 10^{-6}$ g, and $F = 10^{14}$ W cm $^{-2}$, giving an initial heating rate of 10^{25} erg g $^{-1}$ s $^{-1}$. The optical depth parameter (α) in Eq. (14) is 0.55, as in Ref. 45. If $F(t)$ is constant at late times, when $\tau(t) \ll 1$, we note that $T(t) \propto t^{-4/5}$ for $Y_0 < L_2$. This result can be derived from Eq. (10) using the appropriate form of H and the fact that, at late times, the scalelengths grow linearly with time. Similar arguments show that $T \propto t^{-n}$, where $n \simeq 0$ for 1-D expansions,⁴⁵ and $n = 8/5$ for 3-D expansions, as summarized in Table 2-3.

Adiabatic Expansions. After heating ceases, the flows continue to expand adiabatically. Thus, the temperature evolution may be expressed as

$$T(t) = T_0 \left(\frac{L_{10} L_{20} L_{30}}{L_1 L_2 L_3} \right)^{2/3}, \quad (17)$$

where the subscript zero now refers to the time when heating ceases. Moreover, there exists an energy integral [i.e., an equality derived from Eqs. (9) and (17) that represents energy conservation] of the form

$$\begin{aligned} \sum \dot{L}_i^2 + 3V_{s0}^2 \left(\frac{L_{10} L_{20} L_{30}}{L_1 L_2 L_3} \right)^{2/3} \\ = \sum \dot{L}_{i0}^2 + 3V_{s0}^2. \end{aligned} \quad (18)$$

Although we have been unable to find general solutions for 3-D adiabatic expansions, we have solved the symmetric (i.e., planar, cylindrical, and spherical) expansions.

For 1-D planar expansions, Eq. (18) can be integrated to yield the implicit result for the expansion scale factor as a function of time:

$$\begin{aligned} 3L \left\{ \left[1 - B \left(\frac{L_0}{L} \right)^{2/3} \right]^{1/2} \right. \\ \left. - \frac{2}{3} \left[1 - B \left(\frac{L_0}{L} \right)^{2/3} \right]^{3/2} \right. \\ \left. + \frac{L_0}{L} \left[\frac{2}{3} (1 - B)^{3/2} - (1 - B)^{1/2} \right] \right\} \\ = (3V_{s0}^2 + \dot{L}_{10}^2)^{1/2} t, \end{aligned} \quad (19)$$

where $L \equiv L_1$ and $B = 3V_{s0}^2/(3V_{s0}^2 + \dot{L}_{10}^2)$. For long times, $L_1 \rightarrow (3V_{s0}^2 + \dot{L}_{10}^2)^{1/2} t$, in agreement with earlier work.⁴⁵

For 2-D cylindrical expansion with $L_1 = L_2 \equiv L$, the solution is again implicit⁵³:

$$\begin{aligned} \frac{L_0 b^3}{\sqrt{2}} [F(\epsilon_2, s) - F(\epsilon_1, s)] \\ + L^{1/3} \sqrt{t^{4/3} - b^4 L_0^{4/3}} \\ - L_0 \sqrt{1 - b^4} = \sqrt{\dot{L}_0^2 + (3/2)V_{s0}^2} t, \end{aligned} \quad (20)$$

where $b^4 \equiv [(3/2)V_{s0}^2][\dot{L}_0^2 + (3/2)V_{s0}^2]$, $s = 2^{-1/2}$, $\epsilon_1 = \cos^{-1} b$, $\epsilon_2 = \cos^{-1} (bL_0^{1/3}/L^{1/3})$, and the F s are incomplete elliptic integrals of the first kind. As L/L_0 becomes large, $L \rightarrow [\dot{L}_0^2 + (3/2)V_{s0}^2]^{1/2} t$ and $(\dot{L}_1^2 + \dot{L}_2^2)^{1/2} \rightarrow (\dot{L}_0^2 + \dot{L}_{20}^2 + 3V_{s0}^2)^{1/2} t$, exactly as in the 1-D case.

For 3-D spherical expansion with $L_1 = L_2 = L_3 \equiv L$, we have an explicit solution:

$$L = L_0 \left[D + (1 - D)^{1/2} + \frac{V_{s0} t}{L_0 D^{1/2}} \right]^{1/2}, \quad (21)$$

where $D \equiv V_{s0}^2/(\dot{L}_0^2 + V_{s0}^2)$. For large t , $L \rightarrow (\dot{L}_0^2 + V_{s0}^2)^{1/2} t$, and $(3\dot{L}_0^2 + 3V_{s0}^2)^{1/2} t$.

The power-law indices of the late-time adiabatic solutions are listed in Table 2-3.

In addition to their intrinsic interest, these solutions have been used to verify the accuracy of our numerical scheme for integrating the general, multidimensional similarity equations.

Second Integral for 2-D Adiabatic Flow

Because the adiabatic equations are conservative, there is generally one invariant per degree of freedom. The energy integral expresses one of these invariants. For 2-D flows another invariant describes the asymptotic shape of the density distribution. Because the accelerations approach zero for long times, the velocities must approach constant values. Therefore, as $t \rightarrow \infty$, we have $L_1/L_2 \rightarrow \dot{L}_1/\dot{L}_2$, — a particular constant for each case. As shown in Fig. 2-50, we have found by numerical solutions of the similarity equations that, if the initial velocities are zero in 2-D expansions, $\dot{L}_1/\dot{L}_2 \rightarrow (L_{20}/L_{10})^{1/2}$ with time. The shape of the exploding material at late times is directly related to its initial shape by this relation. In particular, the direction of elongation changes and the degree of elongation becomes less than it was initially.

A rigorous proof of this result is quite subtle. An exact second invariant can be found in the 2-D adiabatic case by first combining a simple rotation with a time transformation upon the Hamiltonian derived from Eqs. (9) and (17).⁵⁴ This puts the system in a form known as the Fokas-Lagerstrom potential, for which a second invariant has been found [see Eqs. (3.3.24) and (3.3.25) of Ref. 55]. In our notation, the second invariant for 2-D, adiabatic similarity expansions is

$$\phi = \dot{L}_1 \dot{L}_2 (L_1 \dot{L}_2 - L_2 \dot{L}_1) + \frac{3V_{s0}^2 (L_{10} L_{20})^{2/3}}{(L_1/L_2)^{2/3}} (L_1 \dot{L}_1 - L_2 \dot{L}_2). \quad (22)$$

If the initial velocities are zero, Eq. (22) yields $\phi = 0$. For this case, Hietarinta has rigorously proved,⁵⁶ via an asymptotic analysis on the first and second integrals, that $\dot{L}_1/\dot{L}_2 \rightarrow (L_{20}/L_{10})^{1/2}$ as $t \rightarrow \infty$, in agreement with our numerical experiments.

Comparison with Numerical Hydrodynamics Simulations. We have

Fig. 2-50. History of the ratio $L_1^2 L_{20} / L_{10}^2 L_2$ for 2-D adiabatic flows. The flows all begin at rest with $T = 1$ keV. The ratio approaches unity for long times, independent of its initial value.

performed several numerical hydrodynamic simulations using the LASNEX computer code⁵⁷ for comparison with our similarity solutions. The method of using this code is discussed in Ref. 45.

As a representative case for comparison, we considered a long, thin laser-heated foil designed as a recombination x-ray laser. Such targets have recently been used to study x-ray lasing in highly stripped ions of magnesium⁵⁸ and aluminum⁵⁹ (see also "Analysis of Data on Recombination-Pumped Soft X-Ray Lasers," later in this section). The lasing scheme works by recombination in an adiabatically cooled plasma after it has been heated and ionized by a short optical laser pulse. The plasma motion starts out primarily as 1-D, perpendicular to the foil's surface. For optimal performance as an x-ray laser, the motion of the plasma should become 2-D after the peak of the heating pulse, so that the plasma cools quickly. Therefore the 2-D similarity model is well suited to studying this type of target.

We have modeled a simplified version of such a target, consisting of a very long, 0.1- μm -thick by 100- μm -wide aluminum ribbon, irradiated on the two 100- μm sides by 200- μm -wide beams of 0.53- μm laser light in a 0.1-ns, Gaussian shaped pulse. The laser pulse peaks at 0.1 ns with an intensity of 4×10^{14} W/cm^2 per beam. The actual targets used in recent experiments are somewhat more complicated, having a thin plastic substrate and overcoating.^{51,59}

Calculated density and temperature histories at the center of the targets are shown in Fig. 2-51. The high-temperature

Fig. 2-51. Density (a) and temperature (b) histories at the center of an exploding aluminum foil. The importance of 2-D expansion and the good agreement between the similarity model and the 2-D numerical simulation are shown. The similarity solution parameters are: $A = 27$, $Z = 11$, $L_{10} = 4 \times 10^{-6}$ cm, $L_{20} = 4 \times 10^{-3}$ cm, $\rho_0 = 2.7$ g cm $^{-3}$, and $\alpha = 0.55$.

period near the peak of the pulse is when the collisional ionization takes place. Recombination takes place at a time near 1 ns, after the plasma has cooled significantly. It is evident on comparing the 1-D and 2-D LASNEX results that the 2-D expansion significantly affects both the temperature and density in the exploding foil. In the 2-D simulation, the density is about 10 times lower at 1 ns than in the 1-D simulation, while the temperature is about 4 times lower. The good agreement between the similarity solution and the 2-D simulation, particularly at late times, is evident. At early times, near the peak of the heating laser pulse, they differ by 13%.

We have found similar agreement between the similarity model and the hydrodynamics simulations in other cases, except those in which laser burn-through occurs relatively late in the pulse. The disagreement in these latter cases is due to the incorrectness of the similarity solution's laser absorption model before burn-through, and to the fact that the isothermality assumed by the similarity solution does not occur in the simulation.

The similarity solutions require about three orders of magnitude less computer time than the simulation, which took about one hour of Cray XMP time for the 250-zone calculation described here. Therefore, the similarity model appears to be useful for quick, approximate design studies for such targets, including 2-D expansion; in fact, it has already been used in this manner.⁶⁰

Summary. We have extended isothermal similarity models for exploding foils to two and three dimensions. This work included analytical solutions for the cases of constant heating and adiabatic flow for planar, cylindrical, and spherical expansions. We also derived an energy integral for general multidimensional adiabatic flows. For 2-D adiabatic expansions, we have demonstrated a second invariant of the motion. This invariant gives the asymptotic shape of a Gaussian object in terms of its initial shape. Our modeling work also included analytical solutions for laser heating. Finally, we have illustrated the usefulness of similarity models by comparisons with numerical hydrodynamics simulations for laser-heated exploding foils designed as recombination x-ray lasers.

We believe that our similarity model is accurate enough to be cautiously applied to modeling the 2-D (and even 3-D) aspects of the dynamics of exploding foils. Its analytic solutions provide both a check on numerical codes and a quick way to estimate the dynamical behavior of exploding foils in the multidimensional regime.

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Analysis of Nickel-Like X-Ray Lasers with Wavelengths Approaching the Water Window

One of the main goals of the x-ray laser community is to achieve significant gain in the water window (23 to 44 Å), defined by the K-absorption edges of oxygen and carbon. A significant step toward this goal was made in 1987, when gain coefficients ~ 1 cm $^{-1}$ were reported for the 0-1, 4p-4d transitions in

nickel-like europium at 66 and 71 Å (Ref. 61) and in nickel-like ytterbium at 50.26 Å (Ref. 62). The incoming laser intensity for these experiments was 7×10^{13} W/cm² (europium) and 1.4×10^{14} W/cm² (ytterbium). This article summarizes our Ref. 63 report on the calculation and design of the nickel-like targets used in these experiments.

The $J = 0$ states are populated by monopole excitation from the ground state, while the $J = 1$ and 2 states receive a large contribution to their populations through dielectronic recombination from the cobalt-like ground state. A significant outcome of our experimental work on nickel-like ions is the fact that the 0-1 laser transition lines are seen on every shot with their full calculated gain. It should be noted that in nickel-like systems, the 0-1 transitions scale more rapidly than other transitions with atomic number Z to shorter wavelengths.

Trapping is a limitation to the gain achievable in nickel-like systems, decreasing the gain coefficients by a factor of 2. The oscillator strength is much larger for the 3d-4p nickel-like transition because of the latter's lower principal quantum numbers and lower angular momentum. The calculated ratio of these oscillator strengths is 6.

Figure 2-52 shows a simplified energy-level diagram for nickel-like europium that includes collisional and radiative rates connecting some of the levels. Because of the very rapid decay, the population of the $(3d_{3/2}4f_{5/2})_1$ state is much smaller than that of the ground state, and

its contribution to the $(3d_{3/2}4d_{3/2})_0$ state is negligible. Therefore, the only transition populating the latter state is the monopole transition from the ground state. In Fig. 2-52(b), we show the rates that are relevant for populating and depopulating the $(3d_{3/2}4d_{3/2})_0$ state. Laser transitions ($4p$ - $4d$) are indicated as well as the characteristic x-rays used to identify the presence of these nickel-like ions.

The position of the $(3d_{3/2}4d_{3/2})_0$ state is difficult to calculate because of mixing with the 1S_0 ground state. Its energy has been determined through use of the multiconfigurational, relativistic Hartree-Fock method,⁶⁴ including orbital relaxation effects. The remaining energy levels and decay rates were obtained from Hartree-Fock calculations using a superposition of configuration.

Our x-ray laser experiment uses the exploding-foil technique.⁶⁵ The target's expansion upon irradiation in the Nova two-beam chamber is calculated with the Lagrangian hydrodynamics code LASNEX.

Then the time-dependent plasma conditions and spectra for different zones, along with the atomic model, are used as input for the kinetics code XRASER, from which we obtain the local gain coefficients. Spectra and average gains are obtained from the XRASER postprocessor SPECTRE.

Our nickel-like model includes the M- and N-shell states in detail and Rydberg states up to $n = 10$ (137 levels total). The 3-4 collisional excitation rates were computed in the relativistic distorted-wave approximation,⁶⁶ and the 4-4 collisional excitation rates were calculated in

Fig. 2-52. Simplified energy level diagrams for nickel-like europium: (a) properties of the levels and (b) collisional and radiative rates pertaining to the laser transition $(3d_{3/2}4p_{3/2})_1$ - $(3d_{3/2}4d_{3/2})_0$ at $n_e = 3 \times 10^{20}$ cm⁻³ and $T_e = 700$ eV. Energies are given in electron volts.

Fig. 2-53. Comparison of 4-3 spectra from the 60-μg/cm² EuF₂ target at 0.7 ns: (a) calculated with instrument broadening of 3.0 eV and (b) measured.

the classical path model. In the copper-like sequence, the $n = 4$ and $n = 5$ states are treated in detail using atomic data from the code YODA. Hydrogenic Rydberg states up to $n = 10$ are included. The other sequences were modeled using largely hydrogenic physics, except for the zinc-like sequence, which has detailed $n = 4$ levels.

The dielectronic recombination coefficients for the nickel-like to copper-like sequence were calculated in the quantum defect approximation based on the relativistic distorted-wave results of Ref. 66. The resulting total recombination coefficients agree with the results of Ref. 67 to within about 20%.

Before dielectronic recombination was available in the data file, its effect was simulated by a rate coefficient of 1×10^{-10} cm³/s connecting the ground states of successive ions.⁶⁷ Using the data file including dielectronic recombination instead of these ground-state-to-ground-state coefficients increased Z by almost one unit. Determining the appropriate value of the coefficient resulted in reducing the dielectronic recombination rate to 3×10^{-11} cm³/s because of density effects. Dielectronic recombination from a cobalt-like ground state (followed by decay) is a major population mechanism for the $(5/2, 5/2)_2$ level.

We report here on our simulation of the 60-μg/cm² EuF₂ target used in the Nova gain-length experiment (for a summary of that experiment, see "Progress in Nickel-Like Lasers" in Sec. 3). If we arbitrarily set the peak of the 1-ns-FWHM input pulse at 1.0 ns, the laser burns through the foil at ~600 ps. The electron density profile at the peak of the input pulse resembles a Gaussian distribution with a FWHM of 232 μm and a peak value of 3.4×10^{20} cm⁻³. This should ensure good beam propagation down the length of the target without severe refraction. The electron temperature in this region ranges from 650 to 700 eV.

To investigate the degree of ionization, we measured the 3d-4f lines, spatially integrated but time dependent. Identifications of spectral lines from nickel-, cobalt-, and iron-like europium have recently been published for a laser-produced plasma.⁶⁸ In addition, 4-3 spectra from iron-, cobalt-, nickel-, copper-, and zinc-like gadolinium measured on the Princeton Large Torus (PLT) Tokamak have been recently presented.⁶⁹

Because our atomic data file does not treat detailed cobalt lines, we can only compare cobalt and nickel emission qualitatively. In Fig. 2-53, the calculated and measured 4-3 spectra for the 60-μg/cm² target (plotted at a time corresponding to the peak of the x-ray pulse) indicate that we are predicting the ion abundance correctly. The calculated copper abundance is $\leq 2\%$. Furthermore, the atomic model does not contain a detailed treatment of the copper-like transition shown in the

spectrum in Fig. 2-53(b). The ionic abundances for the $60\text{-}\mu\text{g}/\text{cm}^2$ target calculated at 1 ns are 18% manganese-like, 25% iron-like, 32% cobalt-like, and 16% nickel-like.

In Fig. 2-54 we show the simulated trapped and untrapped gains near the center of the foil for the 0-1 line at $71\text{ \AA}$ and the 2-1 line at $100.39\text{ \AA}$. The untrapped case is calculated in XRASER by neglecting radiation transfer for the nickel-like resonance lines.

The time- and space-averaged gains (including refraction) are obtained by comparing intensities calculated by SPECTRE at lengths $L = 1.67$ and 3.48 cm . The calculated and experimental average gains for the $60\text{-}\mu\text{g}/\text{cm}^2$ target with an incoming laser intensity of $5.7 \times 10^{13}\text{ W}/\text{cm}^2$ are given in Table 2-4. We find that the gain values are in agreement for the 0-1 lines at 71 and $65.83\text{ \AA}$, but we initially predicted gains $O(1)$ for the 2-1 and 1-1 lines at 100.39 and $104.5\text{ \AA}$, whereas linear growth was measured.⁶¹ Newer gain numbers, obtained using a different version of LASNEX, are in good agreement with experimental values, however.

To optimize the 0-1 yield, we considered the same $60\text{-}\mu\text{g}/\text{cm}^2$ target but with an irradiation of $3.6 \times 10^{13}\text{ W}/\text{cm}^2$. The temperature was then 480 eV and the nickel-like fraction was 45% instead of the previous 16% at 1 ns (peak of the input pulse). In Fig. 2-55, the trapped and untrapped 0-1 and 2-1 lines are plotted. Comparing Figs. 2-55(a) and 2-54(a), we see a larger gain because of the greater abundance of nickel-like ions because refraction does not allow propagation of the x-ray laser beam for times $t < 0.64\text{ ns}$ for the 0-1 lines.

Comparing the trapped and untrapped gains in Fig. 2-55, we see that the most dramatic effect is on the 2-1 line. The gain becomes positive at 0.8 ns in the trapped case and 0.6 ns in the untrapped case. The average gain in the trapped case is 0.6 cm^{-1} ; in the untrapped it is 2.6 cm^{-1} . For the 0-1 line, the average gain is 1.9 and 4 cm^{-1} in the trapped and untrapped cases. Therefore, as we optimize the 0-1 emission, the 2-1 emission decreases and the 2-1 gain becomes more sensitive to trapping.

Fig. 2-54. Calculated gain curves for the $60\text{-}\mu\text{g}/\text{cm}^2$ EuF_2 target with an irradiation of $5.7 \times 10^{13}\text{ W}/\text{cm}^2$ for (a) the 0-1 line at $71\text{ \AA}$ and (b) the 2-1 line at $100.39\text{ \AA}$.

Table 2-4. Gains for the $60\text{-}\mu\text{g}/\text{cm}^2$ EuF_2 target.

Line	$\lambda(\text{\AA})$	Gain (cm^{-1})	
		Calculated ^a	Experimental ^b
$(5/2,3/2)_1 - (3/2,3/2)_0$	71.0	1.2 (1.3) ^c	1.1
$(3/2,1/2)_1 - (3/2,3/2)_0$	65.83	0.36 (0.55) ^c	0.6
$(5/2,3/2)_1 - (5/2,5/2)_2$	100.39	1.4 (0.15) ^c	0.08
$(5/2,3/2)_1 - (5/2,5/2)_1$	104.5	1.0	-0.07

^a See Ref. 63.

^b See Ref. 61.

^c Numbers in parentheses were obtained later with a different version of LASNEX.

A 28% decrease in the population of the $J = 2$ state or a corresponding 28% increase in the $J = 1$ state would reduce the 2-1 gain to zero without significantly reducing the 0-1 gain. Therefore, an improvement in either our treatment of trapping or our dielectronic recombination calculations may explain the discrepancy in the gain coefficient. Also, a lower calculated temperature in the foil would help to explain the results. A modified version of LASNEX does produce lower temperatures, bringing the predicted 2-1 and 1-1 gains into agreement with experiment.

Investigations are now being carried out for nickel-like tungsten, where the 0-1 transition is located at $43.11\text{ \AA}$. The

Fig. 2-55. Time-dependent gain curves for the 60- $\mu\text{g}/\text{cm}^2$ EuF_2 target with an irradiation of $3.6 \times 10^{13} \text{ W}/\text{cm}^2$: (a) the 0-1 line at 71 Å, and (b) the 2-1 line at 100.39 Å.

higher laser intensities required for such experiments will be available on the Nova upgrade.

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Analysis of Data on Recombination-Pumped Soft X-Ray Lasers

Introduction. Recombination x-ray laser schemes are a potential way of obtaining lasing in the water window at wavelengths of less than 44 Å. The $n = 3$ to $n = 2$ transition in hydrogen-like aluminum, the $n = 5$ to $n = 3$ transition in lithium-like calcium, and the $n = 4$ to $n = 3$ transition in lithium-like chromium have the desired short wavelengths.

Theoretical calculations show that a short driving pulse of $\lesssim 20$ ps is required to obtain significant gain, $\sim 4 \text{ cm}^{-1}$, in the $n = 3$ to $n = 2$ hydrogen-like aluminum scheme.⁷⁰ While such short pulses are not currently available, we performed a series of aluminum shots in the Nova two-beam chamber with a pulse length of

100 ps or greater to determine ionization balance and to see if gain could be measured in other lines. In particular, lines associated with the $n = 4$ to $n = 3$ transition in hydrogen-like aluminum and the $n = 5$ and $n = 4$ to $n = 3$ transitions in helium- and lithium-like aluminum are of interest. In this article, we compare data from these experiments with detailed two-dimensional modeling.

To explore a relatively large parameter space or to study particular physics effects, a simple model for recombination x-ray laser targets is useful. We describe such a model and discuss the use of that model to explore the effect of vertical scalelength and additional cooling on the gain for two lines in lithium-like aluminum. Our study is motivated by experiments using aluminum-coated, 7- μm -diameter carbon fibers performed at the Rutherford Laboratory.⁷¹ Researchers there have measured gains of $\sim 3 \text{ cm}^{-1}$ for both the 5f-3d and 4f-3d lines in lithium-like aluminum. An understanding of these results will allow for more reliable predictions of lasing in lithium-like calcium and chromium.

Nova Two-Beam Experiments. The x-ray laser target consists of a strip of aluminum that is 100 μm wide; 1000, 2000, or 4000 Å thick; and 0.5, 1, or 2 cm long sandwiched between two 500-Å-thick layers of Lexan. An initial analysis of our experiments is contained in the 1986 Laser Annual, where more details of the experiment can be found.⁷² One of the conclusions of our initial analysis was that the measured ionization balance was in disagreement with theoretical predictions. This disagreement has been largely resolved by two factors. First, we have refined our modeling and have modeled the actual targets and laser parameters more precisely. Second, the stated experimental intensities have been reduced because of a wider beamline focus than initially assumed.

We will compare measurements of ionization balance with calculations for three shots. Two of the shots were at an intensity of $4 \times 10^{14} \text{ W}/\text{cm}^2$, the highest possible. The third was at an intensity of $1 \times 10^{14} \text{ W}/\text{cm}^2$. For the low-intensity shot and one of the high-intensity shots, we used targets that were 1000 Å thick.

For the other high-intensity shot, we used a target that was 4000 Å thick. All three shots were with green light and a pulse length of 100 ps.

The ratio of upper-state populations in hydrogen-like and helium-like ion states of aluminum can be determined using two instruments. The time-integrated Henway camera provides high-sensitivity and high-resolution data, while the x-ray crystal streak spectrograph (XCSS) provides time-resolved data with lower sensitivity and resolution. Results obtained from the modeling codes LASNEX and XRASER are run through a postprocessor called SPECTRE to obtain synthetic spectra that can be compared directly with data obtained from these two instruments. The LASNEX and XRASER modeling as well as the SPECTRE calculation are two dimensional.

Figure 2-56 shows data from the Henway camera for the three shots, along with the theoretical synthetic spectra. The agreement for all three shots is quite good, showing that in at least a time-integrated sense, the modeling correctly predicts the ionization balance between hydrogen- and helium-like states. For the two high-intensity shots, there is significantly more upper-state population of hydrogen-like than helium-like; for the lower-intensity shot, the converse is true. The modeling calculations show that in the lower-intensity case [Fig. 2-56(c)], there is significant recombination from hydrogen-like into helium-like when the $n = 3$ to $n = 1$ emission is the largest. This recombination provides a large upper-state population in helium-like as the electrons cascade down to the ground state. At this time, there is actually more total population of hydrogen-like than helium-like (most of the population is in the ground state). Therefore, in these recombining plasmas, we must be careful to distinguish between ratios of upper-state populations and ratios of total populations between ionization states.

The agreement between theory and experiment in Fig. 2-56 is good; however, some differences between the two should be noted. The experimentally determined ratio of H_{3-1}/He_{3-1} for the thin target at high intensity [Fig. 2-56(a)] is a bit less—1.4 vs 1.8—than the predicted ratio. The

predicted ratios come from integrating the emission out to 3 ns; for this case (thin target, high intensity), there is still significant emission at 3 ns with He_{3-1} being comparable to H_{3-1} . Had modeling been integrated out to much longer times,

Fig. 2-56. Theoretical and observed time-integrated x-ray spectra for $n = 3, 4$, and 5 levels to the $n = 1$ ground state for hydrogen-like and helium-like ion states.

as in the Henway camera, the H_{3-1}/He_{3-1} ratio would decrease and agree better with the data.

For the thick target at high intensity, the converse is true. The experimental ratio is 1.9, while the predicted ratio is 1.5. For this case, the recombination occurs more rapidly, and there is no significant emission predicted for times greater than 3 ns. The higher experimental ratio might indicate that the thick target becomes hotter and more ionized than theory would predict. The modeling does predict a peak electron temperature of 1.9 keV and that the center region becomes 80% fully stripped. (The thin target reaches 2.1 keV, but only becomes 60% fully stripped.) During modeling we find the thick targets to be sensitive to changes in flux-limiter values, while the thin targets are relatively insensitive. (The flux limiter is a parameter used in LASNEX to model the effect of nonclassical electron thermal conduction.) The flux limiter is 0.03 for all the results given in this article.

We made a series of 1-D LASNEX runs to explore the effect of the flux limiter on thin and thick targets. At the high intensity corresponding to Fig. 2-56(a) and (b), the temperature of the thin target did not change significantly, while the temperature of the thick target almost doubled as the flux limiter was increased from 0.03 to 0.2. This result would imply that a small increase in the flux limiter

would give a H_{3-1}/He_{3-1} ratio that is in better agreement with data for the thick target. This is, of course, only an *ad hoc* change, and the difference in the ratio may be due to other effects, such as uncertainties in the actual intensity on target and the amount of energy absorbed. The latter effects may also explain that, for the thin target at low intensity [Fig. 2-56(c)], the ionization state is higher than predicted.

The time dependence of the H_{3-1} and He_{3-1} line emission is quite different for the thin and thick targets. In Fig. 2-57(a) and (b), we give the predicted time histories for these lines for the high-intensity shots. For the 1000-Å target, the initial peak is associated with the ionization phase. The flatness of the H_{3-1} slope following the peak is representative of the relatively slow time scale over which recombination from fully stripped to hydrogen-like ionization state is occurring. The time histories for the thicker target are significantly different. The electron density is higher so that recombination occurs at a more rapid rate. This effect is seen by the steepness of the H_{3-1} slope to the right of the main peak. For the thicker target, there is a predicted, noticeable dip between the initial ionization peak and the onset of recombination from the fully stripped state. Finally, the thick target is predicted to produce substantially more emission in the 3-1 lines compared to that for the

Fig. 2-57. (a) and (b) Theoretical time-resolved x-ray spectra for H_{3-1} and He_{3-1} , using the same conditions as those in Fig. 2-56(a) and (b), respectively. (c) Experimental XCSS data for the same conditions as in (b).

thinner target. The intensity scale for the thinner target is 1/5 that of the thicker target.

Experimental XCSS data are available only for the thicker target, and those data have problems with sensitivity. Figure 2-57(c) gives the observed time histories for the H_{3-1} and He_{3-1} lines. For the H_{3-1} line, the intensity falls off faster than is predicted by theory, particularly for late time. In addition, the large dip in emission during the fully stripped state is not observed. For the He_{3-1} line, only the peak associated with the ionization phase is observed, and the predicted long, low tail of emission is not observed. The lack of this emission and the sharp drop off at the H_{3-1} emission are most likely due to the signal being too weak and thereby lost in the film fog. This effect can be inferred by measuring the areas under the two curves and comparing them with H_{3-1}/He_{3-1} ratio obtained from the time-integrating Henway results. We obtain values of 6 (XCSS) vs 2 (Henway), implying that significant He_{3-1} emission (and, of course, some H_{3-1} emission) is not observed by XCSS. Even with these problems, we can say that for the H_{3-1} emission, the experimental time history for the thicker target is much closer to that predicted for the thick target than to that for the thin target.

A limited length study (consisting of 3 shots) at an intensity of 1×10^{14} W/cm² per side with a target thickness of 1000 Å gave evidence for greater than linear growth in the $n = 5$ to $n = 3$ lines of both helium-like and lithium-like aluminum. The $n = 4$ to $n = 3$ lines of hydrogen-like and helium-like aluminum were observed to vary linearly as the length was increased from 0.5 to 2 cm. The $n = 4$ to $n = 3$ line of lithium-like aluminum was not observed. The target and laser parameters are the same as those shown in Fig. 2-56(c), and modeling shows that only a small amount of the plasma becomes fully stripped, resulting in significant recombination into helium-like and beyond, but not into hydrogen-like aluminum. The predicted temporally and spatially averaged gains obtained via a synthetic length study using SPECTRE for these conditions are 0.2 cm^{-1} for H_{4-3} , 0.5 cm^{-1} for He_{4-3} , 0.04 cm^{-1} for He_{5-3} ,

0.1 cm^{-1} for Li_{4-3} , and 0.00 for Li_{5-3} . These gains are for the case of no trapping, and actual gain could be smaller than these values. A signal from a 2-cm target would be 4 times that of a 0.5-cm target, times a factor associated with gain. This factor is 1.17, 1.52, 1.02, 1.07, and 1.00, respectively, for the lines discussed above. The only line for which we might expect to see nonlinear growth would be the He_{4-3} line. This result is in conflict with the experimental length study and may be related to the discussion of 5-3 vs 4-3 lines later in this article. However, we cannot place too much weight on results of only three shots.

A comment should be made with respect to the predicted gain for H_{4-3} for an intensity of 2×10^{14} W/cm² and a 150-ps pulse. In the 1986 Laser Annual, a gain of 1 cm^{-1} was given.⁷² This is a peak-gain value, and the averaged value needed to compare with the time-integrating McPigs is 0.4 cm^{-1} . Going from 0.5 to 1 cm (the maximum length currently possible at an intensity of 2×10^{14} W/cm²), a factor of only 1.11 over linear growth would be predicted. An averaged gain of 0.75 cm^{-1} is predicted for an intensity of 3×10^{14} W/cm² and 100 ps, but the higher intensity can be achieved only for lengths below 1 cm. These considerations point to the urgent need for a time-resolved instrument that will help measure gains in systems with low gain lasting for a relatively short duration.

A Simple Model for Recombination X-Ray Laser Targets. A simple model for recombination x-ray laser targets allows us to explore a relatively large parameter space or to study particular physics effects. We used the 2-D similarity model discussed in the article "Multidimensional Similarity Solutions for Exploding Foils" earlier in this section and in Ref. 73 to calculate the laser absorption and hydrodynamics of the exploding foil.

The temperature and density histories obtained from the similarity model are used as input for the XRASER code. Because we use only the time histories from the center of the exploding foil, XRASER need solve only the kinetics in one zone. For collisionally pumped x-ray laser schemes, we can study various physics effects using steady-state calculations.

However, recombination schemes are inherently time-dependent, and we believe this is the simplest model that can provide useful information. Dave and Pert⁷⁴ used a similar approach to model hydrogen-like aluminum. It should be noted that detailed calculations⁷⁵ do not always give the same results as the simple similarity models. However, we can still learn useful information and study various effects using such a model.

We are interested in lithium-like aluminum kinetics and, in particular, the gain in the 5f-3d and 4f-3d lines. In general, the gain between two levels is given by

$$\alpha = \frac{1}{8\pi} \left(\frac{M_i}{2\pi k T_i} \right)^{1/2} \lambda_{ul}^3 A_{ul} g_u \times \left(\frac{N_u}{g_u} - \frac{N_l}{g_l} \right) (\text{cm}^{-1}), \quad (23)$$

where M_i is the ion mass, k is the Boltzmann constant, T_i is the ion temperature, λ_{ul} is the wavelength of the transition, A_{ul} is the Einstein spontaneous coefficient, $g_{u,l}$ is the statistical weight for the upper and lower levels, and $N_{u,l}$ is the population of the upper and lower levels.

The lines with the largest gain between 5f-3d and 4f-3d levels are 5f_{7/2}-3d_{5/2} and 4f_{7/2}-3d_{5/2}, respectively. Using the level numbering in the XRASER data file, we denote these transitions as 22-8 and 15-8, respectively. The gain for each of these lines is then given by

$$\alpha_{22-8} = \frac{5.30 \times 10^{-14}}{[T_i (\text{eV})]^{1/2}} \left(\frac{N_{22}}{8} - \frac{N_8}{6} \right) (\text{cm}^{-1}), \quad (24)$$

and

$$\alpha_{15-8} = \frac{5.03 \times 10^{-13}}{[T_i (\text{eV})]^{1/2}} \left(\frac{N_{15}}{8} - \frac{N_8}{6} \right) (\text{cm}^{-1}), \quad (25)$$

where the populations are given in cm^{-3} . If $N_{22} \approx N_{15}$, the gain in the 5f-3d line would be less by a factor of 10 than the gain in the 4f-3d line. Our initial, detailed (LASNEX + XRASER) calculations of the

Rutherford experiment have given $N_{22} \approx N_{15}$, with the resulting gain in the 5f-3d line being much smaller (0.1 vs 3) than that in the experiment. The gain for the 4f-3d line is in reasonable agreement with experiment. (Trapping is not included in the current modeling.) We see that there are two related conflicts with the Rutherford data. First, the calculated 5f-3d gain is too small by a factor of 30. Second, the predicted ratio between 4f-3d and 5f-3d is too large.

The questions we want to address with the simple model are twofold. First, is there a region in parameter space where the gain in the 5f-3d line is at least on the order of unity? Second, is the ratio of gains between the 5f-3d and the 4f-3d levels close to one as observed in the experiment?

The first effect we will explore is the type of expansion the target undergoes after it is heated by the driving laser pulse. Two scalelengths corresponding to the direction along the driving laser beam and to a direction 90° to the beam are given as inputs to the similarity model. The first scalelength corresponds to the target width, which is set at 1000 Å for this study of expansion effects. The second scalelength at 90° to the beam is referred to as the vertical scalelength. The direction 90° to both of the above directions is along the x-ray laser axis, and we assume that there is no expansion in that direction. The target initially expands in the direction of the driving laser beam in a 1-D manner until the scalelength in that direction becomes comparable to the vertical scalelength. After the two scalelengths are comparable, the expansion is 2-D. For an initially large vertical scalelength ($\geq 1000 \mu\text{m}$), the expansion is essentially 1-D for all times of interest. The experiments done in the Nova 2-beam chamber have a vertical scalelength of 100 μm so that the expansion becomes somewhat 2-D late in the expansion. However, for a vertical scalelength of 7 μm representative of the Rutherford experiment, the expansion is 2-D throughout.

Figure 2-58 shows the peak gain in the 5f-3d and 4f-3d lines for a range of

vertical scalelengths. We see that as the scale-length is decreased, the gain in both of the lines increases significantly. Note that the gain scale on the figure for the two lines is different by a factor of 10 and that the ratio of the gains is always greater than 10. The driving laser intensity is $4 \times 10^{13} \text{ W/cm}^2$, and the pulse duration is 100 ps for all these calculations.

One reason for the increase in the gain is that the time taken to recombine to lithium-like decreases as the vertical scalelength decreases. The decrease in recombination time results in a larger flux passing through the upper levels and thereby increasing the populations in the upper levels. From Eq. (23), the gain is proportional to the upper-level population. An additional effect is that for scale-lengths $\geq 100 \mu\text{m}$, the electron density is high enough at the time of recombination into lithium-like that there is collisional mixing of the $n = 4$ and $n = 5$ population. Such mixing has a significant negative effect on the gain for the 5f-3d transitions, and the gain is essentially zero for these scalelengths.

The second effect we wish to study is that of additional cooling. The Rutherford experiment is really an ablating cylinder with a cold remnant, which can be a heat sink and provide conductive cooling of the heated plasma. Of more general interest, the introduction of impurities could be used to cool the plasma by radiative cooling. The recombination time decreases under additional cooling, resulting in a higher density at the time of lithium-like recombination. This has a beneficial effect until the electron density is high enough, $1 \text{ to } 2 \times 10^{19} \text{ cm}^{-3}$, that collisional mixing of the $n = 4$ and $n = 5$ levels occurs and reduces the 5f-3d gain. A decrease in the initial target thickness (the results for Fig. 2-58 are for a thickness of $1000 \text{ \AA}$) causes the electron density to be lower at the time of recombination into lithium-like. A target with a vertical scalelength of $7 \mu\text{m}$ and a thickness of $250 \text{ \AA}$ has a predicted 5f-3d gain of 0.5 cm^{-1} , assuming additional cooling that caused the recombination time to decrease by a factor of 2. The predicted 4f-3d gain is 8 cm^{-1} for this situation.

Fig. 2-58. Gain vs vertical scale-length for 5f_{7/2}-3d_{5/2} and 4f_{7/2}-3d_{5/2} lines, labeled 5f-3d and 4f-3d, respectively, in lithium-like aluminum. Initial target thickness = $1000 \text{ \AA}$.

The conclusions of our study are that the 5f-3d gain can be increased, but not dramatically, and that the 4f-3d gain is usually larger by at least a factor of 10. Both conclusions are contrary to the Rutherford data cited in this article.

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Plasma Physics

Introduction

An important goal of the ICF Program is to model the physics of laser-target coupling more accurately in order to optimize target designs and guide the choice of future drivers. To this end, the Plasma Physics Group carries out theoretical studies and simulations of the interaction processes, develops computational models, and works closely with experimental groups in designing and interpreting laser plasma experiments. During 1987, members of the Plasma Physics Group also participated in the Heavy-Ion Fusion (HIF) Project, an effort partially funded by the Department of Energy's Office of Energy Research.

Our ICF Program work is discussed first. Smoothing the intensity profiles of laser beams by introducing temporal and spatial incoherence is a promising way to improve laser-target coupling. Some

important benefits of such smoothing are examined in "Effects of Induced Spatial Incoherence on Parametric Instability Growth Rates" and "Self-Focusing of Hot Spots in Laser Beams." Greater understanding of how plasma density fluctuations affect laser-driven instabilities is important. Our work on nonlinear effects associated with ion-density fluctuations are discussed in "Role of Stimulated Brillouin Scattering in Reducing Stimulated Raman Scattering" and "Quasi-Resonant Decay of Ion Waves." We also developed a code for estimating instability levels in target designs; this work is described in "PCUBED: A 2-D LASNEX Postprocessor via ALMA."

Much of our work in 1987 involved collaborating with others in the ICF Program on designing and interpreting Nova experiments to characterize laser-target coupling. One example is discussed in "Time-Dependent X-Ray Conversion." Other work on experimentally characterizing Raman scattering, filamentation, and $3/2 \omega$ emission is reported in the "Laser Experiments and Advanced Diagnostics" articles of Sec. 3.

Division-wide activities on the HIF Project consisted of numerical studies of heavy-ion-beam compression, transport, and target physics. A heavy-ion-beam driver is a promising candidate for long-term ICF energy applications.

Simulations of ion-beam coupling with the target are discussed in "Deposition of Heavy-Ion-Beam Energy on Targets." This work has demonstrated gradient stabilization of the ion-electron two-stream instability and shown that a Weibel instability driven by the electrons' energy anisotropy stabilizes at a benign level. We have also shown that target charging by an incident heavy-ion beam is severely limited by expulsion of target ions and/or by creation of a plasma by photoionization. Details are given in "Discharging of Heavy-Ion Fusion Targets." Changes in the charge state of the incident beam ions have also been calculated in some detail, as discussed in "Photoionization of Beam Ions."

In "Self-Consistent Particle Simulations and Emittance Growth of Heavy-Ion Beams During Longitudinal Compression," we report our calculations for

longitudinal and transverse emittance growths. These growth rates are well within tolerable limits. As described in "Surface Waves on Heavy-Ion Beams," particle simulations have confirmed analytical theory and show that no significant emittance growth results. Finally, we made significant headway in developing simulation codes for heavy-ion applications. The section's last five articles describe this work.

Author: W. L. Kruer

The Effects of Induced Spatial Incoherence on Parametric Instability Growth Rates

Experiments at the Naval Research Laboratory⁷⁶ with laser beams that have ISI suggest that these beams can substantially suppress parametric instabilities and consequently reduce both scattering and hot-electron production. Unlike an ideal coherent beam, an ISI beam has a finite autocorrelation time and length, the first characterized by the beam's temporal bandwidth $\Delta\omega$ and the latter by its spatial bandwidth Δk . These bandwidths^{77,78} exert a stabilizing influence on parametric instabilities when they are comparable with, or exceed, the instability's growth rate or length, respectively.

In 1987 we studied this stabilizing effect and its competition with plasma inhomogeneity and linear damping. Although practical non-ISI beams are generally far from ideal, we found that the smoothing effect of ISI reduces hot spots, even when the direct effects of bandwidth, considered here, appear marginal.

In this article, we limit our study to modes that are both weakly coupled and damped and that are far from their turning points. Their description by the standard coupled-mode equations is then adequate:

$$\left(\frac{\partial}{\partial t} + v_1 \frac{\partial}{\partial x} + v_1 \right) a_1 = \gamma_0(x, t) a_2, \quad (26)$$

and

$$\left(\frac{\partial}{\partial t} + v_2 \frac{\partial}{\partial x} + v_2 \right) a_2 = \gamma_0^*(x, t) a_1. \quad (27)$$

Here $a_i(x, t)$ are the mode amplitudes and v_i and γ_i are the corresponding group velocities and damping rates.

Motivated by our interest in ISI, we adopted a simple model for the pump, $\gamma_0(x, t)$, as the sum of incoherent, overlapping beamlets:

$$\gamma_0(x, t) = \sum_j \gamma_{0j} \exp[ik_j x + i\phi_j(t)]. \quad (28)$$

The spread Δk of the wave numbers k_j derives from the angular spread of the beamlets. The temporal bandwidth of the laser $\Delta\omega$ is contained in the time-dependent phase $\phi_j(t)$.

We reported earlier^{79,80} that in a uniform plasma, average temporal growth rates are reduced from $\langle\gamma\rangle = \gamma_0$ to $\langle\gamma\rangle = \gamma_0^2/[\Delta\omega + \Delta k \min(|v_1|, |v_2|)]$ whenever $\Delta\omega + \Delta k \min(|v_1|, |v_2|) \geq \gamma_0$. In 1987 we extended this result in further analytical and numerical studies.

When there are many superposed beamlets, we identified two competing effects: first, only a fraction of the beamlets satisfy the k -matching condition precisely enough to drive the instability; second, the overlapping beamlets generate an interference pattern with a spatially fluctuating intensity. The first effect reduces the growth rate, as predicted by our previous analysis; the second causes spatial fluctuations of the local growth rate around the predicted mean. When $\Delta\omega \ll \gamma_0$, the second effect creates regions of enhanced growth. When $\Delta\omega \gg \gamma_0$, these regions are temporally smoothed away.

To investigate the instability's absolute and/or convective nature, and thus its behavior in plasmas of finite spatial extent, we derived equations for the equal-time correlation functions $\langle a_i(x', t) a_j^*(x, t) \rangle$ in the manner of Ref. 81 but including the effects of spatial as well as temporal incoherence. With some further approximation, we obtained the following dispersion relation:

$$\begin{aligned} & [\omega - kv_1 + 2i(g_1 - v_1)] \\ & \times [\omega - kv_2 + 2i(g_2 - v_2)] \\ & + 4g_1g_2 = 0. \end{aligned} \quad (29)$$

where

$$\begin{aligned} g_i &= \gamma_0^2/[\omega - k(v_1 + v_2)/2 \\ & - i(|v_1 + v_2 + \Delta\omega + \Delta k|v_i|)]. \end{aligned} \quad (30)$$

Although we made a large-bandwidth approximation in deriving Eq. (29), certain formally small terms were judiciously retained so that the correct limit is obtained in the coherent ($\Delta\omega = \Delta k = 0$) limit. Equation (29) thus smoothly interpolates through the small-bandwidth regime, in which one expects the growth rate to depend on the statistics of the beam and to have large fluctuations about its expected mean. We now summarize some of the results obtainable from Eq. (29).

The convective threshold is given by

$$\begin{aligned} \gamma_0^2 &> v_1 v_2 / \left(\frac{v_1}{\Delta\omega + \Delta k|v_1| + v_1 + v_2} \right. \\ &\quad \left. + \frac{v_2}{\Delta\omega + \Delta k|v_2| + v_1 + v_2} \right), \end{aligned} \quad (31)$$

and the absolute threshold (for $v_1 v_2 < 0$) by

$$\begin{aligned} \gamma_0^2 &> |v_1 v_2| \left(\frac{v_1}{|v_1|} + \frac{v_2}{|v_2|} \right) / \left[\left(\frac{|v_1|}{\Delta\omega + \Delta k|v_1| + v_1 + v_2} \right)^{1/2} \right. \\ &\quad \left. + \left(\frac{|v_2|}{\Delta\omega + \Delta k|v_2| + v_1 + v_2} \right)^{1/2} \right]^2. \end{aligned} \quad (32)$$

In obtaining these two equations, we assumed $\Delta\omega + \Delta k|v_{\min}| + v_1 + v_2 > |ik(v_1 + v_2)/2|$. This restricts the validity of Eq. (32), as is evident from the fact that it fails to reduce to the correct coherent threshold, namely

$$\gamma_0 > \frac{\sqrt{|v_1 v_2|}}{2} \left(\frac{v_1}{|v_1|} + \frac{v_2}{|v_2|} \right). \quad (33)$$

when $\Delta\omega = \Delta k = 0$, unless $|v_1| = |v_2|$. The required validity condition for Eq. (32) is

$$\Delta\omega + \Delta k |v_{\min}| + v_1 v_2 > ||v_1| - |v_2||$$

$$\times \left| \frac{v_1}{|v_1|} - \frac{\alpha v_2}{|v_2|} \right| / (1 + \alpha), \quad (34)$$

where α is defined by

$$\alpha^2 = \frac{|v_1| \Delta\omega + \Delta k |v_2| + v_1 + v_2}{|v_2| \Delta\omega + \Delta k |v_1| + v_1 + v_2}. \quad (35)$$

For a finite slab of homogeneous plasma, an additional threshold must be exceeded to compensate for the energy loss through the boundaries. In the coherent case, the slab's length L must satisfy $L > \pi L_g/2 \approx \pi (|v_1 v_2|)^{1/2}/2\gamma_0$. In the incoherent case, this becomes

$$L > L_g^2 D_1 D_2 \frac{\ln \left(\frac{D_1 |v_2|}{D_2 |v_1|} \right)}{2\Delta\omega ||v_2| - |v_1||}, \quad (36)$$

where $D_i = \Delta\omega + |v_i| \Delta k$, which is independent of v_1/v_2 when $\Delta\omega = 0$ and is otherwise only weakly dependent on this ratio. The ratio L/L_g is thus essentially determined only by $\Delta k L_g$ and $\gamma_0/\Delta\omega$.

We have compared growth rates obtained from the numerical solution of the dispersion relation [Eq. (29)] with those obtained from direct solutions of the amplitude Eqs. (26) and (27). Generally, we found good agreement. However, for large group-velocity ratios (e.g., $|v_1|/|v_2| = 100$), growth rates for pump strengths in the range $D_1 < \gamma_0 < D_2$ are somewhat greater than the predicted (amplitude) growth rate $\gamma \approx \gamma_0/(2)^{1/2}$. In this regime, the temporal bandwidth is insufficient to average out the spatial variations of growth.

Plasmas of practical interest in laser fusion are not spatially homogeneous and often have spatial gradients that affect or suppress parametric instabilities.

Thus the combined effects of spatial gradients and ISI are of interest. We considered two cases:

1. A ramp profile: the spatial gradients induce a linear mismatch in the k -vectors of the parametrically coupled modes $\Delta k(x) \approx \kappa' x$.

2. A parabolic profile: κ' locally vanishes and $\Delta k(x) \approx \kappa'' x^2$. The first case is the generic one; the second is the significant exception of a local "flat-spot," where the effects of inhomogeneity are significantly reduced.

Earlier work on the ramp model⁷⁸ predicted that temporal bandwidth does not affect the convective amplification coefficient, the reduction in the spatial growth rate being compensated by an increase in the length of the amplifying region. Our numerical calculations confirm these results, with the observed gain fluctuating within 20% of the nominal $\exp(\pi\gamma_0^2/\kappa' |v_1 v_2|)$.

Additionally, both long-^{82,83} and short-wavelength⁸⁴ spatial incoherence have been predicted to induce absolute instability in the otherwise only convectively unstable ramp model. We verified these results numerically. We discovered that the addition of temporal bandwidth eventually suppresses the absolute instability induced by spatial bandwidth. The amount of bandwidth needed ($\Delta\omega$) was found to be proportional to Δk for fixed κ' . The critical coherence time was found to be the transit time of the faster wave across a coherence length, i.e., $\Delta\omega \approx \Delta k \max(|v_1|, |v_2|)$.

In the parabolic case, we extended the analysis of Ref. 84 and showed that the ensemble average number N of unstable modes in a system of dimensionless curvature $\tilde{\kappa}'' = \kappa'' L_g^3$ and spatial bandwidth $\tilde{\Delta k} = \Delta k L_g$ is given for $\tilde{\Delta k} \gg 1$ by

$$N/\tilde{\Delta k}^2 = (2\pi s)^{-1/2} \int_{-\infty}^{+\infty} d\lambda \alpha^{-1/2}$$

$$\times \sum_{m=0}^{\infty} \exp[-\pi^2 s(2m+1)^2/8\alpha]$$

$$\times \int_0^{\infty} dx \exp(2\alpha u^2/s) \cos[(2m+1)\pi u], \quad (37)$$

where

$$s = (\tilde{\Delta k} \tilde{\kappa}'')^{1/2}, \quad (38)$$

$$\begin{aligned} \alpha(\lambda) &= \int_{-\infty}^{\infty} dx' [1 + (\lambda + x'^2/2)^2]^{-1} \\ &= \pi [(\lambda^2 + 1)^{1/2} - \lambda]^{1/2} (\lambda^2 + 1)^{-1/2}, \end{aligned} \quad (39)$$

and

$$u(x, \lambda) = \alpha^{-1} \int_0^x dx' [1 + (\lambda + x'^2/2)^2]^{-1}. \quad (40)$$

Taking the threshold criterion to be $N = 1$ and numerically solving Eq. (37), we obtain Fig. 2-59 for the threshold curvature $\tilde{\kappa}''$ for an absolute instability as a function of $\tilde{\Delta k}$. As was found in the uniform and ramp cases,⁸⁴ spatial bandwidth is somewhat less effective in suppressing absolute instability than would be predicted by a simple random-phase model. For large Δk , an unstable system is many growth lengths in extent, and there are many unstable modes with similar growth rates in the corresponding coherent system. More spatial bandwidth is required to suppress all the modes than to suppress a typical mode.

We found numerically that temporal bandwidth likewise increases the gradient threshold. This could be anticipated from Eq. (36) by assuming the parabolic model to be roughly equivalent to a homogeneous slab of length $L_{\text{eff}} = 1.25\pi\kappa''^{-1/3}$, as it is in the coherent case. We found that this simple estimate somewhat underestimates the required $\Delta\omega$ to return an unstable parabolic system back to threshold.

Summary. With a wide range of instabilities and plasma parameters of interest, we cannot make sweeping generalizations about the effects of ISI. It appears, however, that for the high-frequency instabilities that historically have posed problems for laser fusion, feasible ISI bandwidths should have only a marginal direct effect. But this does not mean that ISI is not beneficial.

Experiments have shown substantial Raman scattering when the average incident-laser intensity is lower than

Fig. 2-59. Threshold curvature $\tilde{\kappa}'' = \kappa''(v_1 v_2)^{3/2}/\gamma_0^3$ for an absolute instability decreases with effective spatial bandwidth $\tilde{\Delta k} = \Delta k(v_1 v_2)^{1/2}/\gamma_0$. This theory should be valid for $\tilde{\Delta k} \gg 1$.

theoretical thresholds would predict. These experiments have been explained on the basis of hot spots, perhaps enhanced by filamentation.^{85,86} ISI provides a means of smoothing the laser beams to a more uniform intensity. It is probably this, more than the direct bandwidth effects surveyed here, that makes the use of ISI appear desirable.

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Time-Dependent X-Ray Conversion Efficiency

Much has been written about the x-ray conversion efficiency from high-Z laser irradiated targets.⁸⁷⁻⁹⁰ A summary of the present modeling status and comparison to experimental results is given in the articles "Gold Disk Modeling" and "Modeling Conversion Efficiency in Gold-Beryllium Disks," earlier in this section. In general, the LASNEX simulations are successful in reproducing time-integrated results for incident pulse lengths on the order of 1 ns. Here we report on an initial effort to monitor the time-dependent x-ray conversion efficiency with a resolution of 50 to 100 fs: the use of the spatially discriminating streak spectrograph (SDSS) and the data unfolding procedure is described in the article "Time-Resolved X-Ray Conversion Efficiencies of Laser-Driven Plasmas" in Sec. 3. This more detailed data then provides a new, stringent test for our standard LASNEX modeling.

The basic result is for the first several hundred picoseconds, the standard LASNEX simulations overestimate the x-ray conversion efficiency by at least a factor of 2. This conclusion is preliminary; more definitive results await the completion of a detailed comparison with the well-diagnosed experiments that were done later in 1987, some of which are described in the articles "Modeling Conversion Efficiency in Gold-Beryllium Disks" earlier in this section, and in "X-Ray Emission from Gold and Beryllium Mixtures," "Pulse-Length Scaling of X-Ray Conversion Efficiencies," and "Experiments with Embedded Gold Microdots" in Sec. 3.

Two gold disk shots have been selected for this first look at the time-dependent x-ray conversion efficiency: one with a 2.2-ns, flat-topped, incident laser pulse and the other with a three-picket pulse. Nominal incident laser intensities at $0.35 \mu\text{m}$ for these Nova shots are 10^{15} W/cm^2 for the flat-topped pulse shot and $2 \times 10^{14} \text{ W/cm}^2$ for the last (main) 1-ns part of the three-picket pulse, well within the regime successfully encompassed by the standard modeling.

The key diagnostics for these shots were the SDSS and Dante, located at 35° and 45° , respectively, to the disk normal.

Fig. 2-60. Time-dependent emission into the channel whose dominant energy is 790 eV for the 1-ns flat-topped pulse irradiation.

Two-dimensional simulations were used for comparison to the data collected at these angles. An additional motivation for the more complete 2-D simulations was the discrepancy between the experiment and preliminary 1-D modeling. The nominal LASNEX parameter set for gold disks, as detailed in Ref. 90, was used in the simulations described here. The actual laser pulse shape was used in this modeling, since we were comparing to time-dependent data. We assumed normal incidence in the modeling, since the experimental absorption fractions are greater than 90%. Moderately inhibited electron transport, $f_e = 0.05$, and a typical 2-D emission opacity multiplier, $E_k = 0.5$, were used.

After these time-consuming 2-D simulations were done, the standard LASNEX modeling of gold disk irradiations evolved to that discussed in articles "Gold Disk Modeling" and "Modeling Conversion Efficiency in Gold-Beryllium Disks" earlier in this section. As described in those articles, the *ad hoc* emission opacity multiplier has been dropped in favor of the more physical bound-bound and bound-free multipliers. Comparisons between the two approaches were checked in 1-D simulations and no differences affecting the results described here were found. This is not surprising, since both the SDSS and Dante were primarily looking at spectral regions below 1 keV, and the newer modeling is motivated mainly by the spectral region above 1 keV, that is, by frequencies at or above the N-line cutoff.

From these simulations, the fact that the x-ray conversion efficiency data is taken at one angle does not appear to be a problem. The postprocessing results using TDG are consistent with the $\cos \theta$ angular distribution assumed in the data analysis. Also, the x-ray spectrum appears to change only modestly as the detector is moved from 0 to 60° . By 75° the spectrum does change dramatically, but the solid angle is small there.

The calculated emission into the SDSS/Dante 790-eV channel is compared to the data in Fig. 2-60; here the calculated and experimental profiles are normalized to the same point at ~ 2.5 ns. These curves

are for the flat-topped irradiation pulse. At early times, the calculated emission is too high by at least a factor of 2. In fact, the calculated and experimental curves have different shapes for this channel, which is near the peak of the x-ray spectrum. The increase in emission during the second half of the laser pulse is not reproduced in this modeling. Similar, although not as dramatic, discrepancies exist for the other channels. The 290-eV channel is shown in Fig. 2-61. Here again, the theoretical and modeling results are normalized to the same level at 2.5 ns.

It is tempting to identify this early-time deficit in the x-ray emission modeling with difficulties in simulating experiments with short laser pulses. The calculated x-ray conversion efficiencies for 100-ps Gaussian pulses at $0.35\ \mu\text{m}$ (Ref. 90), and $1.06\ \mu\text{m}$ (Ref. 91) incident wavelength are too high by a factor of 2 in the nominal gold disk LASNEX modeling. We note that the irradiation experiments with layered targets that appeared to require extremely severe transport inhibition were done with 100-ps pulses.⁹² And the time-dependent data from Argus experiments with 700-ps Gaussian pulses on gold disks are also consistent with a lower early time x-ray emission as compared to the nominal LASNEX modeling.⁸⁸

For this 2.2-ns Nova irradiation, the nominal modeling leads to an x-ray conversion efficiency, $\sim 50\%$, that is almost twice as large as the experimental result, 27%. By x-ray conversion efficiency, we mean the x rays that the Dante and the SDSS are detecting; that is, all x rays below $\sim 1\ \text{keV}$. As shown in Fig. 2-62, the calculated x-ray emission tends to track the incident laser pulse more closely than the experimental results do. The discrepancy at early times accounts for only part of this difference. Presumably, more energy goes to hydrodynamic expansion at early times, resulting in

Fig. 2-61. Time-dependent emission into the channel whose dominant energy is 280 eV for the 1-ns flat-topped pulse irradiation.

Fig. 2-62. Incident laser flat-topped pulse shape and calculated emitted power in x rays below 1 keV. A rough sketch of the data for the emitted power is included.

density and temperature profiles that then alter the energy accounting during the rest of the laser pulse.

One caveat is that this particular shot was done quite close to best focus; the incident laser spot is $\sim 250 \mu\text{m}$ in diameter. This implies an extremely modulated beam and the effect of that remains to be assessed. Therefore, the differences in the shape of the x-ray emission, both for the total and the individual channels, is more significant than the quantitative difference in conversion efficiency.

The other gold disk irradiation for which we had the complete SDSS analysis was the three-picket one (see Fig. 2-63). Here, the incident laser spot was more than a factor of 2 larger than for the 2.2-ns one. We attempted to account for the lower experimental x-ray conversion efficiency at early times or short pulses by the extremely *ad hoc* procedure described in Ref. 90 of lowering the emission opacity, E_k , to 0.2 for the first two pickets. This parameter was raised to 0.5 for the main pulse; then the modeling for the main pulse is the same as that used in the simulation of the flat-topped pulse discussed above. This procedure resulted in a better match to each

Fig. 2-63. Time-dependent emission into the channel whose dominant energy is 790 eV for the three-picket pulse irradiation.

time-dependent channel; the 790-eV channel is shown in Fig. 2-63. However, the level and timing of the emission during the second picket are still not right with this purely numerical modeling prescription. Then the overall x-ray conversion efficiency that is dominated by the emission during the main picket is in much better agreement with the data. But, of course, this agreement is presumably due in part to the larger spot size and the presumably smoother beam spatial intensity profile.

A richer data set is becoming available from the next conversion efficiency series. Further progress on the time-dependent x-ray conversion efficiency awaits the comparison of those results with both the nominal gold disk modeling and new and improved physics packages in LASNEX.

Author: B. F. Lasinski

Self-Focusing of Hot Spots in Laser Beams

We typically encounter significant structure, or hot spots, in the intensity profiles of current laser beams. In addition to confusing the interpretation of experiments, these hot spots can reduce heat transport, degrade x-ray conversion, and enhance the role of deleterious, collective plasma effects. To assess the benefits of improving beam quality, we need to clarify the role of hot spots in long-pulse-length experiments. A particular concern is the self-focusing of these hot spots in long-scalelength plasmas.

Let us first give some estimates for this self-focusing of hot spots. For simplicity, we consider ponderomotive focusing of a central hot spot with an intensity at least several times larger than the intensity of the background beam. Neglecting the latter's intensity, we use the ray equations to obtain an estimate for the self-focusing length⁹³:

$$\ell_{\text{SF}} \approx 1.7 \frac{\omega_0}{\omega_{\text{pe}}} \frac{v_e}{v_{\text{os}}} r_0, \quad (41)$$

where ω_0 is the laser light frequency, ω_{pe} is the electron plasma frequency, v_e is

Fig. 2-64. A ray-trace plot from a LASNEX calculation. These are hot-spot rays only.

the electron thermal velocity, v_{os} is the electron oscillatory velocity in the light wave, and r_0 is the radius of the hot spot. The numerical coefficient has been chosen to approximate numerical calculations⁹⁴ which relax the usual assumption that the intensity profile of the beam remains Gaussian. We note that improved calculations should account for ion inertia, density gradients, angular breakup of the beam, and competition with thermal self-focusing and with finer scale filamentation.

Equation (41) can also be expressed as

$$\ell_{SF} \approx 0.8 \left(\frac{n_c \theta_{keV}}{n I_{16} \lambda_\mu^2} \right)^{1/2} r_0, \quad (42)$$

where n is the plasma density, n_c is the critical density, θ_{keV} is the electron

temperature in kiloelectron volts, I_{16} is the hot-spot intensity in units of 10^{16} W/cm², and λ_μ is the laser wavelength in microns. As an example relevant to high-gain targets, we consider $r_0 = 10 \mu\text{m}$, $\lambda_\mu = 0.35$, $\theta_{keV} = 2$, $n/n_c = 0.05$, and $I_{16} = 0.1$; then $\ell_{SF} \approx 500 \mu\text{m}$. Since the self-focusing length is substantially less than the absorption length at this density, strong intensification may occur in such a plasma with a scalelength ≥ 1 mm.

We have examined self-focusing of a hot spot with a 2-D LASNEX simulation using the 2-D ray-trace package. In this simulation, a background beam is focused by an $f/4$ lens 1.2 mm in front of a 25- μm -thick gold disk and then strikes the disk with a peak intensity of 10^{15} W/cm² and a focal spot diameter of 308 μm . To emulate a central hot spot, we superimpose a second beam focused onto the target with a peak intensity of 8×10^{15} W/cm² and a

focal spot diameter of $35\text{ }\mu\text{m}$. For both beams, the wavelength is $0.35\text{ }\mu\text{m}$ and the pulse shape is a 1-ns flat top with a 100-ps rise and fall.

The central hot spot does indeed self-focus, as shown in Fig. 2-64, a ray-trace picture at 800 ps. In a vacuum, these rays would fan out in a Gaussian intensity pattern from a zero radius. There is not always intensity multiplication over the initial hot-spot intensity because of inverse bremsstrahlung absorption. Density contour lines at the same time are shown in Fig. 2-65. At this time, plasma density is elevated on the axis, refracting the light to a ring focus at the hot-spot rays that may be related to the work of Ref. 95. At other times and conditions, self-focusing can be on axis. Clearly the underdense plasma conditions have been

strongly modified. Note that the back side of the gold disk near the center line has been blown out.

We attribute the self-focusing in this simulation to the ponderomotive mechanism. For example, if we use Eq. (42), $\ell_{\text{SF}} \approx 200\text{ }\mu\text{m}$ at $n = 0.1 n_c$. For the high intensity used in this example, the thermal self-focusing length⁹⁶⁻⁹⁹ is much longer. If the self-focusing lengths are longer than the region of interest, then self-focusing is not a problem.

We note that focusing can be partly due to refraction from a density funnel,¹⁰⁰ which forms because a hot spot burns deeper into the target.

Finally, reduced x-ray conversion efficiency is another possible consequence of hot spot self-focusing. This conversion efficiency in gold targets is a function of

Fig. 2-65. Contour of electron density vs radius, showing density expansions corresponding to the focusing ring of the hot spot. The spacing between the lines is a factor of $2^{1/2}$. Critical density n_c is contour A, $n_c/8$ is contour H, and $n_c/16$ is contour J.

intensity and decreases for 1-ns pulses of 0.35- μm light from $\sim 70\%$ at 10^{14} to $\sim 15\%$ at 10^{16} W/cm 2 . A typical Nova beam has $\sim 20\%$ of its energy in hot spots with intensities greater than about twice the average intensity. If these hot spots intensity, x-ray conversion efficiency could easily be reduced by 10 to 15%. This reduction could be even larger if the electron transport becomes degraded by, say, magnetic fields driven by the hot spots. Indeed, recent experiments by Kania et al.¹⁰¹ indicate that beam smoothing significantly increases the x-ray conversion efficiency. Further study of the role of hot spots in experiments is needed.

**Authors: K. G. Estabrook
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Role of Stimulated Brillouin Scattering in Reducing Stimulated Raman Scattering

The role of parametric instabilities in laser-plasma coupling has long been a concern in laser fusion research. Both simulations and experiments indicate that interplay among these instabilities partially determines energy coupling and plasma characteristics. Previous researchers^{102,103} have numerically studied the competition between SRS and stimulated Brillouin scattering (SBS) in finite homogeneous plasmas. In 1987 we extended these studies to include plasma inhomogeneity.

We used simulations with the SATIN code¹⁰⁴ to study how SBS-generated plasma density fluctuations affect the SRS instability in inhomogeneous plasma. Simulations in a linear density gradient showed that with SBS effects, SRS growth is reduced near quarter-critical density but is essentially unaffected at lower densities, when compared to results with fixed ions. The simulations described here include only SRS and SBS parallel to the laser.

Some important nonlinear effects can only be addressed via particle simulations. For nonkinetic effects, however, a reduced description provides a relatively economical simulation and a framework in which competing processes can be

easily isolated and studied independently. The SATIN code solves two-dimensional coupled-wave equations for SRS and two-plasmon decay (TPD) in the fluid limit. The model plasma has an arbitrary density gradient in the x -direction but is uniform in the y -direction. The code has been enhanced to include ion acoustic modes to address the effect of ion dynamics on SRS and TPD.

The system of equations solved is

$$\begin{aligned} & \left(\frac{\partial^2}{\partial x^2} + \Omega_j^2 - N - K_j^2 + 2i\Omega_j \frac{\partial}{\partial \tau} \right) \\ & \times \mathbf{E}_j - (1 - 3V_c^2) \nabla (\nabla \cdot \mathbf{E}_j) \\ & = \sum_{k,l} \left[\frac{-N}{2\Omega_k \Omega_l} \nabla (\mathbf{E}_k \cdot \mathbf{E}_l) - \frac{\Omega_j}{\Omega_l} (\nabla \cdot \mathbf{E}_k) \mathbf{E}_l \right] \\ & + N_l \mathbf{E}_j \end{aligned} \quad (43)$$

for the electric field of the high-frequency modes, and

$$\begin{aligned} & \left(\frac{\partial^2}{\partial \tau^2} + 2\nu_i \frac{\partial}{\partial \tau} - C_s^2 \frac{\partial^2}{\partial x^2} \right) \\ & \times N_i = \frac{Zm_e}{m_i} \sum_k \frac{\nabla^2 |\mathbf{E}_k|^2}{\Omega_k^2} \end{aligned} \quad (44)$$

for the ion-density perturbation.

In these equations, $\mathbf{E}$ is the high-frequency electric field component normalized to $m_e c \omega_0 / e$, Ω is the associated mode frequency normalized to ω_0 , K is the perpendicular wave number normalized to k_0 , and their subscripts j , k , and l label distinct frequency wave-number components. The subscript in both ω_0 and k_0 denotes the laser. Other variables in these equations are normalized such that the background electron density N and the ion-density perturbation N_i are in units of the critical density n_c , the ion damping ν_i is normalized to ω_0 , spatial coordinates are in units of k_0^{-1} , the electron thermal velocity V_e and the sound speed C_s are in units of the speed of light c , and τ is time in units of ω_0^{-1} . The sum on the right-hand side of Eq. (43) represents all pairs of indices such that

$\Omega_j = \Omega_k + \Omega_l$ and $K_j = K_k + K_l$. Note that fast time dependence $\sim e^{i\Omega_j \tau}$ is assumed in Eq. (43).

The time step for solving this system of equations is limited by the instability growth rates or the ion-mode frequencies. The high-frequency equations are solved by an implicit finite difference scheme for the wave equation, with the coupling terms calculated explicitly. Langmuir wave damping is included in V_e and collisional damping in N in the manner prescribed by Forslund et al.¹⁰⁵ The Langmuir waves are forced to zero at the boundaries, and the light waves satisfy outgoing-wave boundary conditions. Equation (44) is solved with a second-order, explicit, upwind differencing scheme.

One of the cases studied was that of SRS and SRS occurring simultaneously in a linearly inhomogeneous plasma. Simulation parameters in normalized units are $E_0 = 0.045$, $V_e = 0.044$, $C_s = 0.003$, density scalelength $k_0 L = 500$, and ion-to-electron mass ratio $m_i/m_e = 184$. These parameters were chosen to give approximately comparable growth rates for the two instabilities. The initial levels were

imposed to provide an ion-density perturbation $\sim E_0$ at $t = 0$. These initial conditions enabled us to obtain qualitative information on the SRS/SBS interaction in a density gradient in relatively economical simulations. To obtain quantitative results, we would have to use a realistic mass ratio and evolve modes from physically determined noise levels in a consistent manner. These more costly simulations will be performed in the future.

The results of our simulations of SRS from various plasma densities are shown in Fig. 2-66. SRS growth rates and scattered-light amplitudes are for simulations including SBS plotted as a fraction of the values obtained from corresponding simulations with fixed ions. The growth rates are the maximum values over the interval of the simulation. Since the SRS instability saturates convectively, its growth rate varies in time; consequently, the scattered-light amplitude may be a better measure of SRS suppression by SBS.

For these parameters, SRS near quarter-critical density ($N = 0.25$) is relatively strongly affected by SBS-generated density perturbations; at lower densities, SRS is unaffected by SBS. One contributing factor is that the SRS growth is weaker at lower densities even in the absence of SBS, owing to gradient effects and stronger Landau damping. Consequently, the SRS growth in the low-density region saturates convectively before the ion waves reach high amplitudes. Another factor is that the SRS-instability growth rate, and hence the level of density perturbations, is lower at low density.

Figure 2-67 shows the temporal evolution of SRS and SBS at the normalized density of 0.16. The decreased growth of SBS at late times is due to pump depletion, whereas the decreased growth of SRS in the fixed-ion case is due to convective saturation. For the case of simultaneous SRS and SBS, the saturation of SRS is due primarily to the detuning effect of the ion-density perturbation.

There is some evidence of enhanced longitudinal electric fields in the SBS-generated density cavities late in the simulations. Enhanced electric fields with the spatial structure of the density perturbations appear without corresponding

Fig. 2-66. Ratio of SRS growth rate and scattered-light amplitude in simulations including SBS to values obtained in simulations with fixed ions.

Fig. 2-67. Temporal evolution of the amplitudes of SRS scattered light from density $N = 0.16$, both with and without SBS density perturbations. Amplitudes are normalized to their initial values at $t = 0$. For reference, the temporal evolution of the profile's maximum SBS is also shown.

growth of the SRS-reflected wave. Simulations with adequate resolution to follow the modes' evolution in this highly nonlinear state are planned for the future.

Simulations with fixed, sinusoidal density perturbations showed results similar to those with ion dynamics, while enhancing the SRS growth rate by decreasing the mass ratio did not further reduce SRS. It appears that for these parameters, the size of the local density perturbations plays a more important role than ion dynamics in reducing SRS. However, ion dynamics are still important in determining the spatial dependence of the ion-wave amplitudes in the inhomogeneous case.

Our results differ from earlier homogeneous plasma simulations, in which fixed ion-density perturbations were observed to have little effect on SRS,¹⁰² and the maximum SRS inhibition occurred at low density where Landau damping weakened SRS growth.¹⁰³ These differences are likely due to gradient-induced effects, such as the localization of modes, the density dependence of the SRS spectrum, and the interaction of SRS and SBS modes originating at different densities.

Work is now under way to extend our results to the lower laser irradiances and longer density-scalelength plasmas characteristic of current and proposed laser-plasma interaction experiments. The SATIN code can also simulate two-dimensional processes like SRS sidescatter and TPD. How ion-density fluctuations affect these instabilities will be addressed in future simulations.

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PCUBED: A 2-D LASNEX Postprocessor via ALMA

LASNEX produces FLASH postprocessor files that contain two-dimensional arrays of electron density, laser intensity, ionization state, and electron and ion temperatures, as well as the zoning coordinates of these quantities. In 1987, we developed PCUBED, a subroutine that uses these data to estimate levels of

various plasma instabilities and the observable light emitted by them for comparison with experiments. PCUBED runs via ALMA, a code written by D. Munro.

Objectives and Work. We had two objectives with PCUBED. First, we wanted to plot calculations of the light emitted by Raman scattering,¹⁰⁶⁻¹⁰⁸ two-plasmon decay $2\omega_{pe}$ (Ref. 109), and resonant absorption as a function of radius z and time for comparison with experiments. These data give us density diagnostics over the plasma instability's range of interest. Second, we wanted to estimate the Raman absorption, heated temperature T_{hot} , and scattering and the spectrum of Raman-shifted light. We calculate the inverse bremsstrahlung (IB) absorption of the scattered spectrum from the plasma back to the lens.

To zero order, the wavelength of the Raman scattered light indicates that density (~ 0.03 to 0.25 critical density n_c) is independent of laser intensity and gain, although to first order, the emitted wavelength is also a function of temperature, angle, and refraction. Raman scattering is also an absorption mechanism and a hot-electron source for densities greater than about $0.1 n_c$.

The Raman threshold is calculated from Landau damping, collisional damping, and the density gradient. The code divides each LASNEX quadrilateral into triangles and then subzones each triangle from the direction of laser focus to better resolve density gradients. It solves the Raman dispersion relation for each subzone to estimate gain, using scaling from Krueer¹⁰⁷ and Nishikawa¹⁰⁸ with fits to kinetic simulations.

PCUBED plots contours of the following:

1. Raman absorption (W/cm^3).
2. Raman scattering (W/cm^3) without IB absorption to arbitrary angles.
3. Raman T_{hot} .
4. Raman scattered-light wavelength.
5. IB absorption length of Raman scattered light.
6. Ratio of laser intensity to $2\omega_{pe}$ threshold intensity ($3/2$ light is the rough experimental indicator).
7. $2\omega_{pe}$ T_{hot} .
8. $2\omega_0$ light from resonant absorption (in rough units of W/cm^2).

9. Density ratio n/n_c , electron and radiation temperature T_e and T_r , laser intensity, and grid.

10. Fringe patterns expected from holography (not including refraction, which is small at these low densities).

11. Ponderomotive and thermal filamentation focus lengths.

The program also plots the space-integrated Raman spectrum with and without IB absorption to the lens. At the end of a run, the program graphs contours of time-integrated Raman (J/cm^3) and $2\omega_{pe}$ emitted light.

As a function of time, the code plots:

1. Fraction and amount (J) of laser energy scattered and absorbed by Raman scattering.

2. Time- and space-integrated Raman spectra (both the scattered energy and spectra are plotted with and without IB absorption back to the lens).

3. Raman T_{hot} weighted by the energy absorbed.

4. Peak multiple of $2\omega_{pe}$ intensity threshold vs time.

Caveats on PCUBED. The code's treatment is not self-consistent: there is no feedback from Raman absorption and/or scattering to the plasma. Consequently, high Raman levels will be overestimates. We eventually hope to have the model in LASNEX for self-consistency. PCUBED treats Raman backscattering only, but sidescattering has similar scaling. Errors are introduced by laser hot spots and the still-unresolved convective and/or absolute character of Raman scattering. The error bars on the thresholds are probably less than a factor of 3. However, since small-signal scattering is exponential with gain, the uncertainty is greater than a factor of 3 on total scattering and absorption. Our model is still at an early stage—improvements and bugs are still being added.

The characteristics and limitations of holography also need to be pointed out. The first fringe is at density $n/n_{cp} = 2\lambda_{op}/L$, where L is the plasma length through which the probe beam traverses, and n_{cp} and λ_{op} are the probe beam's critical density and wavelength. For a fairly thick plasma ($500\text{ }\mu\text{m}$), the first fringe shift for $\lambda_{op} = 0.25\text{ }\mu\text{m}$ is

$1.6 \times 10^{19}\text{ cm}^{-3}$. Additional fringes are the multiples of this density. Unless the vacuum region is clearly seen, it is difficult to identify the first fringe from which to count.

Since at most ten fringes are observable, the maximum density that can be diagnosed from interferometry is about $1.6 \times 10^{20}\text{ cm}^{-3}$, or about 0.04 critical density for the $0.53\text{-}\mu\text{m}$ laser light. The maximum density resolved decreases as plasma thickness increases. For a $500\text{-}\mu\text{m}$ -thick plasma, the maximum density that holography resolves is about the minimum density from which Raman scattering emits observable light.

Other interferometry complications are the microscope's focal point and refraction, and the lack of either Cartesian or cylindrical symmetry in some of the geometries.

Future Plans. Experiments measure scattered light and the fraction and temperature of the hot electrons. These quantities are lower bounds because Raman light can be partially absorbed and hot electrons can be partially equilibrated before reaching a high enough density to radiate significantly. These limitations are not yet accounted for with PCUBED, and this is one area for future work.

We are also working on 2-D and streak displays of Raman scattered-light spectra with IB absorption, on rough sidescatter estimates, on more accurate treatments of gain with a statistical treatment of hot spots, and on rough estimates of Brillouin scattering.

Researchers at Garching¹¹⁰ have experimentally observed $5/4\omega_0$ light that might be from $n_c/16$. They have not spatially resolved the $5/4\omega_0$ emission with interferometry but qualitatively found it was emitted some distance from critical density. Localizing $5/4\omega_0$ would be a simple Phoenix or KMS Fusion experiment and might provide another density diagnostic.

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Quasi-Resonant Decay of Ion Waves

Quasi-resonant decay of finite-amplitude ion-acoustic waves has been proposed¹¹¹ as a saturation mechanism for the Brillouin instability. In 1987 we examined this decay of a driven ion wave into lower-wave-number ion waves in theoretical studies and in simulations with a two-fluid code. We found that it is essential to allow for harmonic generation, which is also quasi-resonant and dominates the mode coupling. In general, harmonic generation suppresses decay into lower wave numbers.

The decay of a finite-amplitude ion wave into two lower-frequency ion waves has been predicted.¹¹¹ For ion waves,

$$\omega = \frac{k v_s}{\sqrt{1 + k^2 \lambda_{De}^2}}, \quad (45)$$

where ω is the frequency, k is the wave number, λ_{De} is the electron Debye length, and v_s is the ion sound velocity. (For simplicity, corrections due to ion temperature and damping are neglected.) Since ω is not simply proportional to k , a small frequency mismatch must be overcome, i.e., the decay is quasi-resonant.

The theory of quasi-resonant decay is readily obtained from the two-fluid equations. An equation for the low-frequency

density fluctuation is obtained and then linearized by assuming for the density n :

$$n = n_0 + \delta n + \tilde{n}, \quad (46)$$

where n_0 is the uniform background-plasma density, δn is the density fluctuation associated with the large-amplitude ion wave with frequency ω_0 and wave-number k_0 , and $\tilde{n}$ is the fluctuation associated with the perturbation.

A dispersion relation is obtained by truncating to two modes with lower wave numbers. This dispersion relation predicts that the maximum growth rate occurs for decay into the half-harmonic ($k_0/2$) and is

$$\gamma_0 = \frac{k_0 v_s}{4} \frac{\delta n}{n_0}. \quad (47)$$

If we allow for the frequency mismatch $\Delta\omega$, growth requires $\gamma_0 > \Delta\omega$, or

$$\frac{\delta n}{n_0} > \frac{3}{4} k_0^2 \lambda_{De}^2. \quad (48)$$

However, derivation of this decay instability omits a very important effect, harmonic generation, which is also quasi-resonant and which leads to a steepening of the large-amplitude ion wave. We carried out simulations to include these higher-wave-number waves. In our code, we used a two-fluid description and

Fig. 2-68. Evolution of (a) driven mode ($k\lambda_{De} = 0.05$) and (b) half-harmonic mode ($k\lambda_{De} = 0.025$) in a two-mode simulation. The energy damping rate is 0.02 of the ion-wave frequency. Here ω_{pi} is the ion plasma frequency.

drove up the large-amplitude ion wave by a ponderomotive potential. An ion-wave damping proportional to wave number was also used, as is appropriate for Landau damping. For more details on the code, see Ref. 112.

Two examples illustrate our results. In the first, only two modes are included in the simulations—the driven mode and its half-harmonic. As shown in Fig. 2-68, the driven mode ($k\lambda_{De} = 0.05$) is excited to

an amplitude of $\delta n/n = 0.10$. The amplitude of the half-harmonic mode ($k\lambda_{De} = 0.025$) then exponentiates with a growth rate in very good agreement with the two-mode theory. Indeed, this half-harmonic mode efficiently takes energy from the driven mode and subsequently dominates.

The behavior is qualitatively different if ion waves with higher numbers are included in the simulation. In this second example, the same driven mode is again excited, but now mode numbers up to 20 are retained. Hence this simulation allows the competitive effect of harmonic generation. Evolution of the amplitude of the driven mode, its half-harmonic, and its second harmonic is shown in Fig. 2-69. The amplitude of the driven mode now saturates at $\delta n/n \approx 0.048$, concomitant with significant excitation of the second harmonic. In contrast to predictions of the two-mode theory, the half-harmonic wave does not grow, even though ion-wave damping was reduced by a factor of 2.

Many different simulations were carried out, varying the $k\lambda_{De}$ of the driven mode and the strength of the ponderomotive driver. The above results were typical. No growth of the half-harmonic was observed when higher-wave-number modes were included. Growth of the half-harmonic mode was observed in only one case in which the zero-order state became extremely distorted. Finally, we note that half-harmonic generation was not observed in recent detailed experiments¹¹³ on ion-wave excitation. However, the ion-wave damping would have been sufficient to suppress the half-harmonic generation in these experiments, even if the truncated mode analysis were valid.

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Discharging of Heavy-Ion Fusion Targets

The accumulation of electrostatic potential due to the charge of the incoming ion beams, to a level high enough to repel or deflect the incoming beams, has been a concern for heavy-ion fusion

Fig. 2-69. Evolution of (a) driven mode ($k\lambda_{De} = 0.05$), (b) half-harmonic mode ($k\lambda_{De} = 0.025$), and (c) second harmonic mode ($k\lambda_{De} = 0.1$). In this simulation, 20 modes were allowed, and the energy damping rate was 0.01 of the ion-wave frequency.

(HIF).¹¹⁴ Here, we show that target charging is not a problem. Two mechanisms are considered that reduce the potential to harmless levels:

- If the chamber residual gas density is high, electrons generated by photoionization by blackbody radiation from the heated target provide the conductivity to discharge the target.

- If the residual gas density is low, or if the target surface is too cold, target charging is limited by electrostatic repulsion of target ions to large radii.

We also estimate that electrons accelerated onto the target by the remaining potential do not produce excessive preheat.

For ICF with ion beam drivers, small values of beam ion charge-to-mass ratio Z/A (where Z is the charge state and A is the relative atomic mass) reduce space charge effects in the accelerator; larger Z/A reduces accelerator costs. We are led to consider $A > 200$, $Z \simeq 3$.

Especially for larger values of beam Z , it has been speculated repeatedly¹¹⁴ that the target will charge during the pulse to a potential comparable to the beam energy, reducing energy delivered to the target. Others have estimated this will not occur, but there is no definitive treatment in the literature.

The basis of the concern is in these ratios:

$$\frac{Ze\phi}{\text{K.E.}} = \frac{2(E^2 \text{ field energy})}{W}$$

$$= 0.90 \frac{W Z^2}{(\text{K.E.})^2 r_t} \quad (49)$$

where

- $Ze\phi$ = beam ion energy loss (GeV)
- K.E. = beam ion kinetic energy (GeV)
- W = total energy on target (MJ)
- r_t = target radius (cm).

For example: 6 MJ in a beam of 10-GeV ions onto a 1-cm target results in $Ze\phi/\text{K.E.} = 0.054Z^2$, or $\sim 1/2$ for $Z = 3$. One quarter of the beam energy has gone to electrostatic field energy instead of to the target. Additional energy may be lost by deflection of some ions away from the target.

Fortunately, target charging is reduced by several effects. The positive charge of

the beam will effectively be at a radius much greater than the target radius, reducing the potential for two reasons:

- As it heats, the target radiates and photoionizes any chamber gas. When the photoelectrons are drawn to the target, the remaining positive charge is that of the ionized gas.

- Even in vacuum, target charging is limited, by electrostatic expulsion of target ions, to potentials much less than the beam energy.

This last case, the most pessimistic, is considered here first. It is an interesting problem of transient space-charge-limited flow.

Other mechanisms that may eliminate or reduce target charging have also been suggested:

- The beam may have a halo that is ionized in passing through the limb of the target to a much higher charge state, say $+70$, leaving its electrons at the target as it passes on. If a few percent of the beam were to pass through the limb, this process would make available enough electrons to neutralize the target. Unfortunately, this mechanism depends on details of the beam that are unknown, so no reliable calculations can be made at present.

- The target is discharged by its supporting structure, depending on its impedance.

Lastly, if it is possible to charge and current-neutralize the beam all the way to the target, the question of target charging may not arise at all.

Vacuum Case. For the extreme case of an unsupported target in an evacuated chamber, we use two methods, both assuming spherical symmetry:

- An incomplete but accurate theoretical model.
- Self-consistent particle-in-cell simulations.

A simple model problem predicts the final target potential. For the transient, space-charge-limited discharge of the target, we adapt a Lagrangian method used for ac analysis of radio tubes.¹¹⁵ First the trajectories of expelled ions are computed. Then the final target potential is expressed in terms of these trajectories.

We assume spherical symmetry and assume that target ions are emitted at a

rate that keeps the target nearly neutral and that beam ions are much faster than expelled target ions, so we neglect the ion beam's in-flight charge until later.

These assumptions determine the equation of motion of a spherical shell of expelled target ions. The electric field on the shell is due solely to the charge enclosed inside it. Assuming no crossing of trajectories, shells emitted earlier contribute no electric field. Shells emitted later reduce the target charge, so they add no net enclosed charge, and therefore also no electric field. Therefore the charge inside a shell of target ions is the same as the charge deposited on the target by the ion beam since that shell left the target. Therefore a shell's equation of motion is simply

$$\ddot{r} = \frac{q}{mr^2} \int_{t_0}^t dt' I_1(t'), \quad (50)$$

where q and m refer to target ions, $I_1 = ZI_0$ is the beam electric current, and t_0 is the time the shell left the target.

This problem is remarkably simple for a rectangular current pulse. We have

$$\ddot{r} = \frac{qI_1}{mr^2}(t - t_0), \quad \text{with} \\ r(t_0) = r_1, \quad \dot{r}(t_0) = 0. \quad (51)$$

All shells follow the same $(r, \dot{r})$ trajectory given by solving the normalized equation of motion

$$\frac{d^2x}{d\tau^2} = \frac{\tau}{x^2}, \quad (52)$$

where $x = r/r_1$ is normalized to the target radius r_1 , and $\tau = (qI_1/m)^{1/3}[(t - t_0)/r_1]$.

The target potential at the end of the pulse, $t = T$, has two parts: one part of the final target potential is

$$-\phi[r(T)] + \phi(r_1) = \int_{r_1}^{r(T)} E dr \quad (53)$$

at the end of the pulse, integrated out to the ion that was emitted first. For constant beam current I_0 , all target ions follow the same $r(t)$ trajectory, experiencing the same E at the same radii.

Therefore $\int E dr$ is the same as the $\int E v dt$ experienced by that first target ion along its trajectory, and is given by its final kinetic energy divided by q .

The remaining contribution to the final target potential is due to the electric field at larger radii,

$$-\phi(\infty) + \phi[r(T)] = \int_{r(T)}^{\infty} E dr = \frac{I_1 T}{r(T)}. \quad (54)$$

The target potential at the end of the pulse can be expressed in two ways:

$$\phi = \left(\frac{mI_1^2}{q} \right)^{1/3} \left[\underbrace{\frac{1}{2} \left(\frac{dx}{d\tau} \right)^2}_{r < r_1} + \underbrace{\frac{\tau}{x}}_{r \geq r_1} \right]_T \quad (55)$$

$$= \frac{I_1 T}{r_1} \left[\frac{1}{2\tau} \left(\frac{dx}{d\tau} \right)^2 + \frac{1}{x} \right]_T. \quad (56)$$

The first expression is in Child-Langmuir form. The second form facilitates comparison to the target potential neglecting discharge, $I_1 T/r_1$. Since $dx/d\tau < 3$ and $x > \tau \gg 1$, as we see below for parameters of interest, Eq. (56) shows that the target potential is greatly reduced.

A solution of the scaled equation of motion, Eq. (50), provides a simple evaluation of the final target potential. We integrate from $t = 0$, $x(0) = 1$, $(dx/d\tau)_0 = 0$ to $t = T$, producing a table or graphs of $(dx/d\tau)^2/2\tau + 1/x$ versus x and x versus τ . In Fig. 2-70 we tabulate

$$\left[\frac{1}{2\tau} \left(\frac{dx}{d\tau} \right)^2 + \frac{1}{x} \right]$$

in Eq. (56) versus the normalized time

$$\tau(T) = \left(\frac{qI_1}{m} \right)^{1/3} \frac{T}{r_1} \\ = 0.95 \left(\frac{Z_1 I_{kA}}{A_1} \right)^{1/3} \frac{T_{ns}}{r_{1cm}}, \quad (57)$$

where Z_1 and A_1 refer to target ions.

The final target potential is negligible compared to the ion beam energy. For numerical examples that show the range

of results, take 6 MJ delivered at $I_i = 30$ kA, 10 GeV, in $T = 20$ ns, and assume the expelled target ions are protons (a few nanograms of hydrogen is enough).

• For singly ionized beam ions and $r_t = 1$ cm, we find $\tau = 59$. From the table, we find $x = 137$ and $(dx/d\tau)^2/2\tau + 1/x = 0.064$. Therefore the ion cloud extends to 137 cm, and the target potential is reduced from 540 to 35 MV.

• For triply ionized beam ions and $r_t = 0.3$ cm, we find $\tau = 284$. From the table, we find $x = 760$ and $(dx/d\tau)^2/2\tau + 1/x = 0.016$. Therefore the ion cloud extends to 228 cm, and the target potential is reduced from 16 GV to 253 MV.

ZOHAR particle-in-cell simulations avoid simplifications made in the analytic model, yet give similar results. The analytic model assumes beam ions are so fast that their in-flight charge density can be neglected compared to that of the expelled ions—but their charge density turns out to be nearly as large. The model assumes maximum beam ion energy loss is given by the final target potential, i.e., $\int E dr$ integrated at a fixed time; in fact the potential is $\int E v dt$ integrated along the trajectory. Both these points concern the fact that expelled target ions reach speeds comparable to that of the beam ions. The simulations could also be used with more realistic pulse shapes, if desired. ZOHAR simulation shows beam ion energy spread down only 39 MV vs 33 MV for the simple model, in a representative example.

Photoionization of Chamber Gas. We have used x-ray output spectra from the LASNEX modeling of targets, and photoionization cross sections for lithium atoms and ions calculated by an independent particle approximation (IPA) method, to calculate the discharging of the target by photoelectrons from ionization of the chamber gas. We use a particle-in-cell code built on a spherical version of the ES1 code. All species receive a kinetic treatment: beam ions, photoelectrons, and lithium ions. There are no expelled target ions. All particles reaching the target are absorbed. The beam is created with space-time dependence given by the target drive in the LASNEX calculation, whose x-ray output

Fig. 2-70. Dependence of target potential multiplier on normalized time τ .

is used in ionization rate equations for the lithium vapor at a density of 10^{12} cm^{-3} (3×10^{-5} Torr). At all times during the pulse, the target potential is < 4 MV. For higher vapor densities the potentials will be even smaller.

Discussion. The potentials at positions where photoelectrons are created are ~ 1 MV, so the photoelectrons form a cloud of relativistically hot electrons. This may be of interest in considerations of beam propagation.

Electrons accelerated into the positively charged target cause minor preheat, according to preliminary estimates. In the modeling discussed above, we find less than 10^3 J/g preheat deposited in the DT fuel up to the beginning of the high-intensity part of the pulse. Preheat up to 10^5 J/g is acceptable.

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Photoionization of Beam Ions

In addition to the role of photoionization in the context of ionizing the gas in the target chamber (see the preceding article "Discharging of Heavy-Ion Fusion Targets"), the importance of photoionization of the ion beam itself by thermal radiation emitted by the target as it heats was pointed out by Herrmannsfeldt.¹¹⁶ Rough estimates show this could be of interest because of its effect on the focusing

of the beam. We have used x-ray output spectra from the LASNEX modeling of targets driven by 1 to 5 MJ, and photoionization cross sections for Pb^{+n} to calculate the position and time-dependent photoionization of a beam of Pb^{+3} ions. Then we made estimates of this effect on beam dynamics.

The calculation proceeds as follows: From files written by LASNEX at intervals of about 1 ns, we extract x-ray output spectra in the form of energy per photon bin cumulative from the start of the pulse. Although the beam ion speed, about one-third that of light (c), is not relativistic, it is large enough to require corrections. As viewed by the beam ions, the photon energies are Doppler-shifted up and their number flux increased by the factor $\gamma(1 + \beta)$, but the time of interaction is reduced by the same factor (photon number is invariant). Here $\beta = v_{\text{beam}}/c$, $\gamma = (1 - \beta^2)^{-1/2}$ refer to the beam ions. Retardation is taken into account. Photoionization cross sections for lead ions are calculated by conventional one-electron methods.¹¹⁷ The photon frequencies at which the output spectra and the cross sections are known are different. The cross sections are interpolated, and the Doppler-shifted output spectra distributed, from their respective

frequency bins to a common fine binning on which their product is integrated to form the ionization rates.

Ionization is small at large radii, where deflections due to change in charge state can most easily degrade focusing. However, near the end of the pulse, almost all beam ions are in a higher charge state before they reach the target, and, by the end of the pulse, half are in a higher charge state at about 20 cm from the target, depending on the target's temperature history. For photon energies of interest (most are in the range of a few hundred electron volts) the major contributions to the cross section of Pb^{+3} are from the 14 electrons in 4f states. Because there are 14 more weakly bound electrons in outer shells, Pb can be ionized more than 14 times with little reduction of the cross section. Figure 2-71 shows the results of integrating the ionization rate equations. The curves show the fractional populations of successively higher charge states. At 2 cm from the target, $\bar{Z} \approx 15$.

Unless neutralized, the rising charge of the beam ions would cause the beam to expand and the deposition spot on the target to enlarge. Estimates of this effect depend on one's assumptions about co-moving electrons injected with the beam, and electrons picked up from the chamber gas and from photoionization of the beam ions. We have calculated the evolution of a beam that begins with no accompanying electrons but whose photoelectrons remain close to the parent ions. In this model, the radial evolution of a cross-sectional slice of the beam is calculated by a one-dimensional particle-in-cell method. The beam, carrying 32 kA with emittance 30 mm·mrad, where Z is the charge state, has initially a Kapchinskij-Vladimirskij distribution and focuses to a 3-mm-radius spot without photoionization. With photoionization, 20% of the beam is outside the 3-mm spot and about 2% is outside 5 mm.

More accurate studies await better modeling of the electrons. Better beam neutralization, especially if by co-injected electrons, would reduce the beam blow-up. While no difficulties have appeared clearly, photoionization of the beam and

Fig. 2-71. Fractions of beam ions in successively higher ionization states versus distance from target in centimeters. Curve A is for $Z = 3$, B is for $Z = 4$, etc. The time is at the end of the pulse.

the chamber gas will certainly be a factor in planning beam propagation modes.

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Deposition of Heavy-Ion-Beam Energy on Targets

In 1987 we investigated the possibility that electron anisotropy produced by streaming instabilities might rapidly transport energy away from the incident ion beam, thus dissipating energy in the expanding target corona. We considered transverse instabilities that are driven by these anisotropic electron distributions. Indications are that for parameters of interest to HIF, these modes will not grow rapidly enough to dissipate much energy. In regions of higher density where collisional effects become important in reducing electron anisotropy, we demonstrated gradient stabilization of the ion-electron two-stream mode. We found no significant deleterious instabilities in our study. In this article we describe our models and the rationale behind these conclusions.

Model. The energy source driving the various instabilities in our study is the incident ion beam. We assume that this beam is both charge and current neutralized by a co-moving beam of electrons. The co-moving electrons exist either by design of the accelerator or by the ion beam's first interaction with matter, where the nonneutral beam encounters electrons that are pulled along. Without concern for the front of the ion beam, we considered instabilities that the remainder of the neutralized beam can experience.

As discussed in the 1986 Laser Annual,¹¹⁸ the first interaction will be between the electrons following the ion beam and the electrons of the first plasma encountered. We expect rapid growth and saturation of the electron-electron two-stream instability, producing an elongated tail on the electron distribution that has the monotonically decreasing velocity profile characteristic of the saturated electron-electron mode. The slower, ion-electron two-stream mode is also stabilized by such electron distributions.

Fig. 2-72. Electron and ion distributions (a) as they would appear after the early, energetically insignificant, electron-electron two-stream instability saturates; (b) contours of the saturated electron distribution.

The electron-tail density then becomes slightly greater to allow for the velocity-space bump due to the ions.

The neutralized ion and electron distributions as they might exist after the electron-electron mode saturates are shown in Fig. 2-72(a). Because the electrons have little mass, the energy in this mode is insignificant compared with the ion beam's large kinetic energy. We take this configuration as the given beam that impacts the target corona.

Transverse Electromagnetic Modes Driven by Electron Anisotropy. A distribution like the one in Fig. 2-72(a) is anisotropic and so is subject to transverse instabilities. Our concern was that these modes would negatively affect ion-beam energy deposition. The transverse electromagnetic (EM) mode, in particular, generates a transverse magnetic field that grows in the colder directions and that acts to rotate the velocity vectors and reduce the anisotropy. We can study these transverse instabilities by 2-D, electromagnetic, particle-in-cell (PIC) simulations.

With the implicit PIC code AVANTI,¹¹⁹ we start the simulation with the velocity distribution in $v_{||}$ shown in Fig. 2-72(a). We represent this distribution as the sum of four particle groups:

1. Target ions with a scaled density of 1.0, no drift velocity, and a thermal velocity $u_{th} \sim 0.002c$ (intended to be negligible).

Fig. 2-73. PIC simulations at $t = 75/\omega_{pe}$: (a) electron-beam cross section showing beam filamentations; (b) vector plot of the transverse magnetic field, where filamentation is more evident; and (c) growth in magnetic-field energy resulting from the EM mode, measured at $0.03/\omega_{pe}$.

2. Beam ions with a density of 0.25, parallel drift velocity $u_{\parallel} = 0.25c$, and the same thermal velocity as the target ions.

3. Target electrons with a density of 1.0, no drift velocity, $u_{th} = 0.05c$, and a temperature ratio $T_{e\parallel}/T_{e\perp} = 1.2$.

4. Beam electrons with a density of 0.25, $u_{e\parallel} = 0.25c$, $u_{th} = 0.15c$, and $T_{e\parallel}/T_{e\perp} = 9.0$.

The sum of the distributions from groups 3 and 4 provide the electron distribution shown in Fig. 2-72(b). We chose an ion-to-electron mass ratio in both beam and target of 2000—an unrealistically small value.

We simulated this distribution using $\Delta t = 1/\omega_{pe}$ on a 60×60 uniform mesh covering the range 0 to $10c/\omega_{pe}$ in both x and y . Here ω_{pe} is computed using the target plasma's electron density. We modeled this phenomenon in a plane perpendicular to the beam propagation. The beam (particle groups 2 and 4) was given an initial radius of $4 c/\omega_{pe}$. The target (groups 1 and 3) also has a circular cross section with a slightly larger radius of $5 c/\omega_{pe}$. As an example, we assume a target electron density of $10^{16}/\text{cm}^3$, consistent with the corona's front edge where collisional stabilization is not yet important. This assumption gives $\omega_{pe} = 5.64 \times 10^{12}/\text{s}$. At this density the beam radius simulated is $2.1 \times 10^{-2} \text{ cm}$.

The electron distribution at $t = 75/\omega_{pe}$ or $1.33 \times 10^{-11} \text{ s}$ and at our assumed density is shown in Fig. 2-73(a); some beam filamentation is evident. This filamentation of the fast beam electrons provides a concentration of charge that displaces target electrons; the massive beam ions experience no discernible

transverse displacement. No appreciable deviation from charge neutrality can occur with the relatively small Debye length $\lambda_{De} = 2.66 \times 10^{-4} \text{ cm}$.

The filamentation of the fast-electron component, resulting in a local concentration of current, is even more apparent in the vector plot of the transverse magnetic field at the same time, shown in Fig. 2-73(b). The growth in field energy as a function of time for this configuration is shown in Fig. 2-73(c). The mode has saturated with a magnetic-field amplitude of roughly $4.0 \times 10^{-2} \omega_{ce}/\omega_{pe}$ or $1.3 \times 10^4 \text{ G}$. We measure the growth rate to be $0.03\omega_{pe}$ and the typical transverse wavelength to be $4c/\omega_{pe}$.

Excitation of a transverse EM mode due to anisotropy in the velocity distribution is described in standard texts. From Krall and Trivelpiece¹²⁰ we find that the growth rate and wave number are given by

$$\gamma_{\max} = \left(\frac{8}{27\pi} \right)^{1/2} \left(\frac{T_{e\perp}}{m_e c^2} \right)^{1/2} \times \frac{T_{e\perp}}{T_{e\parallel}} \left(\frac{T_{e\parallel}}{T_{e\perp}} - 1 \right)^{3/2} \omega_{pe} \quad (58)$$

and

$$k_{\max} = \frac{1}{\sqrt{3}} \left(\frac{T_{e\parallel}}{T_{e\perp}} - 1 \right)^{1/2} \frac{\omega_{pe}}{c}. \quad (59)$$

In Eqs. (58) and (59), $T_{e\parallel}$ ($T_{e\perp}$) is the effective temperature in the parallel (perpendicular) direction.

For our parameters, Eqs. (58) and (59) give $\gamma_{\max} = 0.041\omega_{pe}$ and $k_{\max} = 1.64\omega_{pe}/c$, or a typical transverse wavelength of $3.82c/\omega_{pe}$. This agreement is close enough to convince us that Eqs. (58) and (59) represent the correct physics. The theoretical growth time τ_G is $23.4/\omega_{pe}$. If we again assume a corona density of $10^{16}/\text{cm}^3$, $\tau_G = 4.15 \times 10^{-12}$ s, during which time the beam travels 3.11×10^{-2} cm. (The beam has traveled roughly 1 mm in the $75/\omega_{pe}$ required to see the filamentation in Fig. 2-73.)

More importantly, ion movement in the transverse direction due to this process is small; the typical ion radius of curvature due to the maximum magnetic field is roughly 60 cm for $m_i/m_e = 2000$ and is even larger for realistic ion masses. Even if these protons were exposed to the maximum magnetic field throughout the entire corona (≤ 1 cm), they would deflect $\leq 10^{-2}$ cm—an entirely negligible distance. We conclude that ion-beam modification in the target corona due to electron-anisotropy-driven modes is insignificant.

Gradient Stabilization of the Ion-Electron Mode at Higher Densities. At higher target-plasma densities, we expect the target electrons to provide charge and current neutralization. In this case, shown in Fig. 2-74, a two-stream instability between the heavy ions and the target electrons could again produce an electron tail.¹²¹ This electron tail would be similar to that shown in Fig. 2-72(b) but of lower density since it only needs to compensate for the velocity bump due to the ion beam—there are no electrons moving with the beam as was the case with the initial beam model. As we will show, the weaker two-stream instability is readily stabilized by density gradients¹²²⁻¹²⁴ in the target plasma.

We simulated this gradient stabilization by modeling a 1-D inhomogeneous plasma into which a high-velocity ($v = 0.25c$) ion beam propagates. Starting with the unstable ion-electron distribution shown in Fig. 2-74 throughout the simulation region and emitted on the upstream side of this region as time progresses, we find that the ion-electron mode shown in Fig. 2-75(a) goes unstable and saturates in a few growth lengths, given by the

Fig. 2-74. Electron and ion distributions resulting from relaxing the electron anisotropy by the action of transverse EM modes. These distributions are initialized throughout the simulation region and emitted from the left with time.

growth rate times the ion-beam velocity. This growth rate is given by the classical formula

$$\gamma = \frac{\sqrt{3}}{2} \left(\frac{n_{\text{beam}} m_e}{2 n_{\text{target}} m_i} \right)^{1/3} \omega_{pe} \quad (60)$$

For our parameters, Eq. (60) gives $\gamma = 0.087\omega_{pe}$, which agrees well with our simulation value of $0.092\omega_{pe}$. Our parameters were $n_{\text{beam}} = 1.0$, $n_{\text{target}} = 10.0$, and $n_e = 11.0$; numerical parameters were $0 \leq z \leq 30.0c/\omega_{pe}$, 480 mesh points, and $\Delta t = 0.1/\omega_{pe}$. We used an artificially small $m_i/m_e = 50.0$ and a narrow-velocity ion beam to accentuate the instability in this study.

As is evident in Fig. 2-75(a), the distribution is introduced on the left boundary and goes unstable. Before the ion beam has propagated through one-third of the simulation region, the mode is saturated by forming the electron tail, evident by electrons pulled out to large velocities in the v vs z plots. Growth rates can be inferred from this distance and from the ion beam's velocity. But the more accurate value, given above, is obtained by monitoring the histories of electrostatic-field energy.

The energy history from the homogeneous case in Fig. 2-75(a) displays an artifact of our method: the initial configuration, with no field energy, is unstable throughout the entire simulation region. This results in a peak around $40\omega_{pe}$ in the energy history that later relaxes to the steady value by 80 to $100\omega_{pe}$, when only the left third of the region contains

Fig. 2-75. Electron phase space (left) and corresponding histories of electrostatic-field energy (right), showing the effect of gradient stabilization. Density scalelengths are (a) ∞ , (b) 15.0, (c) 6.0, and (d) 3.0. The ion-electron mode is increasingly inhibited and ultimately stabilized as density scalelength decreases.

any unstable distribution. Regions with the stable electron tail have little electric field.

We investigated the effects of the stationary target's density gradient by introducing such a gradient in the direction of the beam velocity. The behavior of the ion-electron mode in the presence of increasing density gradients is shown in Fig. 2-75(b)-(d).

Figure 2-75(b) shows a simulation with the same parameters as in the homogeneous case of Fig. 2-75(a) but with a density inhomogeneity added to the target ions and electrons that increases the density from 10 to 20. The density profiles have the form

$$n(z) = n_0 \left(1 + \frac{z}{L_n} \right), \quad (61)$$

where n_0 is the density on the left and L_n is the density scalelength.

The density of the developing electron tail in Fig. 2-75(b) and its corresponding energy history show evidence of a weaker ion-electron two-stream instability. This trend continues through the next two levels of increasing inhomogeneity. As the gradient scalelength decreases, the mode becomes mismatched and is stabilized. With the largest density gradient in Fig. 2-75(d), no significant electron tail is evident, and the energy history shows no instability at any time.

A heuristic argument characterizes the stabilization process. The formula for the most unstable wavelength of the streaming instability,

$$k_{\text{most unstable}} = \frac{\omega_{pe}(z)}{v_{\text{beam}}}, \quad (62)$$

reveals the dependence on plasma density and therefore on position z . We expect that if the most unstable mode changes significantly before it has a chance to grow, the mode will be stabilized.

We define a mismatch distance

$$L_{\text{mis}} = \left(\frac{2v_{\text{beam}} L_n}{\omega_{\text{pe}}} \right)^{1/2}, \quad (63)$$

where L_n is a density-gradient scale-length. This expression arises from an expansion of Eq. (62) and an assumption that effective mismatch occurs when the relative change in k is 0.5. We expect stabilization when $L_{\text{mis}}/L_{\text{growth}} < 1$, where $L_{\text{growth}} = v_{\text{beam}}/\gamma$. These conditions combine to give the relation for stability

$$L_n \leq 1.058 \left[\left(\frac{m_i}{m_e} \right)^{2/3} \times \left(\frac{n_0}{n_{\text{beam}}} \right)^{1/6} \frac{v_{\text{beam}}}{c} \right] \frac{c}{\omega_{\text{pe beam}}}. \quad (64)$$

For our parameters, Eq. (64) reduces to $L_n < 5.27c/\omega_{\text{pe beam}}$; for the three cases we considered, $L_n = 15.0, 6.0$, and 3.0 . The values of the ratio $L_{\text{mis}}/L_{\text{growth}}$ are 2.13, 1.34, and 0.95 for Fig. 2-75(b) through (d), respectively.

An even clearer demonstration of gradient stabilization is apparent in the histories of electrostatic-field energy shown on the right in Fig. 2-75. In the homogeneous case, we see clear evidence of the instability. As the gradient scalelength decreases, the mode becomes so mismatched that the electrostatic-field energy remains near the thermal level.

These results indicate that, in regions where the target density is 10^{18} , gradient stabilization of this mode will occur for density scalelengths of order 0.02 cm or less for our case with $m_i/m_e = 50$. This length increases with increasing ion mass because the heavier ions simply take longer to move. HIF beams are expected to be focused to roughly a 0.1-cm-radius spot; early in the pulse, typical calculations show the expanding corona to be this depth or smaller. Thus, we expect this instability to pose no threat in terms of degrading beam energy.

Conclusion. We investigated the possibility that streaming instabilities and associated transverse instabilities might degrade the transport of ion-beam energy through the target corona. We found no significant problem in any regime considered. The electron-electron two-stream instability occurs readily but contains no significant energy because of the small electron mass. Transverse instabilities that occur because of the anisotropic electron distribution that results from two-stream saturation stabilize at benign levels. Finally, any possibility that an ion-electron two-stream mode can significantly degrade beam energy is reduced by the mode's gradient stabilization for parameters that seem reasonable for HIF.

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Self-Consistent Particle Simulations and Emittance Growth of Heavy-Ion Beams During Longitudinal Compression

Introduction. It is necessary to longitudinally compress a heavy-ion beam, emerging from an induction linac in order to generate enough power for high-gain implosions. To relieve the constraints in the final focusing lens design, it is probably advantageous for the compressed beam to have uniform line-charge density and uniform longitudinal momentum. The basic principle of the compression scenario for achieving this final beam configuration and the longitudinal dynamics have been described in our previous report.¹²⁵ With the newly expanded capabilities of the CONDOR 2-1/2-D particle code, we are now able to make a much more detailed study of beam dynamics during compression. In this article we will show that, in order to simulate the compression self-consistently, it is necessary to vary the longitudinal temperature, transverse temperature, beam density, and beam radius along the beam. The result of the particle simulations, the emittance growth, and the effect of longitudinal temperature on compression will be discussed.

Self-Consistent Particle Simulations and Emittance Growth of Heavy-Ion Beams During Compression. We start with a 3-m-long beam and let it go through self-compression until the length reaches 1 m. This is because, as mentioned in Ref. 125, the line-charge density λ can be described analytically along the beam longitudinal coordinate z if the beam is less than 3 m long and if the compressed configuration is 1 m long. Thus, the easiest way to calculate the beam initial parameters is to start with a 3-m-long beam. The initial λ of the 3-m-long beam is shown in Fig. 2-76(a). In the first part of this article, we will describe the method of obtaining the initial parameters for a 3-m-long beam in radial equilibrium and of setting up the particle simulation. In the second part of this article, results from the particle simulation will be discussed.

Obtaining the Initial Parameters for a Beam in Radial Equilibrium. Since λ varies along z , the beam radius also varies along z . Near the two ends of the beam, λ decreases to low values and thus the beam radius is predominantly determined by the transverse temperature, i.e., the beam is no longer dominated by space charge. Thus, it is necessary to employ the envelope equation with the emittance term in order to

determine the equilibrium beam radius as one of the initial conditions for particle simulation.

The envelope equation that follows the trajectory of a slice of the beam has the form¹²⁶

$$\frac{\partial^2 \bar{a}}{\partial t^2} = \beta^2 c^2 \left(-k\bar{a} + \frac{\bar{\epsilon}^2}{\bar{a}^3} + \frac{Q}{4\bar{a}} \right), \quad (65)$$

where

$$\bar{a}^2 \equiv (1/N) \sum_{i=1}^N x_i^2. \quad (66)$$

Here x_i is the distance from beam center on coordinate x , which is perpendicular to the beam longitudinal axis, of particle i . The total number of particles in a thin slice of the beam is denoted by N . For a uniform radial density profile, $\bar{a} = (1/2)a$ where a is the beam radius. The root-mean-square (rms) emittance is $\bar{\epsilon}$ where the bar denotes rms value; the averaged magnetic field gradient is k ; the generalized perveance¹²⁶ Q is proportional to the line-charge density λ ; c is the speed of light; and the relativistic parameter is β .

Note that it is accurate to assume that the beam has a uniform radial density profile, except near the two ends, if the

Fig. 2-76. Beam initial parameters. (a) Line-charge density vs z . (b) Radial distribution of beam particles vs z . (c) Longitudinal velocity vs z . (d) Transverse temperature vs z . (e) Longitudinal temperature vs z . (f) Momentum in x direction vs x . (g) Normalized emittance in x direction vs z .

initial equilibrium profile is uniform. This can be proved by the following argument. If Q varies slowly, then $\partial^2 r / \partial t^2 \rightarrow 0$ for any radius r . For most of the beam, the beam is space-charge dominated, i.e., the emittance term in Eqs. (65) and (66) can be neglected. Thus, from force balance, Eqs. (65) and (66) give

$$\tilde{r}_{\lambda} = \text{const}, \quad (67)$$

where λ_r is the line-charge density to an arbitrary radius r . This condition can be satisfied for arbitrary r only if the density is constant. Thus, we have proved that the radial density profile is uniform for a space-charge dominated beam in a linear external field. This condition together with Eq. (67) also implies that the volume charge density does not change during longitudinal compression in the region of the beam that is dominated by space charge. Consequently, the radial expansion velocity of a fluid element inside the beam varies linearly with the radius.

The normalized emittance $\epsilon_n = (\beta\gamma)\epsilon$ is constant along the beam but $\beta\gamma$ is a function of z because of the initial longitudinal velocity v_z tilt. However, the velocity tilt is of order 10^{-2} times the beam center velocity. Thus, we will treat $\beta\gamma$ as a constant. For a sample calculation, we choose a heavy-ion beam at the final compressed configuration having $a = 3$ cm, $\lambda = 6.9 \times 10^{-5}$ C/m, the edge emittance $\epsilon = 3 \times 10^{-5}$ mrad (Ref. 127), and a corresponding transverse temperature of about 6.2 keV. To solve for radius, we set the right-hand side of Eqs. (65) and (66) equal to zero. The calculated particle distribution within the beam envelope as a function of z is shown in Fig. 2-76(b). Near the two ends of the beam, the radius expands to about twice the calculated value (with constant density profile) after approximately 10^{-8} s. This is due to the fact that the density profile is nonuniform near the ends because the beam is no longer space-charge dominated.

The transverse temperature $T_{\perp}$ and longitudinal temperature T also vary along the beam. The transverse temperature varies along the beam because $T_{\perp}$ follows the adiabatic 2-D compression law ($\gamma = 2$) (Ref. 125), i.e.,

$$T_{\perp} = T_0 \left[\frac{a_0}{a(z)} \right]^2. \quad (68)$$

Here, the subscript 0 refers to quantities at beam center. Since we have calculated the beam radius, $T_{\perp}$ can be obtained and is shown in Fig. 2-76(d) as a function of z . Note that Eq. (68) holds only if there is negligible coupling between the longitudinal and transverse energies of any particle. Later in this article, we will show that the coupling is indeed weak.

To estimate the maximum allowable T note that for proper focusing of the beam onto the target, longitudinal momentum variation of the beam $\Delta p_z / p_z$ at the final focusing lens should typically be less than 10^{-3} to 10^{-4} (Ref. 128). Thus the maximum allowable T at the compressed configuration is of order (10 keV).

Before calculating $T_{\parallel}$ as a function of z , we need to estimate the importance of mixing: the smoothing of density and temperature gradients along the beam due to pressure. To do this, we estimate ratio of the pressure force due to the longitudinal density and temperature gradients and the electrostatic force due to the gradient in λ . From the beam longitudinal equation of motion,¹²⁵ the ratio can be expressed as

$$\frac{(1/\rho)(\partial P / \partial z)}{(q/m)(1/4\pi\epsilon_0)g(\partial\lambda/\partial z)} \approx O\left(\frac{8\pi\epsilon_0 T}{q\lambda}\right), \quad (69)$$

where ρ is the density; q is the charge of the ions; m is the ion mass, P is thermal pressure, and g is of order unity.¹²⁶ If we take $T_0 = 10$ keV at the beam center, then Eq. (69) shows that the ratio of the forces is of order (10^{-2}) . Thus, mixing can be neglected and the variation of the longitudinal temperature follows the 1-D adiabatic compression law ($\gamma = 3$), i.e.,

$$T = T_0 \left[\frac{\lambda(z)}{\lambda_0} \right]^2. \quad (70)$$

Using this relation, T vs z is plotted in Fig. 2-76(e). The existence of T is also evidenced on the v_z vs z plot as shown in Fig. 2-76(c). In the absence of T , v_z vs z would be just a zero-width straight line.

Although the above estimate is made for compressing a beam from 3 m to 1 m,

the conclusion should also apply to compressing a beam from 10 m to 1 m. One may tend to think that it will take about four times longer to compress a beam from 10 m to 1 m and thus the mixing should be four times as bad. This is not the case, however. This is because the ratio given by Eq. (69) is inversely proportional to the beam length, while the compression time is proportional to the beam length.

Incorporating the initial parameters of the beam as specified above, the CONDOR particle code is used to simulate the self-compression. The model employed 46 000 particles. The initial transverse momentum p_x is plotted vs x in Fig. 2-76(f); this plot is essentially identical to the plot for p_y vs y because of axial symmetry. Note that on this plot there are particles with relative high p_x around $x = 0$. This is because particles at the two ends have transverse temperature and are contained in areas with small radius. However, the normalized x emittance is constant along the beam, as shown in Fig. 2-76(g); this plot is essentially identical to the plot for y emittance vs z .

To observe whether there is emittance growth, it is necessary to use normalized emittance ϵ_n because particles are being decelerated along the beam due to

space-charge gradient. The normalized rms emittance in the x direction, measured on the coordinate of any slice along the beam, is¹²⁶

$$\bar{\epsilon}_n^x = \frac{1}{c} [\bar{x^2 \dot{x}^2} - (\bar{x \dot{x}})^2]^{1/2}, \quad (71)$$

where dots indicate time derivatives. Although CONDOR measures emittance on the coordinate moving with the beam center, the normalized emittance obtained from CONDOR for each slice of the beam essentially equals that given by Eq. (71) because the velocity difference between the beam head and beam tail is nonrelativistic.

Particle Simulations. We found that when the beam completes the self-compression and becomes 1 m long, no particles have been lost. The line-charge density and radius become uniform along z , as expected, as shown in Fig. 2-77(a) and (b). The slope of v_z vs z becomes flat [Fig. 2-77(c)] and the distribution of v_z is equivalent to having a T_L of 12 keV except at the two ends, as shown in Fig. 2-77(e). The final T_L is 2 keV higher than what we would have expected, but the $\Delta p_z/p_z$ is still well within the constraint of the final focusing lens. The transverse temperature along the beam also reduces, due to radial expansion, to the

Fig. 2-77. Beam parameters at the end of compression. (a) Line-charge density vs z . (b) Radial distribution of beam particles vs z . (c) Longitudinal velocity vs z . (d) Transverse temperature vs z . (e) Longitudinal temperature vs z . (f) Momentum in x direction vs x . (g) Normalized emittance in x direction vs z .

value at the beam center and becomes uniform except at the two ends, as shown in Fig. 2-77(d). Figure 2-77(f) shows that there are no longer particles scattered along the p_x axis around $x = 0$ as in Fig. 2-76(f). This is because particles near the two ends of the beam undergo rotation in p_x - x phase space caused by radial expansion during longitudinal compression. The $\bar{\epsilon}_n^x$ shown in Fig. 2-77(g) maintains approximately the same value, along most of the beam, at the end of the compression as its value at the start. The emittance growth, which occurs mainly at the two ends, is estimated to be less than 10%. The reason that there is some emittance growth at the two ends is probably because the resultant radial electric field there is no longer linear with radius because the beam is not space-charge dominated at the two ends. Nonlinear fields give rise to emittance growth.

The projected radial density distribution of the entire beam is uniform, as shown in Fig. 2-78. This, in addition to the fact that the radial electric field along the beam is almost zero except near the two ends, confirms the assumption—that the radial density profile is uniform—we made earlier.

Finally, we have carried out a simulation otherwise identical to the one discussed above but with initial $T_{\parallel} = 0$. Figure 2-79 shows that there is essentially no longitudinal emittance growth in this case. We also find that $T_{\parallel}$ remains essentially zero along the beam except near the two ends. Thus, there is no significant coupling between the longitudinal and transverse energies of any particle.

Conclusion. CONDOR particle simulations show that the beam compression confirms the predictions of simple 1-D theory provided that the initial beam parameters are calculated correctly for the simulation. The compressed beam satisfies the current and $\Delta p_x/p_x$ constraints of the final focusing requirements. Longitudinal and transverse emittance growths are very small and are well within the tolerable limits.

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Fig. 2-78. Projection of the radial density distribution along the entire beam.

Fig. 2-79. Longitudinal momentum vs z at the end of the compression with initial $T_{\parallel} = 0$.

Numerical Modeling with Internal Boundaries: The Spindt Cathode

Numerical modeling with boundary conditions that are specified within the simulation region is often an important analytical tool. An obvious example can be found in axisymmetric accelerators with some internal anode/cathode arrangement whose precise geometry is essential to properly model its electromagnetic field. For reduced models, such as the radiation-free Darwin model, solving elliptic equations with boundary conditions on internal surfaces is the dominant numerical task. Most computational models are less demanding but still require a general capability to represent internal boundaries.

In 1987 we investigated new ways to incorporate internal boundaries into

Fig. 2-80. Metal cone in Spindt cathode model: I is the silicon substrate, II is the insulating silicon-dioxide layer, and III is the molybdenum cone and the gate film.

elliptic problems associated with Maxwell's equations. We have succeeded with a system of mask arrays that trigger different equations to be solved on specified points within the mesh. The auxiliary equations can be chosen to represent arbitrary mixtures of Dirichlet or Neumann conditions at these internal points in a manner that still allows the algorithm to be vectorized. Loss of both vectorization and simplicity are the main disadvantages of older "brute force" techniques. Further, by treating the external boundaries of the mesh in the same fashion, we gain the added flexibility of applying boundary conditions whose form and type can be functions of position along that boundary.

A straightforward application of these ideas is our modeling of the Spindt cathode.¹²⁹ This cathode consists of an array of microscopic metal cones that function as electron sources. These cones, one of which is shown in Fig. 2-80, can be manufactured in regular arrays with microcircuit technology. We hope that the Spindt cathode will become the basis of a triode capable of switching high-power circuits without the usual ohmic losses associated with cathode heating. Our goal is to model aspects of this triode in order to predict its performance as both the electrical properties and the positions of its internal components are varied.

The Spindt cathode consists of a semiconductor substrate, the metal cone, an insulating layer, and a conducting gate layer. These regions are differentiated by specifying the spatial dependence of various electrical properties across the mesh. We are concerned with the spatial

dependence of the following arrays: the permittivity ϵ , the resistivity, the free-electron density given by the unneutralized background ion density n_i , and the values of the potential ϕ on the external walls and gate conductor.

We are interested in modeling two aspects of this cathode:

1. We can model the electron behavior in the vacuum region surrounding the metal cone and between the gate and anode if we know the potential everywhere in Fig. 2-80.

2. We can model the current streamlines as a function of the substrate's electrical properties if we know the potential contours inside the substrate.

The field model is electrostatic: the electrostatic field $\mathbf{E}$ is determined from

$$\nabla \cdot \epsilon \nabla \phi = -4\pi(n_i - n_e) \quad (72)$$

and

$$\mathbf{E} = -\nabla \phi. \quad (73)$$

These equations are discretized in space in a manner that is second order in $\Delta x, y$.

The most basic form of our internal boundary-condition capability is demonstrated by the specification of Dirichlet values for ϕ on the gate conductor. The general finite-difference form of Poisson's equation is given by the template shown in Fig. 2-81. In this template, i and j denote the matrix coefficient's relative location, which determines the spatial location of the finite-differenced ϕ it multiplies.

In this template, all S are mask arrays that signify boundary points, RHS is an array containing the right-hand side, and BV is an array containing the boundary value to be applied. These arrays are defined as follows:

$S_b = 1$ where a Dirichlet or Neumann condition applies and is 0 everywhere else,

$S_{N_x} = +1(-1)$ where a forward(backward) x -derivative applies and is 0 everywhere else,

$S_{N_y} = +1(-1)$ where a forward(backward) y -derivative applies and is 0 everywhere else,

$RHS = -4\pi(n_i - n_e)$, and

Fig. 2-81. Template matrix for the general finite-difference form of Poisson's equation.

$$\begin{aligned}
 & [1 - S_b(i,j)] \begin{pmatrix} 0 & \frac{\epsilon(i,j + \frac{1}{2})}{\Delta y^2} & 0 \\ \frac{\epsilon(i - \frac{1}{2},j)}{\Delta x^2} & -\frac{\epsilon(i - \frac{1}{2},j) + \epsilon(i + \frac{1}{2},j)}{\Delta x^2} & \frac{\epsilon(i + \frac{1}{2},j)}{\Delta x^2} \\ 0 & \frac{\epsilon(i,j - \frac{1}{2}) + \epsilon(i,j + \frac{1}{2})}{\Delta y^2} & 0 \end{pmatrix} \phi \\
 & + S_b(i,j) \begin{pmatrix} 0 & 0 & 0 \\ 0 & [1 - S_{N_x}^2(i,j)] & 0 \\ 0 & [1 - S_{N_y}^2(i,j)] & 0 \end{pmatrix} \phi \\
 & + \frac{S_{N_x}(i,j)}{2\Delta x} \begin{pmatrix} 0 & 0 & 0 \\ [1 - S_{N_x}(i,j)] \epsilon(i - \frac{1}{2},j) & -\epsilon(i - \frac{1}{2},j) - \epsilon(i + \frac{1}{2},j) & [1 + S_{N_x}(i,j)] \epsilon(i + \frac{1}{2},j) \\ 0 & 0 & 0 \end{pmatrix} \phi \\
 & + \frac{S_{N_y}(i,j)}{2\Delta y} \begin{pmatrix} 0 & [1 + S_{N_y}(i,j)] \epsilon(i,j + \frac{1}{2}) & 0 \\ 0 & -\epsilon(i,j - \frac{1}{2}) - \epsilon(i,j + \frac{1}{2}) & 0 \\ 0 & [1 - S_{N_y}(i,j)] \epsilon(i,j - \frac{1}{2}) & 0 \end{pmatrix} \phi \\
 & = [1 - S_b(i,j)] RHS(i,j) + S_b(i,j) BV
 \end{aligned}$$

BV = the desired value of ϕ (or the desired Neumann value).

This structure was built into an alternating-direction implicit (ADI) routine. It was then a simple matter to specify ϕ on the gate conductor. We set $S_b = 1$, $S_{N_x(y)} = 0$, and the desired value of ϕ in the BV array on those points that represent the gate. We automatically set ϕ to the desired value contained in BV during the Poisson solution with these ADI coefficient definitions: ϕ was set to 0 at $x = 0$ and to a finite value at $x = x_{\max}$; ϕ on the gate conductor was chosen to be greater than 0 but less than ϕ on the far

wall. The vectorized, dynamic ADI routine we used allows the use of a large value for ϵ (10^5) in the cone and the gate conductor. The large ϵ forced the value of ϕ in the cone to be essentially constant, thus producing a negligible electric field in this conducting region.

For the cathode configuration of Fig. 2-80, Fig. 2-82 shows the $t = 0$ potential contours and electric fields consistent with zero charge density. The potential, and subsequent solutions of Eqs. (72) and (73), taking into account emerging electronic charge, could be used to self-consistently advance an ensemble of

Fig. 2-82. Calculated (a) potential contours and (b) electric fields in the plasma region at $t = 0$.

Fig. 2-83. Calculated (a) potential contours and (b) electron-current streamlines in the substrate/cone region at $t = 0$. The lower cone is shorted out.

electrons using the PIC method. In such simulations, the electrons are introduced by field emission from the cathode, which can be controlled by the potential at the cathode's surface.¹³⁰

Another interesting aspect of this problem is the concern that, should an arc short out the cone/gate region, an excessive amount of current will flow through the substrate and melt it, thereby destroying the entire cathode. We considered this situation, in which Dirichlet conditions are applied on the gate conductor and on the tip of the shorted cone in addition to the usual potentials specified at $x = x_{\min}$ and $x_{\max}$. Ideally the potential at the tip has a value that is equal to the gate potential minus the voltage drop across the gap between the gate and the cone. If an arc forms, the gap voltage drop is lost, and the entire gate potential appears across the substrate and cone.

A system with two cones, one of which is shorted out, is shown in Fig. 2-83. We are investigating the changes that can be made by varying the substrate's electrical

properties in an effort to find cathode designs that are less susceptible to damage from arcs.

We are encouraged by the ease with which these boundary masks allow us to adjust the location and physical properties of internal boundaries. We are now considering extending our Spindt cathode work with PIC electron modeling in the plasma region. This extension may require including work functions and field emission from the cone's surface to achieve the necessary realism.

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Reduction of Artificial Emittance Growth in Particle-in-Cell Simulations of Beam Transport

Focusing a particle beam on a small target at a distance requires that the beam have low emittance. That is, its particles must occupy a small volume in phase space. Accurate modeling of

low-emittance beams requires that the self force, in a beam whose interior density is uniform, be proportional to radius r . We show that, for obvious and common methods of field calculation in r, z PIC codes, the radial force at small r is excessive by terms $\propto \Delta r^2$, where Δr is the mesh spacing. This error tends to expand the center of a beam. Then we show methods to cancel this excess.

We assume all quantities are uniform in z and consider only the differencing on the radial mesh $\{r_j\}$, with $r_0 = 0$. In some codes the radial electric force is interpolated from a field $\{E_j\}$ defined on the same mesh, in others from a field $\{E_{j+1/2}\}$ defined at interleaved positions. Either way, a linear force variation will be obtained with linear or higher order interpolation if $E_j \propto r_j$ or $E_{j+1/2} \propto r_{j+1/2}$. So these variations are what we aim to produce from the charge weighting and the field difference equations.

In a continuum with uniform charge density ρ , we have $rE = (1/2)\rho r^2$ (rationalized cgs units). Therefore we want to produce

$$E_{j+1/2} = \frac{1}{2} \rho r_{j+1/2}. \quad (74)$$

The code's field equation (Ref. 131, Ch. 14), when there is no variation in z or θ , is likely to be equivalent to the Gauss law result

$$r_{j+1/2} E_{j+1/2} - r_{j-1/2} E_{j-1/2} = Q'_j / 2\pi, \quad j > 0, \quad (75)$$

and

$$r_{1/2} E_{1/2} = Q'_0 / 2\pi, \quad (76)$$

so that we want

$$\begin{aligned} \frac{Q'_j}{2\pi\rho} &= \frac{1}{2}(r_{j+1/2}^2 - r_{j-1/2}^2), \\ \frac{Q'_0}{2\pi\rho} &= \frac{1}{2}r_{1/2}^2. \end{aligned} \quad (77)$$

In weighting particle charge to the mesh, the most common compromise between computational expense and smoothness of the representation is linear weighting,

That is, for a particle with charge q at radius r between r_j and r_{j+1} , charge $q(r_{j+1} - r)/\Delta r_{j+1/2}$ is assigned to Q_j and $q(r - r_j)/\Delta r_{j+1/2}$ to Q_{j+1} , in which $\Delta r_{j+1/2} \equiv r_{j+1} - r_j$.

But we find that the linear-weighted charge usually differs from Eqs. (75) and (76). For particle spacing in r that corresponds to a uniform density in three dimensions, we find

$$\begin{aligned} \frac{Q_j}{2\pi\rho} &= \left[\frac{1}{2} r_j r_{j+1} + \frac{1}{6} (\Delta r_{j+1/2})^2 \right] \\ &- \left[\frac{1}{2} r_{j-1} r_j + \frac{1}{6} (\Delta r_{j-1/2})^2 \right]. \end{aligned} \quad (78)$$

Using this in the Gauss equation we find

$$\begin{aligned} r_{j+1/2} E_{j+1/2} &= \frac{1}{2\pi} (Q_0 + \dots + Q_j) \\ &= \rho \left[\frac{1}{2} r_j r_{j+1} + \frac{1}{6} (\Delta r_{j+1/2})^2 \right], \end{aligned} \quad (79)$$

whereas we want the value given by Eq. (74). These are equal if we define

$$r_{j+1/2}^2 = r_j r_{j+1} + \frac{1}{3} (\Delta r_{j+1/2})^2, \quad (80)$$

which satisfies

$$r_j < r_{j+1/2} < r_{j+1}. \quad (81)$$

Interpolation from these positions to each particle produces the desired harmonic force variation. Because of other considerations, one often prefers to interpolate from the same positions as for the charge density (Ref. 131, Chs. 8 and 15). In this case we can interpolate from the $\{E_{j+1/2}\}$ to the $\{E_j\}$ and then interpolate from this to the particles.

In codes such as CONDOR that have uniform radial zoning, $\Delta r_{j+1/2} = \text{constant}$, and that interpolate the electric field from $\{E_{j+1/2}\}$, it is most efficient to locate $E_{j+1/2}$ at the evenly spaced radii $r_{j+1/2} = (1/2)(r_{j+1} + r_j)$. Equation (79) can then be written as

$$r_{j+1/2} E_{j+1/2} = \frac{1}{2} \rho \left[r_{j+1/2}^2 + \frac{1}{12} (\Delta r_{j+1/2})^2 \right]. \quad (82)$$

which is too large by the amount $(1/24)\rho(\Delta r_{j+1/2})^2$. This error is as large as 33% nearest the axis.

Two methods cancel this error. We can change the weighting of charge from particles to the mesh or alter the field solution on the mesh; the latter is more economical. To correct by altering the field solution, suppose we form E via Eqs. (75) and (76), in which the corrected mesh charge Q'_j is such that

$$\begin{aligned} r_{j+1/2} E_{j+1/2} &= \frac{1}{2\pi}(Q'_0 + \dots + Q'_j) \\ &= \frac{1}{2}\rho r_{j+1/2}^2, \end{aligned} \quad (83)$$

i.e., Q' satisfies Eqs. (74) through (76). Then we have

$$\begin{aligned} (Q_0 - Q'_0 + \dots + Q_j - Q'_j) \\ = \frac{\pi}{12}\rho(\Delta r_{j+1/2})^2. \end{aligned} \quad (84)$$

We can arrange this by moving a bit of charge outwards between neighboring $\{Q_j\}$ to get $\{Q'_j\}$, i.e.,

$$\begin{aligned} Q'_j &= Q_j - \delta Q_{j+1/2} + \delta Q_{j-1/2}, \\ Q'_0 &= Q_0 - \delta Q_{1/2}, \end{aligned} \quad (85)$$

in which

$$\delta Q_{j+1/2} = \frac{\pi}{12}\rho(\Delta r_{j+1/2})^2. \quad (86)$$

In a code we can only infer ρ in Eq. (86) from the $\{Q_j\}$. This is subject to statistical noise. We have successfully tried a diffusive implicit donor-cell advection obtained by setting

$$\pi\rho = \frac{Q'_j}{(\Delta r_j^2)} \equiv \frac{Q'_j}{(r_{j+1/2}^2 - r_{j-1/2}^2)} \quad (87)$$

in Eq. (86). Equations (85) and (86) are solved by a recursive outward sweep. The computational cost of this procedure is negligible.

When Cartesian coordinates are used to represent the transverse direction (x,y) this problem does not arise. The

property of linear forces in uniform beams is obtained in the usual PIC codes except at the beam edge. Inside a beam of elliptical x,y cross section and of uniform density, the usual particle-to-mesh weighting of charge yields equal and correct values of the mesh charge density ρ_{jk} . The analytic potential $\phi = -2\lambda(x^2/r_x + y^2/r_y)/(r_x + r_y)$ exactly satisfies the usual 5- and 9-point Poisson difference equations. (Here λ is the charge per unit length, and r_x, r_y are the axes of the ellipse.) The electric field evaluated by centered differences of the potential above is also exact, $E_x = 4\lambda x/[r_x(r_x + r_y)]$. Further, the interpolated fields provide the correct linear variation. Lastly, because rotation of this potential introduces only linear terms, these properties persist even when the ellipse axes do not lie along the mesh directions.

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Developments on the ARGUS Three-Dimensional Particle Code for Solving Ion Fusion Beam Transport and Compression Problems

We have studied final beam compression and transport in heavy-ion fusion accelerator drivers. Longitudinal beam compression and the possibility of concurrent pulse shaping have been the subject of previous studies using 1-D and 2-D codes (see, e.g., Ref. 132). The presence of magnetic quadrupole focusing fields is realistically a 3-D effect, however. We are making developments on the ARGUS PIC code as part of our effort to study 3-D effects of magnetic quadrupole beam transport. In particular, we are simulating final compression of a drifting heavy-ion beam pulse. Although magnetic quadrupoles are expected to be an integral part of much of the heavy-ion accelerator driver, restrictions due to space-charge modifications of the beam are particularly acute during final compression and focus (see, e.g., Ref. 132). We include an illustration of an initial benchmarking run.

ARGUS: A Three-Dimensional Hybrid

Simulation Model. ARGUS is a modular, fully three-dimensional, hybrid (particle-fluid) electromagnetic simulation model designed for studying a wide variety of plasma and beam-plasma systems. We are modifying a subset of ARGUS for ion-beam fusion applications. ARGUS is also being developed for modeling physical configurations that involve factors such as complicated geometrical structures, rate processes, radiation, and other processes acting on disparate time scales. Here we describe the overall structure of ARGUS but consider in detail only those modules or modifications specifically useful to our heavy-ion fusion problem.

ARGUS consists of a number of basic, "plug-compatible" modules that communicate through a common data structure. These modules may operate either in conjunction with one another or as stand-alone packages for specific problems. We use the following basic ARGUS modules:

1. Particle-in-Cell Module. This module evolves an arbitrary number of relativistic particle species in time and accumulates the source terms needed by the field-solving module. We are using the collisionless plasma approximation, even though the particle equations of motion can include both elastic and inelastic scattering by background species. Orbit integration can be performed in the usual single-step fashion for solving initial-value problems, or as a complete integration for performing iterative equilibrium calculations.

2. Field Module. This module can actually use a collection of packages, each of which is designed to solve a particular subset of Maxwell's equations and can function as a stand-alone code for multi-dimensional electromagnetics calculations. For our problem, we use predetermined external magnetic fields for beam focusing and self-consistent electrostatic fields computed by the Poisson-solver CYCLOP, which is described below.

3. Diagnostics Module. This module provides both on-line and postprocessing capabilities. For example, we use three-dimensional contour plots of fields and phase-space particle plots where appropriate.

4. System Interface Module. This module provides the necessary input/output, graphics, and memory-management support for the Cray X-MP.

5. Setup Module. This module provides for setup of complicated geometries, internal structures, etc.

External Quadrupole Fields. For our present simulations, a finite-length bunch of beam particles is followed in a rectangular pipe in the beam frame. The external linear quadrupole magnetic fields in the x and y directions are translated past the particles with the negative of the beam velocity in the z direction. Quadrupole magnetic fields are allowed to be of arbitrary length L_B with arbitrary field-free regions in between of length L_V , but the approximation of rapid cutoffs (sharp edges) is adopted as we leave the regions with fields.

CYCLOP: A Fast, Direct Solver of Poisson's Equation for Large Three-Dimensional Problems. We have developed CYCLOP, a fast electrostatic solver that contains a multiblock data structure, for the 3-D ARGUS simulation code. CYCLOP solves a 3-D Poisson's equation for interconnected multiregion problems with boundary structures embedded in the computational domain. Efficiency in dividing a problem into computational subregions (blocks) is an important feature of the solver because it permits attacking large problems on computers with limited core memory. This multiblock feature can also be useful for our future studies of matching straight and beam-bending sections. Solutions of Poisson's equation in these subdivided regions are calculated sequentially by switching data back and forth between central core and auxiliary disk storage. A second solve is necessary to "patch" these solutions together with a block connection algorithm in order to obtain a global solution satisfying global boundary conditions.

As an illustration we consider a problem in Cartesian coordinates with uniformly spaced grids in the x and y directions. This choice of grids allows us to Fourier analyze the problem in these two directions for a speedy computation of the Poisson solution. The grid in the z direction, however, is nonuniformly

spaced and can be subdivided into a number of blocks; therefore, Poisson's equation becomes

$$2 \frac{\left(\frac{\phi_{n+1}}{\Delta z_{n+1}} + \frac{\phi_{n-1}}{\Delta z_n} \right)}{(\Delta z_{n+1}) + \Delta z_n} + f_n \phi_n = S_n, \quad (88)$$

$$f_n = -2 \left\{ 2 \left[\frac{\sin^2 \left(\frac{\pi k}{N_x} \right)}{\Delta x^2} + \frac{\sin^2 \left(\frac{\pi l}{N_y} \right)}{\Delta y^2} \right] + (\Delta z_{n+1} - \Delta z_n)^{-1} \right\}, \quad (89)$$

For N -block problems, potentials adjacent to interfaces (for example, ϕ_M and ϕ_M^- neighboring the interface at z_M , as shown in Fig. 2-84) are linked by the three-point stencil given in Eq. (88). These potentials can be further expressed as linear superpositions of two solutions of the form

The first solution $\phi_{0,M}$ comes from the potential on cells adjacent to z_M by solving Poisson's equation in the associated block, including the source term S and subjecting the solution to zero-potential boundary conditions at block interfaces. The second solution (inside the parentheses) is a mapping that arises due to the nonzero potential on the neighboring interfaces. For instance, setting a unit potential on interface z_M and solving Poisson's equation in adjacent blocks without a source term leads to potentials z_M^+ and z_{M+1}^- that are the mapping quantities $\Phi_{M,M}^+$ and $\Phi_{M+1,M}^-$, respectively. Because these quantities do not depend on source terms, they need to be calculated only once. The particular solution within block M uses sources, but it adopts simple boundary conditions of vanishing potentials. To these must be added a proper solution of Laplace's equation (no sources), which involves finding the proper potential values at block interfaces subject to the global boundary conditions over all the blocks.

$$a_M \phi_{M-1} + b_M \phi_M + c_M \phi_{M+1} = d_M. \quad (91)$$

Here a , b , c , and d are functions of known quantities such as $\phi_{0,M}$, $\Phi_{M,M}$, S_M , and Δz_M , where M varies from 1 to $N - 1$.

This equation has the form of an $N - 1$ by $N - 1$ tridiagonal matrix equation and can be used to solve for all the interface potentials ϕ_M . The solution for the potentials in the entire domain is carried out block by block based on these known ϕ_M .

This electrostatic solver utilizes the "capacitance matrix" technique¹³³ to correct the potentials on the embedded structure points to specified values. Most quantities involved in the solver, such as the connection quantities $\Phi_{M,M'}$ need to be calculated only once during the setup stage and are stored for later use. The algorithm is for problems dealing with evolving sources and fixed geometry, which require repetitive evaluation of the Poisson potentials. Moreover, this algorithm does the field solve in a single sweep (with some overhead for block connection); that is, it does not employ an iterative method that might create a problem of numerical convergence.

Data Management. A key feature of ARGUS is that it can treat problems too large to fit entirely into core. Data for fields and sources are broken into "field blocks" representing specific spatial regions, which are simply rectangular solids for the Cartesian case. Since ARGUS has been a three-dimensional code from its inception, data are organized in this fashion rather than as two-dimensional slabs, an approach we find to be much more efficient. Nonuniform gridding is allowed in all three directions, and particle data are organized into blocks of fixed length called "segments." Field blocks and particle segments are then transferred between disk and core as needed, and all input/output is completely overlapped. This block/segment structure is well suited to a multiprocessing environment.

Fields. All field blocks are organized on a Cartesian "supergrid," which becomes a logical grid for non-Cartesian geometries. The placement of field blocks on the supergrid must allow for the connection of blocks that have been solved separately; therefore, field blocks overlap by one or two cells at block interfaces. The interface regions contain data from adjoining blocks, making block connection straightforward if the field and fluid algorithms are properly chosen.

For example, the CYCLOP fast Fourier transform (FFT) solver uses the capacitive matrix technique¹³³ to connect regions and to embed structures. It is unique because, to our knowledge, it is the only fast, direct solver that allows for distinct regions to be matched at interfaces.

Particles. The particle algorithms must allow for particles that may not be in

Fig. 2-85. Plots of space-charge potential contours in a series of x and y transverse cross sections. The beam at different z (same time) is elongated alternately in y and x directions because of phases in the quadrupole transport channel.

core simultaneously to cross between blocks. To accomplish this, particles are first pushed as described earlier. Those particles remaining in the current field block are written to an output buffer, and any particles moving to other field blocks are written to a dynamically managed cache. After all the particles have been advanced, this cache is sorted for the next time step and for attribution of the particles to the correct source points. The sorted particle data are simply appended at the appropriate place in the input stream on the next push.

Discussion. We have been benchmarking the code using parameters typical of a design under study at the Lawrence Berkeley Laboratory for an accelerator for testing ICF ion-beam physics (acceleration, transport, final compression, bending, and focusing). This so-called ILSA accelerator design envisages about 10 A of ~ 10 -MeV C^+ ions with a velocity tilt (head-to-tail velocity difference) of 5 to 12%. Figure 2-85 illustrates typical plots of space-charge potentials in the transverse (x, y) plane, showing the beam at different z elongated alternately in the y and x directions because of the quadrupoles. Although our test runs use coarse grids and small particle numbers to fit within our daily computer-time allotment, these runs already illustrate some of the fundamental 3-D features of quadrupole focusing in a machine comparable in length to that at LBL and to near-term designs for the next stage. Typically, the particles reach their outermost transverse position where the quadrupole focusing is strongest and reach minimum where the de-focusing dominates.

Our major purpose in this article has been to describe the basic ARGUS modules and algorithms for evolving particles and fields and for handling concurrent data and memory-management needs in the heavy-ion fusion beam compression problem. Although preliminary results are encouraging, we expect to incorporate additional features into this code before obtaining definitive results.

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Surface Waves on Heavy-Ion Beams

Theoretical calculations by Smith¹³⁴ using cold fluid equations show that a perturbation on the longitudinal particle velocity $v_z(z)$ will result in surface charges (to be referred to here as surface waves) moving away from the point of disturbance. These surface waves are moved along the beam by a combination of axial and radial electric fields with zero divergence and, correspondingly, zero internal charge density. What is left behind at the point of initial disturbance, therefore, is an envelope oscillation with no net change in line-charge density λ . This article reports on the use of CONDOR PIC simulations to check the theory, to examine the effect of temperature, which is not considered in the theory, and to determine whether any emittance growth results from surface waves.

We assume initial conditions $\rho_i = 0$ (where ρ_i is the perturbed density; the subscript i refers to initial conditions) and $\mathbf{v}_i = v_{zi}(z) \hat{\mathbf{e}}_z$, where $\hat{\mathbf{e}}_z$ is a unit vector in the z direction, corresponding to an error in the accelerating wave form. Then the solution in the long wavelength limit¹³⁴ (in which the axial extent of the perturbation is large compared to the beam radius) for a beam of radius a surrounded by a perfectly conducting cylinder of radius b is

$$\rho = -\frac{1}{\omega_p} \frac{\partial u_i}{\partial z} \sin \omega_p t, \quad (92)$$

$$\sigma = \frac{a}{2\omega_p} \frac{\partial u_i(z)}{\partial z} \sin \omega_p t + \frac{a}{4v_p} [u_i(z - v_p t) - u_i(z + v_p t)], \quad (93)$$

$$v_z = \frac{1}{2} [u_i(z - v_p t) + u_i(z + v_p t)], \quad (94)$$

$$v_r = \frac{r}{2} \frac{\partial}{\partial z} \left\{ u_i(z) \cos \omega_p t - \frac{1}{2} [u_i(z - v_p t) + u_i(z + v_p t)] \right\}, \quad (95)$$

where σ is the surface charge, v_r is the radial velocity, $v_p^2 = (\omega_p^2 a^2 / 4) \ln(b^2 / a^2)$

gives the wave speed, and ω_p is the plasma frequency.

The uniform beam used in the simulation is 5 m long, has a radius of 0.03 m, is in force equilibrium with a linear radial electric field, and is surrounded by a perfectly conducting cylinder with a radius of 0.06 m. Periodic boundary conditions are imposed at the two ends of the beam.

The beam has a uniform initial line-charge density λ of 1.26×10^{-8} C/m; correspondingly, $v_p = 5 \times 10^5$ m/s. The transverse temperature is 1900 eV, which corresponds to a "tune depression" σ/σ_0 of 0.1, where $\sigma(\sigma_0)$ is the single-particle phase advance per quadrupole period with (without) space charge. The rms emittance in the x and y directions of 1.25×10^3 m²/s is uniform along the beam. Note that the CONDOR PIC code measures emittance using

$$\epsilon_{rms}^x = \left[\dot{x}^2 \dot{x}^2 - (\dot{x}\dot{x})^2 \right]^{1/2}, \quad (96)$$

where dotted values mean derivative with respect to time.

To observe whether the perturbation affects only regions near the beam surface, we plot λ near the beam center (from beam center to 83% of the beam radius) and λ near the beam edge (covering the outer 17% of the beam radius) vs the longitudinal coordinate z . Figure 2-86(a) shows that the initial imposed

longitudinal velocity perturbation has a Gaussian shape with peak value $v_{z1}(0) = 2 \times 10^5$ m/s. At half of maximum, this perturbation has a full width of about 0.2 m and thus satisfies the long wavelength limit. The initial disturbance takes place only in the longitudinal direction, with no disturbance in line-charge density or particle position.

After the simulation has advanced for 3.3×10^{-6} s, we observe that perturbation in v_z essentially causes no perturbation in λ for the inner part of the beam, as shown in Fig. 2-86(d). But, as expected from the theory, the charge density near the surface is strongly perturbed [see Fig. 2-86(e)]. The total λ is perturbed [see Fig. 2-86(c)], but λ remains essentially constant at the original point of disturbance. We calculate the velocities for the left- and right-going waves and find the values to be within about 5% of those given by Eq. (94) [see Fig. 2-86(f)]. As shown in Fig. 2-86(g), no significant emittance growth is caused by the surface waves.

We repeated the calculations assuming a longitudinal temperature of 2500 eV (roughly equal to the transverse temperature). No significant differences from the above simulation result, and the speed of propagation of the surface waves is essentially the same as before, as shown in Fig. 2-87.

Fig. 2-86. Results for a beam with zero initial longitudinal temperature. (a) Initial longitudinal velocity perturbation vs z . (b) Particle distribution vs z . (c) Total line-charge density vs z . (d) Line-charge density from beam center to 83% of the beam radius vs z . (e) Line-charge density from 83% of the beam radius to beam edge vs z . (f) Longitudinal velocity perturbation vs z . (g) Emittance in x direction vs z .

Fig. 2-87. Longitudinal velocity vs z with 2500-eV longitudinal temperature.

Particle simulation shows the analytical theory to be correct. The inclusion of temperature does not qualitatively alter results derived from Smith's computations, and no significant emittance growth is observed.

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Modifications of the 2-1/2-Dimensional CONDOR Code for Heavy-Ion Fusion

We briefly outline a continuing collaborative effort to make various modest modifications of the 2-1/2-D particle code CONDOR for the purposes of solving a number of particle beam physics problems, as part of a program to develop HIF. This effort has been pursued at a low level for several years and the code was used for our HIF beam compression studies.^{135,136} The ion pulse emerging from a heavy-ion reactor driver will have the energy required to drive fusion targets, but

it will require longitudinal compression to give it enough power. We are using this code to study techniques for power multiplication and pulse shaping as part of the U.S. induction accelerator development effort for HIF.

CONDOR is a modular, 2-1/2-D electromagnetic PIC code where the particles are followed in 3-D but the fields only in 2-D. CONDOR was specifically designed for combining basic plasma simulations with complex geometries and internal structures applicable to particle beams in various accelerating structures and ion sources. It also provides capabilities for incorporating various detailed particle-particle, particle-wall, and other physics or electrical engineering effects (such as particle scattering, stopping, injection, and absorption on surfaces, as well as external fields, coils, circuits, and loads). We expect increased use of this code for HIF energy research, initially in its electrostatic version because the beams are not relativistic.

Here we briefly mention the modifications of this code for our applications. Increased funding from energy research allows some improvement in the level of development effort, particularly for the 3-D counterpart ARGUS outlined in the earlier article "Developments on the ARGUS Three-Dimensional Particle Code for Solving Ion Fusion Beam Transport and Compression Problems." Unlike ARGUS, CONDOR is a code where we have made some modest improvements (listed below) to make it useful for ion beam fusion. We are pleased, however, to point out that CONDOR is also an example of a collaborative endeavor that we believe provides a cost-efficient benefit to energy research.

Specific Improvements in CONDOR.

The code capabilities were actually improved for electrostatic problems, specifically in the following areas:

- Replacement of the Poisson correction step of the original electromagnetic code with a Poisson solver of structure similar to that described in the preceding article on ARGUS (except for the multi-block formalism). The original method of running CONDOR in the electrostatic

mode consisted of disabling the field propagator and keeping only the Poisson correction step. When the ion beam approached the ends of the simulation region, the Poisson correction became unstable. A Poisson solver is now used for the electrostatic simulations; this solver benefits ion fusion and other users. The direct solve avoids stability problems encountered by the Poisson correction.

• The development of boundary conditions appropriate to an isolated beam bunch. The boundary conditions appropriate for a finite-length beam bunch in an essentially infinite-length conducting pipe are a good first approximation to the effects of a beam propagating and compressing in length near the end of an accelerator. In the empty region outside of the beam bunch, the electrostatic potential of beam in pipe is

$$V = \sum_{n=1}^{\infty} C_n J_n(k_n r) \exp(-k_n z), \quad (97)$$

which decays rapidly and exponentially from the region of the beam pulse. Here (r, θ, z) is a cylindrical coordinate system with the z axis centered on the pipe's axis and r, θ the radius and azimuthal angles. The constants k_n are related to the n th zero of the Bessel function, $J_n(k_n b) = 0$, where b is the pipe's radius. Because of this exponential decay of V with z , it is sufficient in numerical simulations to set the potential V to be periodic between the $z = 0$ and the $z = 2L$ positions in a Fourier transform of the potential, where L is beam pulse length. This is because the constant $k_{\min} \sim b^{-1}$ or a^{-1} , where a is beam radius and beam length $L \gg b \gg a$. This method can also be used to get an approximate free space boundary condition.

• A new data base management system to allow optimization of memory utilization for electrostatic problems. CONDOR is used most often as an electromagnetic simulation code. Mesh-wide arrays are stored separately for the electric and magnetic fields in each direction. For electrostatic simulations, logic was added to the memory manager to prevent the

allocation of any unused mesh-wide arrays. This resulted in reducing the size of the CONDOR controllee by about a factor of 4 for electrostatic problems.

• Modifications of initial particle distribution. The ion beam is placed into the simulation through the initial particle distribution coding in CONDOR. Thus the initial loading of the beam must match the beam's equilibrium at that point. Earlier, simple analytical formulas were added to the initial particle distribution code to create the equilibrium state. It soon became apparent that more flexibility was needed to control the initial beam loading. Simple 1-D tables are now available for use to control density, temperature, momentum, and beam radius as a function of z . Variations in R can also be controlled by means of the table input. These tables have proven to be easy to implement and adequate to specify the initial equilibrium. Even greater flexibility is being implemented. The user is given the flexibility to write a small input Fortran code to specify the densities, temperatures, and local drift velocities of the initial particle loading. These numbers can be specified on physical grids that could be different from those used by the main simulation. Thus, for example, a linear function of coordinates requires only a specification of values at the four corners of a rectangle. A skeletal code is provided so that the user need only give coding for placing numbers on these input grids. Additional coding is provided for plotting the input and for writing it into CONDOR-generator format and/or NACF format wherever generator line input becomes prohibitive in size.

• Root-mean-square particle diagnostics. Two different uses (snapshots or time history) of the same form of rms beam emittance diagnostics were added to the code. The beam emittance of selected regions as a function of time were added to the standard time history data. Snapshots of beam emittance and other rms quantities as a function of z were also added. Emittance is a measure of beam temperature, which for the transverse case measures area (divided by π) in the phase space made up of particle

position and angle of velocity vector. It gives the typical product of transverse position and velocity vector angle of the particles. The total rms transverse emittance of a particle is defined as

$$\epsilon_{rms} = \left(\langle (\gamma^2 - 1)^{1/2} \right) \left[\langle x^2 + y^2 \rangle \langle x'^2 + y'^2 \rangle - \left(\frac{d}{2dt} \langle x^2 + y^2 \rangle \right)^2 - \langle xy' - yx' \rangle^2 \right]^{1/2}. \quad (98)$$

Here (x, y) are Cartesian position coordinates transverse to the beam axis and $x' = dx/dz = (dx/dt)/v_z$, $y' = dy/dz = (dy/dt)/v_z$. This particular form of rms emittance chosen corresponds to one-third the total transverse edge emittance in a uniformly filled elliptical region in phase space. Of course the emittances differ for general distributions.

• The θ component of the particle positions. CONDOR is a 2-1/2-D code. For example, in cylindrical (r, θ, z) coordinates about beam axis, the fields cannot have variations in the θ direction (i.e., the problem must be axially symmetric) but the particles can be followed in 3-D. For x - y and r - θ geometries, the particle coordinates were already followed in 3-D. In r - z (the geometry most often used for CONDOR simulations of ion beam problems), the particles were originally given a θ position of 0. Even though the particles could develop a θ velocity, the θ position remained 0—i.e., for axially symmetric calculations the particles' θ positions are ignorable. To allow x - y projections of the particle quantities for diagnostics such as the beam emittance, code was added to allow the randomization of the particle's θ component. This θ position can be either constant throughout the simulation or updated at each time step.

• Particle scatter plots. Snapshot particle plots with x - y projections of the r - z particle data were added to the

standard CONDOR phase space plots. Additionally, particle data actually used to calculate emittance can be dumped and v_x vs x plots made in the time history postprocessor.

• Volume weighting. Simulations of problems with r - z (axis included) geometry have grid cells whose volume varies as a function of r . If important quantities, such as the ion beam, come within about five grid cells of the axis, the standard area weighting scheme used in CONDOR becomes inaccurate. An alternative volume-weighted particle interpolation and field-smoothing algorithm is available that prevents the simulation errors from distorting the physics of the ion beam.

Summary. CONDOR is a 2-1/2-D plasma PIC code that has been modified gradually over the past few years for application also to HIF energy research.

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LASNEX and Atomic Physics

Introduction

During the past year the LASNEX group made two major improvements in its atomic physics modeling.

Our earlier work on the Compton scattering kernel was extended to the calculation of various coefficients that are used in the Fokker-Planck and diffusion approximations for Compton scattering. We obtained, for the first time, simple, closed form, fully relativistic analytic expressions for these coefficients, and these new, improved values have been put into LASNEX. The next article, "Integrals of the Compton Scattering Cross Section," describes our new method for calculating these coefficients. The article "Rapid Calculation of the Energies of

Plasma Atoms and Ions" describes an analytic method for obtaining the Taylor series coefficients.

Mixing of materials due to large amplitude hydrodynamic instability growth is important in many ICF design problems. Exact modeling of mixing has proved enormously complicated, and for a design code like LASNEX it is very valuable to have a simple parameterized model that satisfies the basic conservation laws and can fit the results of many more complicated models by an appropriate choice of the parameters.

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Integrals of the Compton Scattering Cross Section

In the past year we evaluated various integrals (moments) of the Compton scattering kernel (CSK). These integrals represent physical quantities associated with Compton scattering. In this article we give the total Compton cross section, the Fokker-Planck diffusion coefficient, and the Compton transport mean free path in terms of single integrals valid for arbitrary values of the electron temperature and photon frequency. The above integrals are coefficients of the radiation diffusion equation, which is one of the most commonly used means of approximating radiation transport in LASNEX. Here we present a brief outline of the results. Details are given in Refs. 137 and 138.

Neglecting emission and absorption, the radiation diffusion equation, in the (normalized) units used in Refs. 137 and 138, is

$$\begin{aligned} & \gamma^2 \frac{\partial n}{\partial t} - \gamma^2 \nabla \cdot \left(\frac{\lambda_{tr}}{3} \nabla n \right) \\ &= \int_0^\infty d\gamma' [S(\gamma' \rightarrow \gamma, \tau) |1 + n(\gamma)| n(\gamma') \\ &- S(\gamma \rightarrow \gamma', \tau) |1 + n(\gamma')| n(\gamma)], \end{aligned} \quad (99)$$

The nondimensional photon energy γ and electron temperature τ are in units of the electron rest mass energy. The photon distribution function n is normalized to the occupation number of the photon gas, and t is the normalized time variable ($t = \sigma_T N_e c t$, where σ_T is the classical Thomson cross section, N_e is the electron density, c is the speed of light, and t is the time). In Eq. (99), the spatial operator ∇ has been normalized to the inverse of the Thomson length, $N_e \sigma_T$. The dependence of n on the spatial variable $\mathbf{x}$ and the temporal variable t has been suppressed in Eq. (99); that is, $n = n(\gamma, \mathbf{x}, t)$. The spectral energy density may be identified with $\gamma^3 n$. The factor $[1 + n(\gamma)]$ describes induced scattering. $S(\gamma \rightarrow \gamma', \tau)/\gamma^2$ denotes the zeroth moment (over final photon direction Ω') of the CSK.

Consider a general integral of the CSK of the form

$$\begin{aligned} S_{m,n} &\equiv \int_0^\infty d\gamma' \left(\frac{\gamma'}{\gamma} \right)^m \\ &\times \int_{4\pi} d\Omega' \xi^n \sigma_s(\gamma \rightarrow \gamma', \xi, \tau), \end{aligned} \quad (100a)$$

$$\sigma_s(\gamma \rightarrow \gamma', \xi, \tau)$$

$$\equiv \frac{3}{16\pi} \frac{1}{\gamma^2} \int d\mathbf{v} \frac{f(v)}{\lambda}$$

$$\times \left\{ 1 + \left[1 - \frac{1 - \xi}{\lambda^2 DD'} \right]^2 \right.$$

$$\left. + \frac{(1 - \xi)^2 \gamma \gamma'}{\lambda^2 DD'} \right\}$$

$$\times \delta \left(\xi - 1 + \lambda \frac{D}{\gamma'} - \lambda \frac{D'}{\gamma} \right), \quad (100b)$$

where

$$f(v) = \frac{\lambda^5 e^{-\lambda/\tau}}{4\pi \tau c^3 K_2(1/\tau)} \quad (100c)$$

is the relativistic Maxwellian distribution at electron temperature τ , K_2 is the modified Bessel function of the second kind, and

$$D \equiv 1 - \mathbf{\Omega} \cdot \mathbf{v}/c, \quad D' \equiv 1 - \mathbf{\Omega}' \cdot \mathbf{v}/c, \\ \lambda \equiv 1/\sqrt{1 - v^2/c^2}, \quad \xi \equiv \mathbf{\Omega} \cdot \mathbf{\Omega}'. \quad (101)$$

Using the above definitions, we have

$$S_{0,0} = \int_0^\infty d\gamma' \frac{S(\gamma \rightarrow \gamma', \tau)}{\gamma'^2}. \quad (102)$$

We refer to Eq. (99) as the Boltzmann diffusion equation. The right-hand side of Eq. (99), the Compton scattering operator, conserves the number of photons, $\gamma^2 n$, but its unwieldy integral form has led previous authors to approximate that operator by a diffusion expression in frequency space, the Fokker-Planck approximation.¹³⁹⁻¹⁴⁴ Momentarily neglecting spatial transport in Eq. (99), the detailed-balancing property of the interaction kernel,¹⁴²

$$S(\gamma \rightarrow \gamma', \tau) e^{\gamma'/\tau} = S(\gamma' \rightarrow \gamma, \tau) e^{\gamma/\tau}, \quad (103)$$

implies that if induced scattering is neglected, then the Wien distribution $n = C e^{-\gamma/\tau}$ ($C = \text{const}$) is the equilibrium solution. If induced scattering is included, then a Bose-Einstein distribution $n = (C e^{\gamma/\tau} - 1)^{-1}$ results. Requiring that the Fokker-Planck equation satisfy these criteria leads to a unique candidate:

$$\gamma^2 \frac{\partial n}{\partial t} - \gamma^2 \nabla \cdot \left(\frac{\lambda_{tr}}{3} \nabla n \right) \\ = \frac{\partial}{\partial \gamma} \left\{ \alpha(\gamma, \tau) \left[\tau \frac{\partial n}{\partial \gamma} + n(1 + n) \right] \right\}. \quad (104)$$

To fix the Compton Fokker-Planck diffusion coefficient $\alpha(\gamma, \tau)$, we require that Eq. (104) give the same rate of change of the total energy as the Boltzmann equation when induced scattering is

neglected. This requires¹³⁷ that α be a solution of the differential equation

$$\alpha - \tau \frac{\partial \alpha}{\partial \gamma} = \gamma^3 (S_{0,0} - S_{1,0}). \quad (105)$$

Since $\gamma^3 n$ is the spectral energy density, if we consider the total energy change of the radiation field due to the Compton process we have

$$\frac{dE}{dt} = \int_0^\infty d\gamma \gamma^3 n(\gamma, t) (S_{1,0} - S_{0,0}) \\ \equiv \int_0^\infty d\gamma \gamma^3 n(\gamma, t) R(\gamma, \tau), \quad (106)$$

where we define $R(\gamma, \tau)$ as the Compton rate of energy transfer.

Integrating the CSK over all γ' and $\mathbf{\Omega}'$ gives $S_{0,0}$, the inverse of the Compton mean free path λ_0 (the average distance a photon travels before scattering):

$$\frac{1}{\lambda_0} \equiv S_{0,0}. \quad (107)$$

Multiplying $S_{0,0}$ by $\sigma_T N_e$ puts the result into the required length units.

In the diffusion approximation, the specific intensity is expressed as a truncated series in surface harmonics:

$$I(\gamma, \mathbf{\Omega}, \mathbf{x}, t) = \frac{1}{4\pi} I_0(\gamma, \mathbf{x}, t) + \frac{3}{4\pi} \mathbf{\Omega} \cdot \mathbf{I}_1(\gamma, \mathbf{x}, t). \quad (108)$$

We may identify I_0 with $\gamma^3 n$. Substituting this *ansatz* into the transfer equation and integrating over all $\mathbf{\Omega}$ gives the zeroth moment, an equation for I_0 . If stimulated scattering is neglected, $\mathbf{I}_1$ enters the zeroth equation only through the transport term, since

$$\int_{4\pi} d\mathbf{\Omega} \mathbf{\Omega} \cdot \nabla I(\gamma, \mathbf{\Omega}, \mathbf{x}, t) = \nabla \cdot \mathbf{I}_1(\gamma, \mathbf{x}, t). \quad (109)$$

Removal of the offensive $\mathbf{I}_1$ term in the zeroth equation requires deriving a Fick's law that expresses $\mathbf{I}_1$ in terms of a gradient of I_0 . The results of Ref. 138 show that

$$\frac{1}{3} \nabla I_0(\gamma, \mathbf{x}, t) = - \frac{N_e \sigma_T}{\lambda_{tr}(\gamma, \tau)} \mathbf{I}_1(\gamma, \mathbf{x}, t) \quad (110)$$

is the desired Fick's law. This expression involves the Compton transport mean free path λ_{tr} , which is given in terms of the integrals

$$\lambda_{tr}^{-1} \equiv S_{0,0} - S_{1,1}. \quad (111)$$

In Ref. 138, λ_{tr} is defined by integrating the first moment equation over all γ (the first moment equation is the transfer equation multiplied by Ω and integrated over all Ω). Cooper¹⁴⁵ derived Eq. (111) by a different argument but presented only an approximation to λ_{tr} which he obtained by fitting rational expressions to the tabulated results of Stone and Nelson.¹⁴⁶ Our contribution is that we produce the various $S_{m,n}$ integrals in closed form.

The required expressions for λ_0 , α , λ_{tr} , and R are derived in Refs. 137 and 138:

$$\begin{aligned} \frac{1}{\lambda_0} &= \frac{1/\gamma}{2K_2(1/\tau)} \\ &\times \int_0^\infty dz \gamma z \sigma_0(\gamma z) \exp\left[-\frac{1}{2\tau}\left(z + \frac{1}{z}\right)\right], \end{aligned} \quad (112a)$$

where

$$\begin{aligned} \gamma \sigma_0(\gamma) &= \frac{3}{8\gamma} \left[\frac{\gamma^2 - 2\gamma - 2}{\gamma} \ln(2\gamma + 1) \right. \\ &\quad \left. + \frac{2\gamma^3 + 18\gamma^2 + 16\gamma + 4}{(2\gamma + 1)^2} \right]; \end{aligned} \quad (112b)$$

$$\begin{aligned} \alpha(\gamma, \tau) &= \frac{\gamma^4}{2K_2(1/\tau)} \\ &\times \int_0^\infty dz z^2 \alpha_0(\gamma z) \exp\left[-\frac{1}{2\tau}\left(z + \frac{1}{z}\right)\right], \end{aligned} \quad (113a)$$

where

$$\begin{aligned} \alpha_0(\gamma) &= \frac{3}{8\gamma^4} (\gamma - 3)(\gamma + 1) \ln(2\gamma + 1) \\ &\quad - \frac{10\gamma^4 - 51\gamma^3 - 93\gamma^2 - 51\gamma - 9}{4\gamma^3(2\gamma + 1)^3}; \end{aligned} \quad (113b)$$

$$\begin{aligned} \frac{1}{\lambda_{tr}} &= \frac{1/\gamma}{2K_2(1/\tau)} \int_0^\infty dz \left[\frac{1}{2} + (\gamma - \tau)z + \frac{z^2}{2} \right] \\ &\times \gamma z \alpha_n(\gamma z) \exp\left[-\frac{1}{2\tau}\left(z + \frac{1}{z}\right)\right]; \end{aligned} \quad (114)$$

$$\begin{aligned} R &= \frac{1/\gamma}{2K_2(1/\tau)} \int_0^\infty dz \left[\frac{z^2}{2} + (\tau - \gamma)z - \frac{1}{2} \right] \\ &\times \gamma z \alpha_0(\gamma z) \exp\left[-\frac{1}{2\tau}\left(z + \frac{1}{z}\right)\right]. \end{aligned} \quad (115)$$

Despite appearances, $\sigma_0(\gamma)$ and $\alpha_0(\gamma)$ are analytic and equal unity at $\gamma = 0$ (see Ref. 138).

Equations (105), (106), (107), and (111) show that λ_0 , α , λ_{tr} , and R are defined in terms of appropriate $S_{m,n}$ integrals, defined in Eqs. (100). In Ref. 138, we show how to reduce such integrals to the form

$$\begin{aligned} S &= \frac{a\gamma^l}{2K_2(1/\tau)} \\ &\times \int_0^\infty dz \exp\left[-\frac{1}{2\tau}\left(z + \frac{1}{z}\right)\right] G(\gamma z), \end{aligned} \quad (116a)$$

$$a = \text{const and } G(y) = \sum_{j=0}^\infty g_j y^j. \quad (116b)$$

We are also able to manipulate expressions such as Eq. (116a) to obtain useful forms in the limits of small or large λ and τ . We derive asymptotic series for low electron temperature and arbitrary frequency. For example,

$$1/\lambda_0 (\tau \ll 1) \approx \sigma_0(\gamma) + \tau \sigma_1(\gamma), \quad (117)$$

where

$$\begin{aligned} \sigma_1(\gamma) &= -\frac{3}{8} \left[\frac{3\gamma^2 - 8\gamma - 6}{2\gamma^3} \ln(2\gamma + 1) \right. \\ &\quad - \frac{1}{\gamma^2(2\gamma + 1)^4} [4\gamma^5 - 124\gamma^4 - 231\gamma^3 \\ &\quad \left. - 157\gamma^2 - 50\gamma - 6] \right]. \end{aligned} \quad (118)$$

Such series expressions are useful since in our units $\tau = 1$ translates to an electron temperature of 511 keV.

For application in LASNEX, integrals such as Eq. (116a) are computed by Gauss-Hermite quadrature. Changing variables in Eq. (116a), we have

$$u = \frac{1}{\sqrt{2}\tau} \left(\sqrt{z} - \frac{1}{\sqrt{z}} \right),$$

$$0 \leq z < \infty, -\infty < u < \infty; \quad (119)$$

then

$$S = \frac{\alpha \gamma^{\frac{1}{2}} \sqrt{2}\tau}{e^{\frac{1}{\tau}} K_2(1/\tau)} \int_{-\infty}^{\infty} du e^{-u^2} \frac{z^{3/2}}{z+1} G(\gamma z). \quad (120)$$

For electron temperatures less than the electron rest mass energy, the integrals are evaluated by eight-point Gaussian quadrature, which gives sufficient accuracy in most applications. In computing λ_{tr} over the range $0 < \gamma < 100$, $0 < \tau < 1$, eight-point quadrature has a worst relative error of 3.59×10^{-4} at $\gamma = 0.1$ and $\tau = 1$. A vectorized Fortran subroutine computes λ_{tr} in a time (in seconds) given approximately by $5.8 \times 10^{-5} + 6.4 \times 10^{-6} N_r$, where N_r is the number of temperature values considered.

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Rapid Calculation of the Energies of Plasma Atoms and Ions

Calculations of the behavior of plasmas depend, in part, on accurate atomic data for the many ions in various states of excitation that are present. In the past, such data have been obtained from a rather crude, screened hydrogenic model (XSN); currently a much improved model based on relativistic, self-consistent field calculations is being developed.¹⁴⁷

In our new model, atomic energies are expressed in terms of a Taylor series

$$E(n) = E^0 + \sum_i (n_i - n_i^0) E_i^0 + \frac{1}{2} \sum_i \sum_j (n_i - n_i^0)(n_j - n_j^0) E_{ij}^0 + \dots, \quad (121)$$

where n_i is the number of electrons in the i th orbit, and the expansion is about a reference configuration n_i^0 . The term E^0 is the energy of that reference configuration, and $E_i^0 = \partial E(n^0)/\partial n_i^0$ turns out to be an eigenvalue in the self-consistent field calculation of the reference configuration.

The coefficients of the quadratic term in the Taylor series

$$E_{ij}^0 = \frac{\partial^2 E(n^0)}{\partial n_i^0 \partial n_j^0} = \frac{\partial E_i^0}{\partial n_j^0} = \frac{\partial E_j^0}{\partial n_i^0} \quad (122)$$

have been computed up to now by finite differences. A series of self-consistent field calculations is done in which, one-by-one, the reference configuration occupations are altered slightly, $n_i^0 \rightarrow n_i^0 + \Delta n_i$, and then $\partial E_j^0/\partial n_i^0$ is approximated by

$$\frac{E_j^0(n_i^0 + \Delta n_i) - E_j^0(n_i^0)}{\Delta n_i}. \quad (123)$$

This approximation has worked well enough, but an alternative method is now available that we can briefly describe.

What we propose is to use perturbation theory to compute the change in the eigenvalues when one of the occupation numbers is altered. Thus if n_i is changed to $n_i + \Delta n_i$, the charge density is changed to

$$\rho^0(r) + \Delta \rho_i^a(r) = \rho^0(r) + \Delta n_i |\psi_i(r)|^2, \quad (124)$$

where $\psi_i(r)$ is the i th orbital function, and the potential function is changed to

$$v^0(r) + \Delta v_i^a(r) = v^0(r) + \left[\int \frac{\Delta \rho_i^a(r')}{|r - r'|} dr' + \frac{\partial v_{xc}^0}{\partial \rho^0} \Delta \rho_i^a(r) \right], \quad (125)$$

where v_{xc} is the exchange-correlation potential. First-order perturbation theory then gives

$$\Delta E_j = \int \psi_j^*(r) \Delta v_i^a(r) \psi_j(r) dr, \quad (126)$$

where $j = 1, 2, \dots$

However, Eq. (126) is not quite right. When n_i is changed, the self-consistent field is altered, and the orbital functions relax; there is an implicit variation in the charge density,

$$\Delta\rho_i^b = \sum_k n_k [\psi_k^*(r) \Delta\psi_k(r) + \Delta\psi_k^*(r) \psi_k(r)], \quad (127)$$

and a corresponding change in the potential function $\Delta v_i^b(r)$. The expression for ΔE_j should have $\Delta v_i^a + \Delta v_i^b$ in it.

The calculation of $\Delta\psi_k(r)$ by perturbation theory is itself fairly complicated. Fortunately a computer program already exists,^{148,149} which does this calculation

for the case where $\Delta v^a(r) = r \cos \theta \cos \omega t$. This form for Δv^a is used for photoabsorption calculations. We have replaced $r \cos \theta$ by $\Delta v_i^a(r)$ given above and taken the frequency ω to be very small (a problem exists with singularities in the Green functions when $\omega = 0$). It should be noted that the equations for $\Delta\psi_k(r)$ use the combination $\Delta v_i^a + \Delta v_i^b$; both external and induced perturbing potentials are involved.

The results appear to be more accurate than those obtained by our original scheme; the partial derivative $\partial E_j^0 / \partial n_i^0$ is found to be more nearly equal to $\partial E_j^0 / \partial n_i^0$. In addition, we have observed that the contributions of Δv_i^b are significant and should not be neglected.

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Nova Experiments

3

Section Editor: E. M. Campbell

Nova Experiments

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Nova Experiments

Activities of the Nova Experiments Program

R. L. Kauffman and E. M. Campbell

Our activities are directed toward the experimental demonstration of the feasibility of inertial confinement fusion (ICF). The principal experimental tool for pursuing that objective is the Nova laser facility, which has been fully operational since 1985. During the past year, we made critical advances in laser science and in developing new technologies to improve Nova's routine performance to its original design levels. During our intense effort in laser development, we also maintained an active experimental program on Nova, emphasizing new experimental and diagnostic techniques.

Since its completion, we have limited the routine performance of Nova because of platinum inclusions in the original large-amplifier glass. New manufacturing techniques have been developed to produce amplifier glass with few platinum inclusions. Indeed, glass that reduces the concentration of platinum inclusions by 3 orders of magnitude has now been demonstrated. Thus, we have committed ourselves to replacing the original glass in the 31.5- and 46-cm amplifiers. A quality-assurance program is being initiated that ensures the new glass will meet current design specifications. When replacement is completed, Nova will be able to routinely operate at its 1ω design energy. Articles under the heading, "Amplifier Development for the Nova Laser," later in this section, review the status of Nova amplifier development.

A new edge-cladding technique for the disks has also been developed. The new technique epoxies the cladding, which suppresses amplified spontaneous emission (ASE), to the edge of the disk. The technique has reduced the cost and difficulty of applying the edge cladding. The new edge-cladding has been tested for over 2000 shots, which is near the expected lifetime of Nova, without failure in a prototype amplifier. A quality-assurance program has been established, and production and installation is scheduled for 1988.

During amplifier development, Nova continues to operate productively as a 50-kJ (1ω , 1-ns) laser for performing target experiments and laser science. The group of articles under the heading, "Nova Operations and Laser Science," describes those activities. Many of our activities were directed toward identifying other areas requiring improved performance for Nova to deliver full frequency-converted energy to a target with the required precision. We constructed a laser experimental test station on beamline 7 for full-aperture testing of Nova performance at high power with new amplifier glass. Subsequent articles discuss the design of this station and our planned experiments. Those experiments planned for full energy require higher accuracy and control of Nova than do present experiments. Two such areas are laser-beam diagnostics and pointing and alignment accuracy of both beams and target, topics that are also discussed in this section.

In 1987 we formed a new Systems Information Group. The formation of this Group, comprised mostly of the Nova Controls Groups and the Data Management and Analysis Group, recognizes the need for a single,

integrated system for Nova operations and data acquisition and management. The activities of this Group, described in articles under the heading, "Nova Computer Systems," include formulating a plan to integrate the numerous existing computational facilities in a single network and upgrading the facilities to a state-of-the-art research tool. The goal of our network is to provide a faster and more responsive flow of information for the complex and diverse needs of the Nova Experiments Program.

Toward the end of this section, we describe the activities of Laser Experiments and Advanced Diagnostics (LEAD). Our emphasis during the past year has been on developing new experimental and diagnostic techniques for better understanding target performance at present Nova operating levels and preparing for more advanced experiments when Nova is at full power. Preliminary experimental results from indirect-drive implosions indicate that fusion yields are close to predictions for one-dimensional spherical compressions (see "Diagnostics Development"). We plan more detailed experiments next year to verify these results and to better understand the compression dynamics. In other experiments, we developed techniques for quantitatively measuring mix at the interface of accelerated materials (see "Target Experiments"). We also conducted an extensive set of experiments to understand the physics of x-ray production from high-Z plasmas (see "Experiments on Multikilojoule Laser-Plasma Interactions").

We significantly enhanced our diagnostic capabilities on Nova in 1987 (see, for example, "Neutron Measurements"). The 22 × Wöfler microscope on the ten-beam chamber was activated and now provides high-resolution, time-resolved imaging capabilities for implosion and hydrodynamics studies. We enhanced our time-resolved framing capabilities by installing two, 4-frame, microchannel-plate x-ray framing cameras with <200-ps resolution to be used for imaging and spectroscopy applications. In preparation for implosion experiments, we developed several new diagnostics for measuring the implosion interval times and for characterizing the imploded core. In areas of advanced diagnostics, we continue to make significant advances in multilayer technology for producing highly reflective coatings for high-energy mirrors and x-ray laser cavities.

In another research area, we completed a significant set of x-ray laser experiments to evaluate various lasing schemes to shorter wavelengths. In separate experiments, we demonstrated, using the 20.6- and 20.9-nm selenium laser, the potential for building an x-ray laser cavity using mirrors and beam splitters. We also used the selenium laser to produce the world's first x-ray laser hologram. This effort is a major step toward developing useful applications of soft x-ray lasers.

In the final group of articles in Section 3, we describe recent developments in target fabrication. In support of our high-Z plasma experiments, we developed new target mixtures of gold and beryllium. These mixtures stretch the technology limits for making atomically mixed components. We also describe two new technologies for ICF targets. The first is a method for producing pure-plastic shells for near-term experiments; the second is development of high-quality foams for more advanced applications.

Nova Operations and Laser Science

Introduction

By 1987 Nova had become a truly operational laser facility. The efficiency of operation had increased dramatically, so that an unexpected reduction in budget and operating personnel resulted in only a minor decrease in the total number of experiments, compared to work during the previous year.

In preparing for the installation of new, platinum-free laser glass, we decided to designate beamline 7 as a prototype beamline. Thus, we would install the new glass first on beamline 7, and we would first test all changes in hardware, settings, and operation on this beamline before implementation on the remaining Nova arms.

As part of our extension of beamline 7, we also built a new experimental facility in the two-beam target-chamber room, which allows us to document the effect of changes to the prototype beamline. The new facility is basically a large optical bench on which large-scale diagnostics can be placed for experiments involving the 70-cm-diam Nova beam.

In the following series of articles, we describe the work done to make Nova a more efficient and precise facility, the improvements made to existing subsystems, and our scientific investigations leading to improved Nova performance both as a laser and in the way the laser beams interact with a target.

Author: D. T. Kyrazis

Operations Summary

During 1987, the Nova Operations Group reexamined its procedures because Nova was then a fully operational laser. The emphasis had changed from making Nova work as a laser to making Nova a tool for conducting scientific investigations in target physics and laser science. All aspects of Nova operations

were examined, including length of operating day, maintenance procedures, shot procedures, personnel functions, and facility utilization. The attention we gave this activity in the very beginning of 1987 helped moderate the impact of the 20% budget cut and corresponding personnel reduction Nova Operations had to implement in February of that year.

Facility operations were made as efficient as possible. Much of the organization and responsibility for the facility was turned over to the senior operators. As part of an efficient operational routine, the program PRESHOT provided a convenient way to define, archive, display, and obtain a hard copy of Nova experiment requests. PRESHOT allows the assignment of a preshot number and a target number to each experimental definition and target design before the experiment is performed. Thus, all groups associated with the experiment can communicate effectively. To further enhance operations, we designated a person responsible for target transportation and tracking, and for organizing the film data processing and distribution. In the past, these problem details had created numerous delays and confusion.

We created a detailed laser system maintenance schedule that specifies weekly tasks. Monday is the one day each week that is dedicated to system maintenance; no experiments are scheduled. To oversee and follow through on these tasks, a position of Maintenance Director was created. The four Shot Directors alternate this position each week. These changes and a more concentrated effort toward efficient system operations resulted in a higher-than-expected shot rate.

Nova Operations changed its operating day from 16 to 12 hr. With these new operating rules, we fired 493 system shots; 78.5% were target shots, and 21.5% were laser science shots. We fired an additional 182 shots on a "ride along" basis. Figure 3-1 shows that in spite of the 25% decrease in operating time, a 15% reduction in personnel, and the imposition of a virtual shutdown for three weeks in September for air conditioning maintenance,

Fig. 3-1. Average shot rate per month for Nova from 1985 to 1987 and for previous systems.

Fig. 3-2. Monthly distribution of Nova shots by number and type for 1987.

our average monthly shot rate was only 15% lower than last year. Our target shot rate was only 9.2% lower. Figure 3-2 shows the monthly distribution of system shots. A shot rate of 40 to 70 shots per month could be realized when a fair percentage of the shots used one or two arms, thereby allowing quick setup and execution. System reactivation and a series of opposed-pert diagnostic (OPD) shots (see "Measurement of Third-Harmonic Pulse Shape On Target," later in this section) limited the number of target shots during October 1987. In November and December, an increase in the number of dedicated ten-beam shots required longer setup and resulted in a reduced shot rate.

In pursuing our goal for routine, dependable operation of the Nova laser as a high energy experimental facility, we had to take precautions. To prevent damage to optical components resulting from platinum inclusion, we limited the output power. We adopted a routine operating

limit of 5 TW/beam for most physics experiments and 6 TW/beam for implosion shots and a few other exceptional experiments. Within these limits, we conducted experiments at pulse widths of 100 ps to 4 ns, with a maximum of 23 kJ of 3ω light delivered on target. Pulse shapes included Gaussian, square, and a few high contrast shaped pulses (see "Demonstration of Shaped 3ω Output Pulses," later in this section).

Figure 3-3 shows the energy ranges and the number of shots taken on Nova in 1987. Beams 6 and 7 were fired more often because they can be fired to both the ten- and two-beam target chambers.

To bring Nova to its full performance specifications, it is necessary to replace the 31.5- and 46-cm amplifier disks with platinum-free glass. During 1987, we dedicated a number of laser science shots to improve the facility and reduce the risk of optical component damage at the higher fluences. We investigated amplifier gain asymmetry, beam spatial-intensity variations,

Fig. 3-3. Distribution of energy and number of shots per arm for Nova in 1987.

target chamber optics transmission, frequency conversion, and damage thresholds of virtually all the optics in the system. The results of these experiments are discussed in the other articles in this subsection.

**Authors: G. L. Hermes
and T. L. Welland**

Comparison of Cost Factors for Shiva and Nova

The optical components in high-energy, high-fluence lasers are subject to damage due to particulate contaminants on optical surfaces.¹⁻³ The evolution of higher-energy lasers at LLNL has necessitated a

continuous reduction in damage to optical surfaces as a result of such surface contamination.

During the conceptual design phase for Nova in 1977, we recognized that it would be expensive to maintain the Nova laser chains at less than 1% obscuration unless we could make significant improvements in the rate at which damage occurred. Figure 3-4 shows the rebuilding costs vs acceptable power loss.

Nova has now been operational for three years, and we have had ample opportunities to analyze damage rates due to contaminants in the system. The design goal for Nova was to improve the damage rate by a factor of 10 over that for Shiva. As shown in Fig. 3-5, the

Fig. 3-4. Rebuilding cost vs acceptable power loss.

improvement we have achieved is a factor of approximately 500.

After investigating the causes of damage in the Shiva laser,³ we developed a plan for Nova that has proven to be successful. We can divide the improvements into the following categories:

- Nova box amplifiers were designed and fabricated with greater attention to reducing contaminants within the amplifier. Our emphasis was on ensuring adherent silverplating and eliminating screw fastness and sliding surfaces.
- Interstage hardware that connects amplifiers with other beamline equipment was a continuing source of contamination on Shiva. The adjustable portion was constructed from Viton, whereas the Nova system is all metal. Nova interstage hardware is cleaned to the same level as that for amplifiers.
- Laser disks in Nova amplifiers are cleaned by a spin-drying technique, which leaves surfaces devoid of particles and films to a significantly greater degree than on Shiva. The disks used in Shiva were cleaned and dried by a common drag-wiping technique. We are using high-pressure spray techniques⁴ much more extensively for cleaning Nova structural components.
- The work-flow pattern in the clean assembly room has been systematized, and greater attention is being given by technicians to contamination-free assemblies.
- On-line laser-bay contamination control is managed to a higher degree on Nova than on Shiva. Laser components are opened and removed from the laser chains in a controlled fashion, with emphasis on preventing the intrusion of contaminants.
- The Nova nitrogen system supplies high-purity nitrogen to the amplifiers. It is designed and constructed as a closed-loop system and provides a cleaner nitrogen environment.
- Nova mechanical design, fabrication assembly, and installation ensured that the mechanical assemblies containing optics or requiring clean surfaces were designed as a system.⁵ This approach prevented adjacent components from

Fig. 3-5. ICF laser disk damage sites relating to surface contamination vs amplifier firings.

cross-contaminating each other and ensured that assembly and manufacturing methods were consistent with our goals.

The program-wide attention we have given to the topic of damage to optical elements from surface contamination has resulted in dramatic improvements in the lifetime of Nova optical components. It is reasonable to expect that many Nova components, but primarily amplifiers, need not be removed and reworked because of surface damage during the projected lifetime of Nova. Such improvements are especially critical in projecting ahead to devices that may be built in the future.

Author: H. G. Patton

Power Conditioning

With over 1000 shots on the capacitor bank, the Nova system has operated longer than any previous large-pulse-power system in the Laser Program. Although the system has proven to be highly reliable, inspections reveal signs of wear on some components.

The capacitor-bank dump resistors continue to fail at a 2% annual rate. The failures appear to be related to the voltage-handling capability of the ceramic resistor assemblies. We are testing a new design that will replace the 2- by

2-in. stack with a 5- by 1-in. stack of ceramic resistors. These resistors are used to discharge portions of the bank not used for a laser shot.

The pulse-forming inductors used for each amplifier circuit have also begun to fail. Only two of the 1576 inductors have failed catastrophically; however, many others have cracks in the epoxy that contains the coil. The cracks are caused by electromagnetically induced forces on the terminals. A brace, which solves this problem, has been designed and tested and will be installed over the next year.

The least-reliable components in the pulse-power system have proven to be the flashlamps. The problems are related to both cracks in the quartz envelopes and to failures of the end seals. The seal failures occur only on lamps from one of our two vendors and primarily on the shorter of the two types of lamps. More than 13% of the 2300 lamps of the latter type have failed. Cracks in the quartz envelopes occur on all types of lamps due to stress in the quartz caused by flaws or contamination. We have initiated an effort in conjunction with the Advanced Driver Group to find solutions to these problems.

We have also added a personnel sweep system to the Nova Personnel Safety Interlock System. This system enables a single operator to sweep the various hazardous areas prior to a shot to provide confidence that no one has entered the area during the sweep. This system enhances both facility safety and operations efficiency.

Several improvements to the Power Conditioning Control System software were implemented this year. New releases of software included control and monitoring of the Facility Sweep System and other facility-oriented monitoring functions. Software was written to allow the Power Conditioning Control System Interface hardware to take on a more global role in the control and monitoring of remote devices, such as diagnostic shutters. The operator interface was also improved through additional, automatic preshot checks of hardware status.

Finally, installation of the platinum-free glass in the power section of the Nova amplifiers began this year. We also

activated the facility for measuring the gain of each amplifier prior to installation on Nova. This facility will operate as part of the glass-replacement quality-assurance plan and will provide detailed information about the differences in performance of individual amplifiers. Such information can be used to help improve laser-performance parameters, such as arm-to-arm power balance.

Author: D. W. Larson

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Beam Diagnostics

Figure 3-6 shows our incident-beam diagnostics, which are designed to measure second- and third-harmonic energy and power delivered to Nova targets. The diagnostic package resides between the potassium dihydrogen phosphate (KDP) frequency-conversion array and the focus lens on each beamline; because of its outside shape, we refer to the package as the diagnostic spool.

Variations in the effective sensitivity of the spool-energy vacuum calorimeters inside the target chamber increase the uncertainty of energy delivered to a target. Disagreement between the spool-intensity-diagnostic and the opposed-port-diagnostic (OPD) intensity vs time streak measurements (see the article "Measurement of Third-Harmonic Pulse Shape On Target," later in this section) has limited the usefulness of these diagnostics in determining pulse arrival time. Therefore, we have devoted significant effort to improving the spool-energy diagnostic, its calibration, and its temporal diagnostic.

Incident-Beam Diagnostics. The second surface of the focus lens provides a primary reflection ghost that acts as a beam splitter to sample the incident beam. This reflection converges quickly because the light in it traverses the $f/4.3$ lens twice. Two fundamental constraints to operation of the diagnostic spool are the result of having chosen the focus lens as the beam splitter and integrator. First, considerable spherical aberration results from the second traverse of the

lens. The caustics that are formed in this beam create intense rings in near-field energy distribution. Microdensitometry of photographs taken at the calorimeter filter plane using the 3ω alignment laser indicates that the caustics are on the order of 40 times the average intensity at this location. The high-intensity ring causes solarization and occasional fracturing of the colored-glass filters in front of the calorimeter, which are used to eliminate fundamental and second-harmonic light. No method has yet successfully eliminated these caustics. Second, reflectivity of the sol-gel-coated second surface of the focus lens can

change if contaminated by vacuum pump oil or vaporized target debris. Variations in reflectivity directly change the amount of energy directed to the diagnostic-spool calorimeter and, hence, its effective sensitivity. It is also possible that reflectivity changes during transmission of a pulse because of laser cleaning of the focus lens.

The original design for our diagnostic spool included a fiber-optic-coupled streak camera as the temporal diagnostic to sample the central 3% (15-cm diam) of the beam. However, recent depolarization experiments have shown that this sample is not representative of the full-aperture

Fig. 3-6. Components of the harmonic-output diagnostics spool for 2ω and 3ω operation. All distances are in centimeters.

beam. We installed a full-aperture sampling configuration using diffuse surfaces in the diagnostic spool on beamline 10. On two shots, we used apertures placed in the beam to verify that responses of this configuration to the inner and outer portions of the beam were similar. Even though spatial sampling of this arrangement appeared to be adequate, when we compared streak data to OPD data, we found agreement in pulse width but not in pulse shape. Plausible causes of this result are transient reflectivity from the focus lens, solarization of absorbing glass filters, or an artifact of ultraviolet radiation in optical fibers. In the near term, we will use the output-sensor streak records to infer 3ω intensity vs time

Segmented Calorimeter Design. We have designed a new calorimeter to replace the 44-cm-aperture vacuum calorimeters used on the Nova target chamber. The calorimeters are used to calibrate incident-beam diagnostics to the energy delivered to target-chamber center and to assess effective transmission of output optics.

Several operational difficulties have arisen with our current calorimeter design. Partial delamination of the absorbing glass from the heat-sink backplate caused a change in sensitivity. The non-equivalence of the electrical heaters intended for in-place calibration is due to the heaters not being properly positioned in the calorimeters to simulate laser absorption. Thus, off-line laser calibration is required.

At high intensities, spatially nonuniform depolarization, along with polarization ellipse rotation, cause depletion of the pump beam that results in back conversion of the third-harmonic intensity in the frequency-conversion crystals. Thus, it is necessary to determine the performance of each of the nine sets of frequency-conversion crystals. (We are considering the use of large-aperture polarizers or Type I/Type II frequency-conversion crystals to correct this decrease in conversion efficiency with increasing intensity.) Our new design for a segmented calorimeter eliminates the shortcomings of our present calorimeters (discussed above) and allows us to assess the performance of individual

frequency-doubling and frequency-tripling crystal sets.

The segmented calorimeter uses absorbing glass, as in the present design; however, the heat sink is sputtered and electroplated onto the glass instead of being glued in place. This process results in a much stronger bond that should prevent delamination. Each segment is shaped and positioned in the target chamber to correspond to the image of one of the crystal sets. The segments are thermally isolated from each other so that measured 3ω energy corresponds to that produced by the crystal set imaged onto it.

Detailed thermal analysis has revealed that the decay time constant of our present calorimeters is controlled by thermal diffusion from the circular beam pattern laterally to the corners of the square calorimeter. By making the new calorimeter the same area as the beam, the decay time constant of the calorimeter is reduced by nearly 50%. This modification reduces the uncertainty of extrapolating back to zero time for determining the initial energy before any thermal losses take place. Thus, noise during data acquisition will pose less of a problem.

A thin copper grid is patterned into the electroplated heat sink near the glass interface to resistively heat the calorimeter for electrical calibration. The grid is insulated by encapsulation in sputtered layers of aluminum oxide. The current and voltage used to heat the grid will be measured with National-Bureau-of-Standards-traceable instrumentation to minimize uncertainty in the calibration. Fresnel-reflection losses from the uncoated absorbing glass are accounted for from measurements on index of refraction. After making a detailed thermal analysis, we believe that the accuracy of this calorimeter is better than 1% in the 1- to 5-kJ (full-aperture) range. A three-dimensional, time-dependent thermal analysis shows that the electrical heating arrangement closely simulates the exponential absorption in the glass of a laser beam. From this analysis, we found radiation losses to be dominated by long-term (tens of seconds) losses, not the short-term front-surface temperature rise. This surprising

result means that a reduced calorimeter response time will also be more accurate.

The temperature rise of the heat-sink plate is measured by thermistors, which have sufficient sensitivity to resolve one millidegree centigrade with lower amplifier gain than the present thermocouple-monitored calorimeters. We also included in the signal-conditioning electronics a reference voltage source for front-to-back electrical calibration of the measurement system. This source should yield precision approaching the 0.05% digitization error.

When beam modulation is included, third-harmonic fluence at the present 44-cm calorimeter position will exceed 5 J/cm^2 when Nova is fully upgraded with platinum-free glass. Therefore, we will position the new calorimeter farther from chamber center to lower the maximum fluence to less than 4 J/cm^2 . This value is below the damage threshold for Schott NG-4 absorbing glass. If damaged, individual segments can be replaced from inside the target chamber without removal of the entire calorimeter assembly. The calorimeter-support system is adjustable both across the beam diameter and toward target-chamber center to align the calorimeter with the array of frequency-conversion crystals.

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Target Alignment

Current target-alignment procedures are acceptable for today's target experiments on the Nova laser but will have to be improved to meet future alignment accuracies with complex targets and beam geometries. To begin to meet the requirements of these more complex targets and geometries, we have made improvements to the optics and mechanics of the prealignment system,⁶ to the optics and illuminators for the target-alignment viewers (TAVs) on Nova,⁷ and to the cameras on both systems.

Current Alignment Procedures. Today's target-alignment procedures on

Nova involve several steps. First, the target is assembled and photographed, and has its physical dimensions recorded with an optical comparator. Then the target and its identifying data are delivered to the Nova laser experimental area, where the target is installed on the Nova target positioner. There an alignment technician manipulates the target positioner while visually comparing the dimensional data and photographs with the television image produced by the TAV. The target is presumed to be aligned when the technician, or the physicist who is conducting the experiment, judges that it is positioned correctly.

To alleviate the difficulty of visually judging when targets are aligned, we have begun integrating a prealignment system on Nova for future experiments requiring beam-to-target alignment accuracies of $\pm 25 \mu\text{m}$. Backlighting targets, for example, will need focused beams aligned to 10- to $50\text{-}\mu\text{m}$ -diam fibers and target rotational alignment to within 0.1 to 0.5° , which are difficult to achieve with the present system. Although integration is not yet complete, the preparations for implementing prealignment have driven many improvements in both Nova target alignment and the prealignment system.

Prealignment System. To meet the more stringent target-alignment requirements, we have been using an alignment technique involving image comparison between two similar TAV systems. An optical system constructed in the target fabrication assembly area emulates the three TAVs in Nova. This system, known as the prealignment system, is similar to the Nova system both in optics for target imaging and mechanics for target positioning. In addition to the three orthogonal linear axes and one rotational axis of motion found in the Nova target positioner, this system introduces an additional goniometric axis. This new axis permits $+45^\circ$ of rotation about the optical axis of TAV 1 (the polar TAV), which allows the target to be so positioned that TAV 2 or 3 can simulate the view of the target from any beam or diagnostic port.

After a target is fabricated, it is taken to the prealignment system and placed on the target-positioner emulator. Under the direction of the shot physicist, the

target is positioned in the correct orientation as viewed from the three TAVs. The images are then digitized and stored on magnetic media and delivered to Nova with the target. The target is placed on the Nova target positioner and moved into the TAV fields of view so that the live images can be compared through real-time image subtraction with the prealigned images that represent the desired target orientation. When the absolute value of the difference images is null or at a minimum, the target is declared aligned.

The prealignment computer imaging system can potentially use every pixel (512×512 pixels) in the field of view or the target silhouette as an alignment fiducial. Complex target geometries, which previously were the hardest to align in Nova, have become good candidates for prealignment. They provide a sufficiently unique geometric image and readily show linear or rotational positional errors with the image subtraction method.

Prealignment System Changes. We replaced the original manual positioning stages used for the linear axes in the prealignment station with motorized, encoded stages that more closely duplicate the positioning controls found on Nova and allow more precise target positioning. A range of ± 25 mm in x and y and ± 12.5 mm in z was chosen. The controller and digital encoder readout allow the target to be moved in three axes with a backlash of $6 \mu\text{m}$. If target travel is kept unidirectional, the backlash is reduced to less than $2 \mu\text{m}$.

Tests on the prealignment system early in the year indicated that geometric distortions in the images in Nova and the prealignment TAVs would preclude effective use of the image-subtraction method. The video cameras in both areas were of the vidicon type, which placed a limit on the amount of geometric distortion that could be corrected. While linearizing the sweep circuits in the driving electronics worked initially, drift eventually rendered this correction useless. We decided then to change all the Nova and prealignment TAV cameras to high-resolution (604×488 -pixel), solid-state, charge-coupled-device

cameras so, with a fixed-array silicon-chip sensor, any geometric distortion between the two systems could be attributed to the optics. With the new cameras in place, the reticle pattern mismatch between Nova and prealignment is less than 1% of the full field of view.

Target Alignment System Changes. The optical resolution of TAVs 2 and 3 in Nova was not sufficient to allow us to position target components to the accuracies required for future experiments. An analysis of the optical system showed a single relay optic was used to transfer the image of the reticle/target plane to the magnification section of the TAV. When we replaced this relay optic with a pair of longer focal-length lenses used as a 1:1 relay pair, resolution doubled in the 2-mm field of view and tripled in the 1-mm field of view.

The original illuminators for TAVs 2 and 3 used an He-Ne laser source. Although the illuminator optical system was designed to eliminate laser speckle and edge diffraction patterns on target, we could not reduce these effects enough to prevent unacceptable image degradation in the TAVs. Therefore, we removed the laser and replaced it with a narrow-band, incoherent illuminator. Currently, all three Nova TAVs utilize incoherent illuminators.

Progress Summary. Our goal with the prealignment system is to position targets to their required accuracies. Positioning will be done by comparing an image of the target in the target chamber to the same target prealigned in the prealignment system. Although integration of the prealignment system on Nova is not yet complete, we have been able to use the system to perform other useful tasks. For example, we have used the system to ensure that when a target has been aligned prior to a shot and moved away, it returns to its pre-shot position. Since this results in a direct subtraction of the Nova TAV images from themselves, the null-signal image gives a very strong indication of misalignment. For a 2-mm field of view, we can readily see a positional error with one to two pixels of misalignment. An error of one pixel

corresponds to a 4- μm positional error for a 2-mm field of view.

We have also used the prealignment system in the target-fabrication assembly area to review prototype target designs and to preview final target assemblies before they are sent to Nova. This system accelerates target design and inspection procedures considerably.

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Target Chamber Optics

Damage to Target Chamber Optics.

Laser-related damage to the optical components associated with the ten-beam Nova target chamber has recently become an issue and may limit the amount of 3ω energy that can be delivered to targets. In the past year, we have observed two types of damage to the chamber optics: high-energy shot damage and accumulated shot damage. High-energy shot damage occurred in September 1986 during two high-energy (>3.2 kJ) 3ω calibration shots into the target chamber. These shots severely damaged the focus lens and the retroreflector⁸ located on the single beamline used to conduct both shots. Accumulated shot damage, on the other hand, occurs continually as the laser is repeatedly operated at relatively moderate (≤ 2.5 kJ per beamline) 3ω fluences. This accumulated damage is

responsible for the deterioration of all chamber optics, including the focus lenses, debris shields, reflectors, and 44-cm-aperture calorimeters. As we study the causes of both types of damage, we are taking steps to protect the optics from further damage.

High-Energy Shot Damage. In September 1986, we fired two 2.5-ns calibration shots on beamline 10. These shots, which had 1ω energies of 10 and 12.5 kJ, were propagated to the ten-beam target chamber, converted to the third harmonic, and measured with the 44-cm-aperture chamber calorimeter. Inspection of the chamber optics following these shots revealed an ~ 2 -cm-diam fracture centered on the vacuum surface of the focus lens and a much larger fracture that passed completely through and extended across the face of the retroreflector (see Fig. 3-7). Concerned that the fracture depth in the focus lens, which serves as a vacuum barrier for the target chamber, had reached the critical size for growth as determined by modeling, we removed the lens from the chamber for structural testing.

To determine the cause of the damage, we developed a computer model of the chamber optics and investigated it using ray traces and a program designed specifically to track ghost reflections and locate caustics.⁹ We also performed several laser tests. To produce beam photographs at the center of the lens and retroreflector, we fired low-energy, 100-ps

Fig. 3-7. Fractures in the beamline-10 (a) focus lens and (b) opposed port retroreflector, incurred during two >3 -kJ, 2.5-ns, 3ω shots in September 1986.

laser shots into the target chamber. We also used the harmonic alignment beams to map observable ghosts both inside the target chamber and on a full-scale model of the chamber optics constructed in the adjacent two-beam experimental facility.

The computer model identified a 3ω reflection from the flat surface of the chamber calorimeter as a possible cause of the focus lens damage. The process is diagrammed in Fig. 3-8. Here a large percentage of the beam reflected from the calorimeter passes around the pinhole at the center of the target chamber and re-enters the lens, where it undergoes several internal reflections before focusing on the axis of the lens. The intensity of the axial caustic is highest (roughly 10 times the average beam intensity) near the vacuum surface of the lens. This calorimeter reflection is also responsible for damaging the target-bay final turning mirrors. In this case, the lens refocuses

Fig. 3-8. Ray trace showing a ghost focus on the lens axis called by an opposed port calorimeter reflection.

the beam from the calorimeter directly onto the surface of the mirrors. Although this same refocusing occurs on all chamber calibration shots, the damage incurred is localized, and no steps have yet been taken to eliminate it. To prevent future damage, a large-diameter beam block will have to be placed around the pinhole, or the calorimeter will have to be tilted to prevent the reflected beam from reentering the lens. Bead-blasting the surfaces of the calorimeters is also an option. Until one of these steps is taken, high-energy calibration shots have been discontinued.

The source of the retroreflector damage remains unexplained. Recent evidence suggests that stimulated opto-acoustic coupling in the glass may be the damage mechanism (and the cause of the crack in the focus lens). Therefore, until the damage is better understood, we have removed all reflectors from the target chamber, reinstalling them when needed to calibrate beam diagnostics. At that time, calibrations are performed at normal (<2 kJ/beamline) 3ω energy levels.

Accumulated Shot Damage. The optical components directly exposed to the target chamber's vacuum environment accumulate damage at a steady rate. The focus lenses, debris shields, and 44-cm-aperture calorimeters (the reflectors have been removed from the chamber) show a consistent damage pattern. After a brief exposure to the chamber environment, each optic develops a surface haze. After a limited number of laser shots, the surface of the optic is altered enough to distinguish which regions of the optic are exposed to the high-power beam. Eventually, small surface fracture sites or "pits" (Fig. 3-9) begin to develop in the regions exposed to the beam and grow in size and number with successive laser shots until the exposed surface takes on a frosted appearance. This damage is 3ω dependent and occurs during shots with aperture-averaged 3ω fluences below 1 J/cm^2 .

Figure 3-10(a) shows a badly damaged focus lens. On the sol-gel-coated surface of the lens, haze is clearly visible in the beam shadows caused by the crystal array and the 46-cm-diam split disk. Damage

is also visible as a frosted ring that surrounds the central apodized region of the lens and as isolated frosting sites scattered across the aperture. The frosted ring is about 1 cm wide by 1 mm deep on some lenses, and the isolated frosting sites are about 5 mm in diameter and 1 mm deep.

Figure 3-10(b) shows a damaged debris shield. Debris shields are nominally exposed to the highest 3ω fluences on the laser (i.e., to an average aperture fluence of $\sim 0.9 \text{ J/cm}^2$ on 2.5-kJ, 3ω shots) and are placed between the focus lenses and the target to prevent target debris from damaging the lenses. Damage caused by the impact of target debris is clearly visible at isolated sites in the periphery of the shield. These sites are few in number and do not pose a threat to laser operation. Severe shield damage is evident only in regions exposed to the 3ω beam and, in some instances, is

restricted to beam segments that correspond to specific, high-conversion-efficiency KDP (potassium dihydrogen phosphate) crystals. The individual pits on the shield surface eventually become so numerous in these regions that they grow together and form the severe frosting shown in the figure. In some instances, the damage has been so extensive that shield transmission has decreased to less than 50%.

The 44-cm-aperture calorimeters are the primary energy diagnostic of the Nova target chamber. Although exposed to 3ω fluences ~ 2.3 times higher than those incident on the debris shields, the calorimeters are exposed to fewer shots and therefore sustain less damage. Damage occurs in those regions of the aperture where the beam is highly modulated by diffraction. The retroreflectors exhibited similar damage before they were removed from the target chamber.

Fig. 3-9. Optical micrographs (a and b) and SEM scan (c) of pit formation on the output surface of a Nova debris shield.

Fig. 3-10. Damaged (a) focus lens and (b) debris shield, showing frosting and pitting.

To understand the damage, we have been conducting a series of tests. We have obtained swipes from a damaged debris shield and analyzed them chemically, using gas chromatography and mass and FTIR spectroscopy. We have also sectioned a damaged shield and analyzed the pieces both optically under a microscope and with a scanning electron microscope (SEM) and an Auger spectrometer. To investigate the chamber vacuum environment directly, we stored both bare and sol-gel-coated witness samples inside the target chamber, later removing them and analyzing them, using the above techniques. We also measured the optical transmission of these samples, using a spectrophotometer. Direct measurements of vacuum quality were also made with the target chamber's quadrupole mass spectrometer.

Several of the above tests have identified organic contamination of the target chamber to be a problem. Chemical analysis of the debris shield swipes has uncovered ether and phthalate compounds, indicating the presence of plasticizers or oils in the chamber. The presence of long-chain hydrocarbons in the chamber is confirmed by the residual gas analysis scans made with the mass spectrometer. More conclusively, Auger depth profiles of witness samples exposed to the target chamber's vacuum environment for a short period of time reveal that carbon has penetrated the sol-gel-coated samples to a depth of ~ 40 nm. Transmission measurements of similar samples show a significant increase in absorption at wavelengths below 400 nm after only a few weeks inside the chamber. Specifically, we have measured a 3% drop in transmission at 350 nm after a 30-day exposure with no change in transmission at 530 nm.

We believe that the increased absorption at the third harmonic, resulting from organic contaminants in the chamber, is responsible for lowering the damage threshold of the optics below the $\sim 1\text{-J/cm}^2$ 3ω fluences at which we currently operate the laser. Damage studies to confirm this hypothesis are currently being conducted. Although we do not understand all aspects of the damage (e.g.,

the ring structure of the damage on the focus lens), we are taking steps to protect the optics from further damage. We are removing damaged focus lenses from the chamber and refinishing them as spares become available, periodically cleaning the chamber optics to extend component lifetimes (see the following article), and making an effort to clean the chamber by adding traps to the vacuum system and improving system operating procedures. We realize that a damage problem at the current Nova operating levels threatens our ability to achieve Nova Phase II energy-level goals. By increasing our understanding of current damage mechanisms, we hope to identify the chamber operating procedures that must be observed to meet those goals.

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Structural Damage and Analysis of Focusing Lenses. The Nova final focusing lenses act as a vacuum barrier between the target chamber and the KDP frequency-conversion crystal arrays. The lenses are made of Corning 7940 silica glass, and a pressure difference of 100 kPa exists across them under normal operation. We routinely inspect the lenses because the glass is brittle; should failure occur, potential implications regarding personnel safety and possible damage to expensive equipment may arise. During our routine inspections, we discovered two forms of damage on the vacuum (tension) side of the lenses.

First, we found a 3-cm-long, star-shaped crack at the center of the vacuum face on one lens. The crack may have occurred because of back reflection from a calorimeter, which concentrated laser energy at the damaged location. However, the exact cause remains to be determined. We removed this lens immediately and pressure tested it to destruction. Failure occurred when a pressure difference of 150 kPa was placed across

the lens for 100 min (Ref. 10), $1.5 \times$ the magnitude of pressure under normal operating conditions.

We then made and tested four specimens from one of the remaining pieces of glass; these specimens were chevron-notched for testing fracture toughness. We found that the minimum fracture toughness¹¹ was $770 \text{ kPa} \cdot \text{m}^{1/2}$, with an average value of $820 \text{ kPa} \cdot \text{m}^{1/2}$. We also performed one flexure test and found the tensile strength to be 39 MPa. These results compare with published values^{12,13} of $750 \text{ kPa} \cdot \text{m}^{1/2}$ and 49 MPa.

Second, we observed pitting on the vacuum face of all final focusing lenses. This pitting was opposite a groove that was machined into the face, exposed to 100-kPa pressure, and then apodized by bead blasting. The pitting had an annular form, with a diameter slightly larger than the edge of the apodizer. The pitting increased in width and depth with laser operation. Damage occurred as light passed through the lens near the edge of the apodizer and was diffracted such that it concentrated in the damage area. Such concentration heated the glass, produced thermal stress, and chipped off shards. Once started, the previously damaged area was partially opaque and absorbed more light during the next laser shot, producing additional thermal stress and chipping off more shards. The damage rate was associated with carbon contamination and was slowed by frequent cleaning, as described in the previous article.

We were initially concerned that stress concentrations present in the sharp corners of the damaged area could result in catastrophic crack propagation should a flaw be present there. However, after more refined inspection, we found that small shards of glass were being chipped away during laser operation. Thousands of these shards were produced, and in all cases the cracking that produced the shard propagated to the vacuum surface of the lens without causing catastrophic failure.

Lens Stresses and Safety Factor.

Williams^{14,15} used a finite-element stress-analysis code to predict the stress state throughout a final focusing lens with and without an annular apodized groove on

the compression (pressure) side. Williams found that the peak tensile stress with a 100-kPa pressure difference across the lens was 3.5 MPa and occurred on the lens vacuum face at the center. Stress on the vacuum face was nearly constant near the center of the lens and reduced to 2.3 MPa near the edge. Williams also found that tensile stress on the vacuum face was about the same with a groove as without one. A peak compressive stress of 9 MPa occurred at the bottom of the apodized groove; however, compressive stresses are not of concern because fracturing starts only where tension exists.

Hauber¹⁶ used a safety factor of 8 based on the material ultimate strength with brittle, fused silica and other glasses in other LLNL applications. Comparing Williams' predicted peak tensile stress of 3.5 MPa to our experimental ultimate tensile strength of 39 MPa shows that we have a safety factor of 11—larger by $1.4 \times$ than that used by Hauber. However, fracture mechanics is a method more likely to predict failure in brittle materials. The residual crack radius a (critical crack length) necessary to cause failure under our conditions of peak tensile stress, with $\sigma = 3.5 \text{ MPa}$, and a fracture toughness $K_{Ic} = 750 \text{ kPa} \cdot \text{m}^{1/2}$ is $a > (K_{Ic}/\sigma)^2/\pi = 15 \text{ mm}$. This flaw size required to cause failure is large compared with what we envision to be present and suggests that Hauber's criterion is reasonable.

We applied fracture mechanics to calculate K_{Ic} in the lens that we pressure tested to failure. Visual inspection showed that the initial crack depth was about 1.5 cm. The fracture toughness so calculated was $1.2 \text{ MPa} \cdot \text{m}^{1/2}$, a value 50% higher than that determined with the fracture-toughness specimens. This result suggests that the actual crack depth was less than our assumed value. If we used this higher value of K_{Ic} , we would calculate an even longer critical crack length that would result in an even larger factor of safety.

Finally we addressed the concern that stress concentrations at the corners of the pitted damage area in Fig. 3-11 could result in catastrophic failure. The highest tensile stress occurs at the vacuum face

Fig. 3-11. Nova final focusing lens showing stress values and the damaged area. (Negative values of stress denote compression, positive values denote tension.)

of the lens and decreases with depth into the lens. If a small flaw were present in the glass at the corner of the damage area, and a crack started to propagate, the crack would tend to curve toward the vacuum face where the highest tensile stress is located. Further, flaws oriented parallel to the lens face or oriented so that the tip of the flaw is closer to the lens face tend to propagate more easily because the tip region where fracture occurs is located in a field of high tensile stress.

Alternately, if the flaw were oriented so that the tip is located further into the lens in a field of lower tensile stress, then propagation is less likely. The latter crack orientation could cause catastrophic failure because the crack would most likely not be able to reverse its direction before propagating through the thickness of the lens.

Consider an example of a damaged area 1-mm deep and a flaw oriented into the lens with its tip located 15 mm away from the lens face. Dowling¹⁷ suggests that for crack lengths greater than about 0.1 the depth of the damaged area, a

value of crack length equal to the sum of the depth of the damaged area plus the length of the crack be used for the term a without consideration of stress-concentration factor. In this example, the length of the crack would be $14 \times$ the depth of the damaged area—well beyond the region where stress-concentration factors need be considered. If the flaw propagates, it would need to be in a tensile-stress field of about 3.5 MPa. The nominal tensile-stress field calculated by Williams at a depth 15 mm away from the lens vacuum face is about 1.5 MPa. We conclude that no crack propagation is expected, and our experimental evidence verifies this conclusion.

Summary. Nova final focusing lenses are inspected routinely because catastrophic failure involves issues of potential personnel safety and could damage expensive equipment. We observed two types of damage. A 3-cm-long, star-shaped crack appeared on one lens, which may have occurred because of back reflection from a calorimeter. The lens was removed immediately; however, the exact cause of the crack remains to

be determined. The lens was tested to determine structural properties of the lens glass.

The second type of damage occurs continually with laser operation and consists of pitting on the vacuum face opposite the apodizing groove. Damage results because refracted light is concentrated in the area. Small shards of glass are chipped off; however, both analysis and experimental evidence show that this type of damage should not result in catastrophic failure.

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Nova Optical Science

Introduction. In Nova optical science, we are trying to understand the constraints that presently limit output from the Nova laser and frequency-conversion system and to apply this knowledge so that we can increase the Nova capabilities to irradiate fusion targets. Our principal near-term goal is to demonstrate that 40 to 70 kJ of 0.35- μm light can be delivered to targets in the ten-beam target chamber after Nova Phase II is implemented. To reach this goal, we must understand the physics and engineering limits on the 1.05- μm laser output after the platinum-particle-containing glass in the high-fluence portion of the system has been replaced, and we must demonstrate an efficient, high-power third-harmonic-conversion system.

In the following 11 articles, we describe several experimental and calculation efforts that contributed to meeting our near-term goal. One important item was preparations for testing the energy- and power-output capabilities of a Phase II prototype beam and for testing third-harmonic-conversion options in a full Nova beam during the first half of 1988. Our secondary goals during the year have been to better measure the energy and power we presently deliver to Nova targets and to extend our pulse-shaping

and fourth-harmonic-generation capabilities. We report on measurements of third-harmonic energy and power delivered to the center of the ten-beam target chamber. We took advantage of the newly constructed third-harmonic-conversion station to develop our ability to produce predefined pulse shapes at the third harmonic. We also performed off-line testing to study fourth-harmonic-generation in KDP. Our activities during 1987 significantly contributed to our present and future capabilities to perform precise, high-energy target-irradiation experiments with Nova.

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Nova Phase II Goals and Schedule.

Our Phase II activities will allow the Nova laser to routinely irradiate targets with 40 to 70 kJ at a wavelength of 0.35 μm , the third harmonic of its fundamental wavelength. Phase II includes replacement of phosphate-glass laser disks and BK-7 glass components in the high-energy portion of the laser amplifier system with glass that is free from metallic-platinum particle inclusions. Phase II also includes improvement of the frequency-conversion system to increase its high-power conversion efficiency.

Work toward replacing the laser disks was started in calendar year 1985 and is discussed in the 1985 and 1986 Laser Annuals. This effort has progressed well, and we expect to complete replacement of the laser disks in late 1988. Likewise, we have placed orders for fused-silica lenses to replace the BK-7 glass lenses at the output of the final two spatial filters in each of the amplifier beamlines and fused-silica substrates for the split-disk apodizers. Delivery and installation of these components is scheduled to be completed by the end of 1988. After replacement is completed, we project that the laser will be able to routinely provide 100 to 120 kJ at the 1.05- μm wavelength to the frequency converters in a 2.5- to 5.0-ns pulse without damage to optical components. We will make a decision on any necessary upgrade to the frequency-conversion system after we have completed testing of the first upgraded beamline.

The initial assessment of Phase II performance will be done on the first completed beamline (beam 7), which is scheduled for early February, 1988. On this prototype beam, we plan to measure the component and system characteristics useful as input to computer models of system performance, to demonstrate the high power and energy capabilities at 1.05 μm , and to test possible third-harmonic-conversion options at full aperture. At the 1.05- μm wavelength, we expect to be able to demonstrate repeated shots at 8.0-kJ output in a 1.0-ns pulse and at 12.0-kJ output in a 2.5-ns pulse. The third-harmonic-conversion options that we plan to test are the present Type I/Type II KDP system, the Type I/Type II KDP system, and Type II/Type II KDP with repolarization. Our measurements will allow us to choose the most cost-effective way of obtaining high-power third-harmonic conversion that will meet Nova third-harmonic energy goals and to provide precise pulse shapes at the third harmonic for target experiments.

Our progress toward these goals during the last year included construction of a third-harmonic-conversion station with 3 ω beam diagnostics on beam 7, the purchase and characterization of a 74-cm-aperture polarizer, the start of procuring a set of Type I KDP doubling crystals for building a Type I/Type II array, and formulation of a plan for the prototype beam measurements.

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Nova Phase II Projected Target-Irradiation Capabilities.

Introduction. We have estimated the energy and power at 0.35 μm available from Nova Phase II for irradiating targets. Our estimates include the future 1 ω drive available to the frequency-conversion arrays and the efficiency of converting the 1 ω drive to 3 ω and delivering it to the target. After the new glass is installed in the power section of the laser-amplifier system, the energy delivered to the frequency-conversion arrays will be limited by B-induced self focusing, the threat of damage to optical components, and stimulated rotational Raman scattering

(SRRS) in the air. We used our current understanding of these effects to estimate the energy available for frequency conversion for pulse lengths between 100 ps and 4.0 ns.

To obtain the resulting 3 ω on target, we assumed that the beam depolarization is corrected, and the presently observed back conversion of 3 ω at high power will be minimized. Transmission of the third harmonic from the frequency-conversion arrays to the target, including the effects of obscurations and Fresnel losses, was assumed to be 85%, close to the maximum value expected for clean, freshly coated optics.

Energy Limits at 1.05 μm . The most serious issue limiting high-energy operation of Nova is multishot, beam-induced damage to optics. When such damage occurs, it accumulates with repeated shots, degrades beam quality, and leads to further damage and an enhancement of other undesirable nonlinearities. The maximum energy that can be extracted from a beam without unacceptable damage to components is difficult to determine precisely. It depends upon the distribution of fluence in the beam at the threatened components and on the damage thresholds of the surfaces and/or materials in the component, both of which have large uncertainties and may change from beam to beam.

We predict the spatial distribution of the fluence in a typical Nova beam at threatened components with the MALAPROP code. We carefully chose input data to MALAPROP to match the average chain parameters and included a noise-generation model to account for imperfections in optical components. The code models the effect of amplification, beam propagation, spatial filtering, and nonlinear growth of modulation resulting from self focusing.

After the platinum-containing glass is removed, the components in the laser amplifier and beam-transport system most subject to damage will be the input lenses to the final spatial filter and the thin-film coatings on the large turning mirrors in the beam-transport system. The final spatial-filter input lenses experience the highest fluence in the laser. The substrate material of these lenses is

fused silica, and they have SiO_2 sol-gel antireflecting coatings on their surfaces. Our best estimate of the damage threshold for these coatings is 11 J/cm^2 at 1 ns , and the damage threshold scales with pulse length as $t^{1/2}$ (Ref. 18). The thin-film coatings on the turning mirrors are $\text{ZrO}_2/\text{SiO}_2$ multilayer coatings designed to give high reflectivity ($>99\%$) for $1.05 \mu\text{m}$ and $>30\%$ reflectivity for the second and third harmonics of this wavelength. Three or four such mirrors are located in each of the beamlines between the amplifier output and the target chamber. The damage threshold of these mirrors is 6 J/cm^2 , which is constant with pulse lengths from 1.0 to 10 ns .

In determining the maximum energy permissible from a Nova beamline, we estimated the peak fluence experienced by the threatened components, using the MALAPROP computer code, varying the energy and pulse duration of the beam. Using the average damage thresholds, we limited the output energy accordingly. Figure 3-12 summarizes our analysis. For Gaussian pulses of lengths less than 0.6 ns , the primary constraint to the output power is self focusing, i.e., beam breakup. We assumed this limit was reached when MALAPROP predicted that the peak-to-average fluence modulation exceeded 2 to 1 at the KDP arrays. For square pulses with durations between 0.6 and 2.5 ns , beam energy is limited by damage to the coatings on the input lenses to the final spatial filter. For durations greater than 2.5 ns , the limit is imposed by the coatings on the turning mirrors. Upper-limit energies are 80 kJ at 1 ns , 110 kJ at 2.0 ns , and 125 kJ at 3 to 5 ns . It should be emphasized that these energies are limits at which the calculated peak fluence in the beam reaches the average damage threshold.

Some conservatism is built into these estimates from the assumptions used in MALAPROP. On the other hand, the damage thresholds have uncertainties, and real beams have nonuniformities that are not accounted for in the calculations. There is substantial danger that repeated shots at the upper-limit energies will cause significant damage. One strategy to minimize the danger would be to limit routine shots to lower energies

Fig. 3-12. Limit of energy at $1.05 \mu\text{m}$ available from Nova vs pulse duration after replacing platinum-containing glass.

Table 3-1. Thresholds for 1% conversion to SRRS.

Pulse length, τ (ns)	Threshold IL (TW/cm)
0.5	17.0 to 22.0
1.0	12.0 to 16.0
2.5	9.6 to 12.0
5.0	8.8 to 11.5

(say, $0.80 E_{\text{max}}$) and to allow the limit to be exceeded only for a small fraction of shots where programmatic or experimental goals require the highest energy. Such a strategy would keep the cost of replacement optics low and still allow us to reach important milestones requiring high energy.

Nova beams propagate 45 to 60 m through air in traveling from the output of the amplifier beamlines to the frequency-conversion arrays on the ten-beam chamber. Such propagation presents the possibility of significant losses from SRRS. Using a Nova beam, we measured the threshold for the onset of beam perturbations from SRRS in air for square pulses with lengths from 0.5 to 5 ns and characterized the behavior well over threshold.^{19,20} We defined the threshold as the product of intensity times path length (IL) at which 1% of the beam is converted by SRRS. Because SRRS is transient in nitrogen for pulse durations less than about 10 ns , it was necessary to explicitly measure the time dependence.

Table 3-1 shows the threshold IL products at several pulse lengths deduced from experiments. From these data, we estimated the limitation that SRRS might impose on energy available to the frequency-conversion arrays at the ends of the

longest air paths of the beams to the ten-beam target chamber. Beams 3 and 7 have air paths of approximately 61 m from the last spatial-filter output lens to the frequency-conversion arrays.

Figure 3-13 summarizes our estimates and compares them with the maximum energy limits. The shaded area in the figure shows the uncertainty in the maximum energy available to the KDP arrays with <1% of the beam energy converted by SRRS. For pulse lengths between 0.5 and 1.5 ns, the band of uncertainty overlaps the maximum performance limits,

discussed above. Because it is probable that we can satisfactorily operate slightly above IL products that convert 1% of the beam energy, we conclude that SRRS is presently not a constraint on delivering energy to the ten-beam target chamber.

Lengths of the beam paths in air to the two-beam target chamber are significantly longer (85 and 100 m) than the value of 61 m considered in Fig. 3-13. Thus, SRRS will limit the energy that can be delivered to the frequency-conversion arrays on this chamber. The SRRS threshold occurs at 4 to 5 kJ for a 1.0-ns square pulse. To overcome this limitation, we have made provisions to replace the air with argon in the beam tubes over a large fraction of the path. Argon will allow us to deliver the full energy available from Nova to the two-beam chamber.

Third-Harmonic Conversion. Efficient third-harmonic conversion at high intensity is presently prevented by small amounts of nonuniform polarization in the Nova beams.²¹ Such polarization causes a nonoptimal ratio of intensity mixing for Type I/Type II conversion and thereby degrades conversion efficiency. We have demonstrated that greater than 70% conversion can be attained by repolarizing the output beam²² or by changing to a Type I/Type II scheme for third-harmonic conversion.²³ Here, we assume that the high-intensity conversion efficiency (2.0 to 2.5 GW/cm²) will be improved to the range of 60 to 70%, as shown in Fig. 3-14. The upper end of this range can be reached with either scheme; the lower end might be reached by using a low-contrast polarizer or partially correcting the birefringence in the optics.

From the 1ω drive limits in Fig. 3-12 and the estimated conversion efficiency in Fig. 3-14, we derived the 3ω energy available to the target, as shown in Fig. 3-15. In calculating 3ω energy, we assumed that the 3ω transmission to the target, including obscurations from the KDP array and target shadow shield, is 85%. This value is near the maximum that can be expected with a clean, well-coated lens and debris shield. Figure 3-15 shows the maximum 1ω drive and the routine experimental limit of $0.80 E_{\max}$, suggested above. The bands of 3ω energy result from the upper and lower

Fig. 3-13. Threshold for conversion of 1% of beam energy by SRRS in the longest beam paths to the ten-beam target chamber (61 m).

Fig. 3-14. Assumed third-harmonic conversion efficiency of Nova KDP arrays. ▼

Fig. 3-15. Third-harmonic energy available to targets in the ten-beam target chamber after implementing Phase II.

limits of frequency conversion from Fig. 3-14. Maximum third-harmonic energy occurs at a pulse duration of about 2 ns, where the maximum energy to the target is 55 to 65 kJ; the routine limit is 40 to 45 kJ. At 1 ns, the corresponding energies are 40- to 50-kJ maximum and 30- to 40-kJ routine.

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Diagnostics for the Third Harmonic of the Phase II Prototype Beam. We have constructed and activated the Phase II diagnostic station, a full-aperture experimental facility for the Nova Phase II experiments. The experiments on the diagnostic station are as follows:

- To calibrate the diagnostics and make frequency-conversion measurements for 5-kJ, 1-ns square pulses.
- To characterize the beamline with the new high-damage-threshold laser glass in the 31.5- and 46-cm amplifiers.
- To investigate third-harmonic conversion efficiencies with a prototype (74-cm) KDP Type I/Type II crystal array. This article discusses only the first of the three experiments.

Diagnostic Station Design.

The Phase II diagnostic station was built in the Nova two-beam target bay on beam-line 7. The optical configuration (see Fig. 3-16) allows us to obtain simultaneously full-aperture data from several instruments: two calorimeters, a streak camera, a spectrometer, and a near-field camera.

The KDP crystal array is approximately 69 m from the last output spatial filter. This configuration approximates the longest path from the final spatial filter lens to KDP crystal array on the Nova ten-beam chamber.

The fused-silica beam splitters were fabricated with a 35° wedge, and sol-gel was coated on the back surface for optimum antireflection (AR) at the third harmonic. This scheme allows the main beam to be sufficiently attenuated and separated from unwanted secondary Fresnel reflections before reaching the diagnostics. The 3 ω energy and temporal diagnostics are placed in the path of front surface reflections. As a result, their readings will be unaffected by reflectivity changes in the sol-gel AR coatings.

The negative lens placed after the Nova focus lens extends the focal length to give us a slowly converging beam.

Fig. 3-16. Phase II prototype beamline diagnostic station.

Additional optics then direct the beam into the diagnostics.

Calorimeters. To calibrate the energy diagnostics in the output sensor and in our diagnostic station, we compared them against a full-beam reference calorimeter (in these cases a Nova 74-cm calorimeter). Both the 1 and 3 ω energy diagnostics are calibrated against the same 74-cm calorimeter; some of the uncertainties in absolute energies should therefore be unimportant in determining relative quantities such as conversion efficiency.

We calibrated the output sensor diode against the 74-cm calorimeter temporarily placed in front of the KDP crystal array on the diagnostic station. From four calibrations, we estimate that we can measure the 1 ω energy entering the array to within 8% (2σ) accuracy and 2.4% (2σ) precision.

A large array of UG-11 filter glass placed directly in front of the 74-cm calorimeter filtered out 1 and 2 ω light during the calibration. We accounted for losses in the beam splitter and the UG-11 array in determining the 3 ω energy out of the KDP array. From five calibrations, we estimate that we can measure the 3 ω energy exiting the KDP crystal with the 5 cm calorimeter to within 8.5% (2σ) accuracy and 4% (2σ) precision.

All 3 ω calibrations were done at low power (1 to 1.5 TW) to prevent solarization of the UG-11 array.

Streak Camera. The LLNL streak camera uses a charge-coupled device (CCD) as its image recording system. Calibrating the streak camera involved timing and filtering adjustments. Three optical signals are multiplexed onto the streak camera: 1 ω coupled through an optical fiber from the output sensor, a 3-GHz comb generator that serves as a timing reference, and 3 ω (air path) out of the KDP crystal array. The Laser Experiments and Advanced Diagnostics group recently characterized the flat-field response of this camera and found a two-to-one variation in the temporal direction.²⁴ We can correct for this variation without a significant increase in the noise.

Spectrometer. We obtained data to confirm alignment and a proper line focus through the cylinder lens/diffuser configuration in front of the spectrometer. In its current configuration, the spectrometer has a resolution of $\sim 2 \text{ cm}^{-1}$ with a spectral field of $\sim 350 \text{ cm}^{-1}$. After the output amplifiers have been replaced, we will operate at energies of up to 12 kJ in a 2- to 3-ns pulse at 1 ω . The high intensity and long propagation path may cause significant conversion of the beam to stimulated rotational Raman scattering. The spectrometer will give us valuable information as to whether we are above SRRS threshold.²⁵

Near-Field Camera. We photographed a near-field image of the 3 ω beam on

calibrated film. Multiple first-surface reflections from three wedged beam splitters strongly attenuate the beam before it reaches the film plane. Minor adjustments were required to filter out unwanted wavelengths and reflections and to image the plane of the KDP crystal array.

Frequency Conversion Results.

Figure 3-17 shows the intrinsic third-harmonic conversion efficiency we measured for nominal 5-kJ, 1-ns square pulses. Intrinsic conversion efficiency is defined as 3ω fluence out of KDP/ 1ω fluence into KDP. Also plotted are three theoretical curves that represent various degrees of beam ellipticity ϵ and external angular detuning θ_1, θ_2 for 1-cm-thick doubling and mixing crystals. Error bars (2σ) are shown on two data points. These results are in good agreement with previous experiments performed on Nova.²⁶

Figure 3-18 shows data from the streak camera after correction for its flat-field response. The 1ω pulse measured at FWHM is $\sim 5\%$ greater length than the 3ω . This is due to the low conversion efficiency at the leading and trailing edges of the pulse.

Figure 3-19(a) shows a 3ω near-field photo of the beam, and Fig. 3-19(b) shows a relative intensity vs horizontal position profile. The obscurations in the beam are due to the KDP conversion-crystal mounting structure and a central disk placed in the beam path before both focus lenses to simulate energy losses encountered on Nova.²⁷

▲ Fig. 3-17. The 3ω intrinsic conversion efficiency vs 1ω drive intensity data, and three theoretical curves that have various degrees of crystal detuning and beam ellipticity.

Fig. 3-18. Relative amplitude vs time for (a) 3ω and (b) 1ω flat-field streak camera data.

Fig. 3-19. 3ω near-field photograph, and a relative intensity profile of the Phase II prototype beam.

Summary. The Phase II prototype beam diagnostics station has proven to be an excellent facility for measuring 3ω beam characteristics during the initial activation series. With the station's stable alignment and diagnostics repeatability, we will be able to characterize the third-harmonic performance of the Phase II prototype beamline.

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Demonstration of Shaped 3ω Output Pulses. We completed a series of shots this year aimed at proving that we could generate the desired 3ω -output temporal pulse shapes for certain ICF target designs. Our work also provided an opportunity to fine tune our ability to predict square-pulse distortion via our gain-saturation code and to characterize the performance of beamline 7 before amplifier replacement began. Because the old amplifier glass was still in place, 1ω energies were limited to 6.5 to 7 kJ in 3 ns, which corresponds to 3ω energies in the range from 2 to 2.5 kJ.

The 3ω pulse shape we chose is characterized by a 20:1 front-to-back contrast ratio, as described in detail below. We were able to produce third-harmonic output pulses with shapes very close to that desired.

Figure 3-20(a) shows the attempted 3ω pulse shape, which was the starting point

for determining the required 1ω output [Fig. 3-20(b)] and input [Fig. 3-20(a)] pulse shapes from the master oscillator room (MOR). This figure also shows associated pulses that were generated on our best attempt. The proposed 3ω pulse shape, which was similar to a three-stepped pulse suggested by one LLNL researcher, was temporally flat in each region, but had a t^4 function substituted for the middle step. This approximation was necessitated by the limitations of our current pulse-shaping system. The contrast ratio between the two steps was chosen to be about 20:1.

The process of determining all three of the relevant pulse shapes— 3ω output, 1ω output, and 1ω input—involved several considerations. Concern for the safety of the amplifier glass limited 1ω output power to 4 TW, which would be attained in the second step. The expected conversion efficiency to the third harmonic for that power, corresponding to an intensity of $\sim 1.1 \text{ GW/cm}^2$, determined the 3ω power for that part of the pulse. It also determined the power in the initial step because we had chosen a 20:1 contrast ratio. The t^4 function was then inserted in the intervening region, connecting the first and last steps, and giving the complete pulse shape for 3ω . The 1ω pulse shape was calculated point by point from 3ω intensities using the most recent conversion data on beamline 7 and corresponded closely in the middle section to a t^3 shape. To determine the associated 1ω input pulse

▲ Fig. 3-20. Measured and attempted (a) input, (b) 1ω output, and (c) 3ω output pulse shapes.

Fig. 3-21. Measured 1ω output pulse shape and code prediction based on the input pulse.

shape (at the input-sensor location), we ran our gain saturation code "backwards," with the input to the code being the calculated 1ω output. This determination provided the shape to be generated by the MOR pulse shaper and injected into the amplifier chain.

Figure 3-20(a) includes the MOR streak data, corrected for camera nonuniformities, from a shot that produced 6396 J at 1ω and 2161 J at 3ω [see Figs. 3-20(b) and (c), respectively]. Figure 3-21 gives the 1ω output pulse for that shot along with our code prediction based on a digitized version of the MOR streak. The accuracy of our code predictions is highly dependent on our ability to obtain correct values for transmissions and gains in the laser chain. Hence, we attribute discrepancies between our code predictions and the measured pulse either to incorrect transmission estimates or to diagnostic inaccuracies.

The deviation in amplitude between the desired and measured 3ω pulse

shapes reached values on the order of 20%. This result is partly due to imperfect compensation for square-pulse distortion in the amplifiers, leaving a slope in the portion of the pulse that should be flat. In the fast-rising central portion, the deviation is greater, which is due to a slight temporal offset in overlaying the two pulses. In addition, the ideal pulse has zero rise and fall times, but these times are typically ~ 200 ps for Nova pulses, which use the pulse-shaping system.²⁸ It should be possible to reduce the amplitude deviation in the flatter regions to $<10\%$ with proper compensation for square-pulse distortion.

On most of our shots, we were able to obtain temporal data using two different streak cameras at the output. One measured both the air-path 3ω pulse and the fiber-coupled 1ω ; the other measured the 1ω pulse via air path. At the input, our measurement instruments included both a fast photodiode (160-ps system rise time) and a streak camera. The

combination provided for a well-diagnosed pulse shape.

In conclusion, we have successfully used Nova to produce a third-harmonic pulse shape that is relevant for ICF targets. The 2161-J, 3ω energy was the result of an output 1ω energy of 6396 J. The low conversion efficiency implied is due to the nature of the pulse shape. Indeed, the conversion process helped provide the means to attain the high contrast ratio we wanted. Issues we need to address in the future include the generation of pulse shapes of this type at higher energies, where saturation is greater and B-integral and ellipse rotation effects are more severe.

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Polarization Measurements for Beamline 7. The output Nova beam is nominally p-polarized (i.e., linear, horizontal). This linear constraint arises from polarizers in the master oscillator room and in the Faraday rotator assemblies that are present up to the 31.5-cm amplifiers. Beyond the last 31.5-cm polarizer, only the Brewster-angle amplifier disks attenuate s-polarized energy. However, we previously reported measurements showing that the output Nova beam is, in fact, a mixture of linear and elliptically polarized light.^{29,30}

During our characterization of beam 7 prior to installing the new platinum-free glass, we conducted another series of polarization measurements to verify the polarization state of the 1ω beam before frequency conversion. We are concerned with the polarization state at frequency conversion because depolarization can have severe effects on 3ω conversion, stimulating back conversion and drastically reducing conversion efficiency.³¹

We conducted two main types of polarization measurements. Our first measurements consisted of collecting the energy rejected from a Brewster-angle plate into either a photographic plate or a calorimeter. The photographs were taken to measure the spatial distribution of depolarization; the calorimeter

provided a measure of the level of depolarization. Our second measurement was a 2ω null experiment that used the Type II/Type II crystal array installed on the two-beam chamber to mix depolarized light with the nominally polarized beam to produce 2ω light proportional to the level of depolarization. The latter technique was described in detail in the 1986 Laser Annual.³⁰ We collected the 2ω light into a 74-cm calorimeter located immediately behind the crystal array. An array of BG-18 filter glass was placed in front of the calorimeter to remove 1ω light. We made each type of measurement both before and after changing the output lenses of the 31.5- to 46-cm and the 46- to 74-cm spatial filters from BK-7 glass to fused silica.

Table 3-2 shows the results of both series of measurements. Depolarization (E_s/E_{total}) is similar before and after the change of lenses; the level of depolarization was approximately 4% before the change and approximately 3% afterwards. The value of 4% for depolarization is consistent with previous measurements of beam 7 (Ref. 30).

Our photographs showing spatial distribution of the depolarization were also comparable to previous measurements of beams 9 and 10 (Ref. 29). The depolarization is localized into four lobes, with the central region of the beam essentially free from depolarization. Changing the spatial-filter lenses did little to change this distribution.

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Beam Uniformity.

Uniform Beam Illumination. During our effort to improve the spatial intensity uniformity of the beam input into the Nova beamlines, we investigated the performance of the autoalignment system in the preamplifier section. Because many of the beam masks used for references in pointing and centering did not represent the current beam profile, we installed new masks. This increased the accuracy and repeatability of the beam alignment in this section.

To improve the spatial uniformity of the beam intensity profile, we adjusted the pulse-centering parameters in the autoalignment system to achieve the flattest possible spatial intensity profile at the output of the preamplifier section. The profile at the output of the master oscillator room is Gaussian and 22 mm in diameter at the $1/e^2$ point. We use a 6-mm hard aperture in the preamplifier to clip the Gaussian profile and propagate a relatively uniform intensity spatial profile to the individual Nova beamlines. By using calibrated photographs scanned with a microdensitometer, we were able to adjust the center of the Gaussian intensity peak to the center of the hard aperture. The previous and current beam profiles, shown in Fig. 3-22, reflect the improvement in the uniformity of the aperture illumination. We are looking into methods of monitoring the beam profile on a more regular basis.

Apodization. We studied the propagation characteristics of the Nova beam with the diffraction code MALAPROP. Much of the circularly symmetric modulation in the intensity can be traced to hard apertures at the beam input. Spatial filtering and relaying keep the diffraction under control but leave ripples in the

intensity that can have a peak-to-mean modulation of 1.2:1 to 1.3:1 at some points in the beamline. Since various optics in the beam chain operate at or near the damage threshold on high-power shots, any reduction in this systematic modulation would be a benefit. One way to reduce this modulation is by apodizing the hard apertures. Softening of the transmission profile should minimize the effects of diffraction and reduce the depth of the ripples created by the spatial filtering.

The first step in verifying this hypothesis was to model the preamplifier section of Nova, where the initial hard aperture is located. We wrote a complete model that included all propagation distances for beam 7 and incorporated it into the standard model that describes the laser chain. MALAPROP calculations then

Table 3-2. Measurement of percent depolarization before and after changing spatial-filter lenses.

Time of measurement	Brewster-plate experiment (E_s/E_{total})	2ω null experiment (E_s/E_{total})
Before change	4.0 to 4.2	3.8
	—	3.8
After change	3.1 to 3.2	3.0
	3.8 to 4.0	2.9
	3.3 to 3.4	3.4

Fig. 3-22. Spatial profile of input beam (a) before adjustment and (b) after adjustment.

Fig. 3-23. Contour plot and spatial profile of input beam (a) without apodizing and (b) with apodizing.

produced intensity profiles based on the current system configuration, which we verified by photographs taken on Nova at various points in the beamline. The results were in excellent qualitative agreement. The next step was to repeat the calculations, replacing the pre-amplifier and arm hard apertures with apodizers.

The 6.36-mm-diam preamplifier aperture is the first the spatially Gaussian MOR beam encounters. We replaced this aperture in our model with an apodizer that had an approximately 10th-power supergaussian transmission profile. This removed almost all diffraction structure from the intensity profile as it propagated through the preamplifier. The second aperture encountered is 26 mm in diameter and is located after the splitter array, immediately before the 3.75-cm spatial filter. This beamline aperture

defines the fill factor in the high-extraction portions of the laser chain, so we balanced the amount of apodization with the resulting fill. We chose an apodizing function that has roughly a 50th-power supergaussian profile. Calculations done up to this point have ignored the effect of gain saturation on the steepness of the transmission profile, which should be an issue only in the final amplification stages. We are making additional calculations to investigate the magnitude of this effect.

Figures 3-23 through 3-25 show the apodized and hard-aperture cases at various planes in the laser chain. The radially symmetric intensity modulation is noticeably reduced in the apodized case. The dominant source of modulation then seems to be stochastic noise.

Because these apertures are small and because fill factor is an issue, the

Fig. 3-24. Beam entering final spatial filter (a) without apodizing and (b) with apodizing.

specifications for these apodizers are quite stringent. The transmission gradient is complete over 1 or 2 mm. Our present goal is to produce apodizers with the specified transmission profile. We are pursuing four options regarding the method of apodization:

1. The first option, phase-scattering (i.e., bead-blasted) apodization, is very difficult to control to the degree required here, but we are attempting to produce prototypes.

2. The second option is metallic coatings that will produce the required profile. We are concerned with the damage thresholds of the coatings. The fluence at the arm aperture will be high enough (on the order of 10 mJ/cm^2) for routine high energy shots to preclude metallic coatings.

3. The third option is a variable-thickness neutral-density glass, index-matched

to a complementing blank to produce a transmission profile while ensuring a flat phase front. We are investigating whether this can be done cost effectively using diamond-turning or similar shaping techniques.

4. The fourth option, being investigated in cooperation with the University of Rochester Laboratory for Laser Energetics, uses liquid crystals to create crossed, circular polarizers of varying extinction. This option has the potential for controllable transmission profiles and very high damage thresholds.

Gain Uniformity. Nova amplifier beamlines have a measured gain asymmetry in the horizontal plane of approximately 20% across the beam aperture. This asymmetry is a result of the gain nonuniformity of the individual amplifiers (see "Amplifier Gain Asymmetry Measurements on Nova," later in this

Fig. 3-25. Beam incident on first turning mirror (a) without apodizing and (b) with apodizing.

section) and of the symmetry of their installation in the amplifier beamlines. All disk amplifiers are installed with the apex of the V caused by the Brewster-angle disks pointing towards the space frame. This results in the high-gain side of all the amplifiers of a given size being on the same side of the beam aperture. The inversion between amplification stages caused by the spatial filters compensates somewhat for the gain nonuniformity, but, because the 31.5-cm amplifiers have the greatest asymmetry, the cancellation is not complete. To make this cancellation more complete and ensure a more uniform output beam, we

plan to reverse the last 31.5-cm amplifier in each beamline as the amplifier beamlines are rebuilt.

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Measurement of Third-Harmonic Pulse Shape On Target. In October 1987, we made a series of time-resolved, full-aperture, pulse-shape measurements on Nova. The primary goal of this series was to measure the shape of the third-harmonic pulse at the center of the

target chamber vs that of the 1ω laser output pulse on a series of shots similar to those performed in the x-ray-conversion experiments in the summer of 1987. Using a carefully characterized opposed-port diagnostic (OPD)³² as the principal 3ω diagnostic and the output sensor as the corresponding 1ω diagnostic, we ascertained, on nominally square 1-ns shots having 1ω energies of 3 to 6 kJ, that the shape of the third-harmonic pulse in the center of the chamber varies less than 10% from that of the 1ω pulse as measured in the output sensor. The same is true for longer (up to 4-ns) pulses measured at moderate 1ω intensities.

We made our measurements on beamline 10, the principal beamline used in the x-ray-conversion series, and we investigated shots with 1ω energies of 3, 5, and 6 kJ at 1 ns, 7.4 kJ at 2 ns, 8 kJ at 3 ns, and 9.1 kJ at 4 ns. The oscillator pulses on these shots were shaped to match those used in the target series, but we were not able to reproduce the pulse shapes exactly because of amplifier or flash-lamp replacements, which changed the gain characteristics of the beamline. The shots were propagated to the ten-beam chamber, converted to the third harmonic, and collected in the OPD for measurement.

All 1ω measurements were made in the beamline 10 output sensor. The sensor was equipped with a film-back LLNL streak camera and a fast diode with a Tektronix 7250 scope so that we could make two independent measurements of the 1ω temporal profile. The down-collimated 1ω beam was coupled into

the streak camera by passing it through a lens and diffuser and then into an optical fiber. This system was designed to provide uniform sampling over the entire beam, based on individual measurements of the numerical aperture of the fiber and the scattering characteristics of the lens-diffuser combination. Energy was measured with the output sensor's 2-in. calorimeter.

We measured 3ω pulse shape and energy with both the OPD and the diagnostic spool³³ (the spool-shaped diagnostics package between the KDP frequency-conversion array and the focus lens on each beamline). The OPD was used to down-collimate the 3ω beam and direct it, by air path, to a streak camera equipped with a CCD readout. The beam was input to the camera by passing it through a diffuser placed in front of a lens focused on the camera slit. Because the OPD-streak-camera arrangement was the primary 3ω diagnostic, we tested its aperture-sampling characteristics prior to the experiment. The tests showed a maximum 3% sampling variation across the aperture as a result of transmission through the OPD and a possible 7% additional variation contributed by the streak-camera interface. In addition to the streak camera, a fast diode and a 7250 scope were incorporated at the OPD output. The 3ω energy was measured by integrating the signal from a pin 25 diode located in the OPD, as well as by the 3-in. calorimeter located in the diagnostic spool.

All pulse shapes measured by the streak camera were corrected for

Fig. 3-26. Comparison of laser pulse shapes on 6-kJ, 1-ns shot as measured by (a) streak camera, (b) fast diodes, and (c) a plot of the 1ω streak divided by the 3ω streak.

camera flat-field responses³⁴ and filtered with a 2.8-GHz bandpass filter to approximate the ~ 100 -ps response times of the diode systems. The streak-time bases were calibrated using the measured, average sweep speed of each camera. The diode traces were hand digitized from individual photographs of the 7250 scope displays. Figures 3-26(a) and (b) show the streaks and fast-diode traces obtained on the 6-kJ, 1-ns shot.

The streak-camera and fast-diode data correlated well and showed little difference between 1ω and 3ω pulse shapes for all shots measured. This is demonstrated in Fig. 3-26(a) and (b), where the 1ω and 3ω pulse shapes from the 6-kJ, 1-ns shot have been normalized in intensity and carefully overlaid. If the 3ω streak is divided into the 1ω streak and the results are plotted [Fig. 3-26(c)], the difference in pulse shape can be quantified as less than 10%.

From Fig. 3-26 and inspection of all available data, we conclude, from the shots measured, that the 3ω and 1ω pulse shapes differ by no more than 10% over most of the pulse, although variations greater than 10% may occur at the pulse's leading or trailing edges. It is difficult to make conclusions about absolute 3ω power because the OPD energy measurement suffered a large systematic bias and the diagnostic-spool measurement is historically unreliable. The 3ω energy on target was estimated from the measured 1ω energy and a system frequency-conversion efficiency curve calibrated for beamline 10 (Ref. 35). The 3ω energy calculated in this manner and that measured by the diagnostic spool are equivalent to within 20%. Until the diagnostic spools are reconfigured for greater reliability, however, we recommend that the output-sensor 1ω energy measurement be used in conjunction with the system conversion curve to infer the 3ω energy on target. Third-harmonic power for target shots with 1ω parameters similar to those we have measured can be derived by scaling the 1ω pulse shape measured in the output sensor

with the corresponding ratio of 3ω to 1ω energies.

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Damage Threshold of Large Polarizers.

One proposed scheme for increasing Nova's third-harmonic conversion efficiency calls for repolarizing the 1ω output with a large-aperture polarizer, the major liability of which will be its damage threshold. Before beginning our experiment, we had planned to use the same combination of coating and substrate that is used on the 31.5-cm polarizers in the final Faraday rotators on Nova. Therefore, it was important to test this coating ($\text{TiO}_2/\text{SiO}_2$ layers) to determine its threshold damage fluence. We first used a small-aperture test facility and then moved to the experimental station on beamline 9 of Nova, near mid-chain. We observed damage on the polarizer tested (S/N 29) within a few shots at average fluences of $\approx 4 \text{ J/cm}^2$ at 2 ns, as measured with a beam having a peak-to-average (P/A) ratio of about 1.3 to 1.

Small-Aperture Tests. There is a small discrepancy in our conclusions from the small-aperture (2-mm-diam) and larger-aperture (8-cm-diam) tests. On the basis of the small-aperture tests alone, we concluded that the sample would exhibit damage within a few shots at fluences $> 5 \text{ J/cm}^2$ in 2 ns but should withstand irradiation at 4 to 4.5 J/cm^2 . We observed a multiple-shot effect that either reduced the damage threshold or at least made the damage visually apparent and concluded that multiple-shot testing is needed for polarizers.

We do single-shot testing to estimate the single-shot threshold for large components. Then we attempt to confirm this threshold by irradiating other test sites multiple times at a fluence of 80 to 90% of the apparent threshold. For this polarizer, the upper limit was set by a failure at 5.1 J/cm^2 that was observed in a 2.5-ns multiple-shot test. The damage induced

Fig. 3-27. (a) Beam used for large-aperture tests and (b) associated lineout.

in that test was slight, however, and did not grow under repeated irradiation at 4 to 5 J/cm², which makes us believe it was threshold-level damage. We can state with certainty that the polarizer cannot be used at 6 J/cm² since that fluence caused damage in all single-shot, 1-ns tests and in multiple-shot tests at either 1 or 2.5 ns.

Large-Aperture Results. A mirror placed between the two mid-chain turning mirrors on arm 9 of Nova directed the beam to our experimental setup. The beam was down-collimated to a diameter of 11.5 cm, and an 8.2-cm-diam aperture was centered on the beam to remove hot spots in the outer portion. Figure 3-27 shows the beam at a position corresponding to ~2 m after the polarizer plane and the associated horizontal scan. The P/A ratio for several different shots was about 1.3 to 1 and is slightly higher if the diffraction from the aperture is included.

Coating damage was easily visible after two 2-ns shots at average fluences of 5.44 and 5.18 J/cm². This damage appeared on the surface in the form of many small scattering sites per square centimeter (as seen by backlighting the optic with a white light source), but no damage was observed in the bulk during this series. When the beam was positioned at a new location on the polarizer, damage was again visible (although not as heavy) after four shots ranging in

average fluence from 3.67 to 3.95 J/cm². While an average fluence of 3.95 J/cm² implies a peak of about 5.1 J/cm², the damage was over much of the beam area, suggesting that the actual multiple-shot threshold at 2 ns is closer to 4.5 J/cm²; this threshold probably makes the current coating unacceptable for use on full-aperture output polarizers for Nova.

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Stimulated Rotational Raman Scattering in Long Air Paths. This year we studied many unusual and fundamentally important diffraction and spectral phenomena associated with stimulated rotational Raman scattering (SRRS) of intense 1ω light arising from atmospheric nitrogen in long propagation paths. The spatial, temporal, and spectral dynamics of SRRS are important to understand as applied to large-aperture, high-intensity beams for both fusion research^{36,37} and national defense applications³⁸ because the occurrence of SRRS in propagation paths significantly affects beam transport and focusability.^{39,40} In addition, the SRRS effect might be of use in the generation of superbroadband, spatially incoherent light for target-irradiation experiments.^{39,40}

1. Near-field camera
2. Streaked spectrometer (time-resolved spectrum)
3. Near-field calorimeter
4. 1-m spectrometer (time-integrated spectrum)
5. Far-field calorimeter
6. 1-m spectrometer (spatially resolved far-field spectrum)
7. Multiple-image, far-field camera
8. Multiple-image, far-field Schlieren camera

Fig. 3-28. Layout of SRRS diagnostics station.

We have obtained evidence (for the first time, to our knowledge) of the suppression of SRRS in a collimated beam as a result of the interplay of four-wave parametric mixing with SRRS. This phenomenon, referred to as parametric gain suppression,^{41,42} has previously been observed only with focused beams.⁴³⁻⁴⁵ We also observed anomalous combinations of low- J rotational, anti-Stokes, and Stokes-shifted lines at low intensities. We are currently analyzing these results. In addition, we obtained high-quality time-resolved spectra of the scattered Raman light showing the transient build-up of the first anti-Stokes and higher-order Stokes lines.

We conducted our experiments using beamline 7 of the Nova laser at a wavelength of $1.053 \mu\text{m}$. The 70-cm-diam Nova beam was propagated 33.5 m through the Nova switchyard to a 23-cm-diam aperture. A spatially smooth region of the beam was apodized with a feathered-edge apodizing plate and then down-collimated to an 11.5-cm diameter with a 2-to-1 telescope. The beam then propagated 67 m through beam tubes to a diagnostics station (see Fig. 3-28) located beyond the two-beam target bay.

We obtained time-integrated and time-resolved spectra, near- and far-field beam photographs, and a few spectrally resolved far-field measurements at intensity times path-length (IL) products from 13 to 25 TW/cm with 1- and 2-ns square

pulses and 100-ps Gaussian pulses. A telescope with an effective focal length of 13.38 m focused the beam through a 5-mm hole in a Schlieren mirror placed 21 cm ahead of focus. The divergent Raman-scattered light was brought to focus by the mirror and re-imaged by a multiple-image camera onto Tri-X film. A wedged beam splitter was placed ahead of the Schlieren mirror, sending 4% of the entire beam to focus in the plane of a second multiple-image camera. The entrance slit of a 1-m spectrometer was placed in a third, equivalent focal plane and provided a spectrally resolved far-field image on a few shots. In addition, an LLNL film-backed streak camera was close coupled to a 1-m spectrometer to provide high-quality, time-resolved spectra. Time-integrated spectra were recorded by a third 1-m spectrometer, and measurements of energy and near-field beam irradiance were made at various locations.

As we previously reported, strong SRRS in large-aperture beams with high Fresnel numbers generally results in near-field beam breakup and the occurrence of discrete, high-intensity speckles that diverge rapidly out of the beam.^{36,37} The far-field irradiance is also highly speckled and shows a divergence comparable to the geometric angle of the pump beam.^{39,40} In the far-field experiments reported here, this scattering is absent in a narrow cone surrounding the

forward direction and appears as highly speckled, structured rings of Stokes-shifted light that radiate at angles of ~ 250 to $750 \mu\text{rad}$ to the unscattered beam direction. A spectrally resolved, far-field image (not shown here) confirmed that the scattered light is predominately Stokes shifted.

Figure 3-29 shows a set of far-field images for a shot at $\sim 1.6 \times$ above stimulated threshold ($\overline{I} \approx 24.7 \text{ TW/cm}$). On this shot, we observed four first-Stokes lines containing $\sim 30.6\%$ of the transmitted energy and three second-Stokes lines with $\sim 11.3\%$ of the energy. The images on the left-hand side of this figure show the entire beam at best focus; the images on the right-hand side show the divergent Raman light.

Figure 3-30 shows horizontal and vertical intensity profiles of the scattered light. The horizontal compression of the far-field distribution is probably due to astigmatism in the Schlieren camera rather than to SRRS. We believe that the bright central spot is an artifact of pump light intercepted and focused by the Schlieren mirror. Just above SRRS threshold, the scattered light appears in the far field as vertical filamentary bands surrounding the central spot. As the $\overline{I}$ product increases, the profile becomes increasingly speckled, and a dense vertical zone of scattering develops along the horizontal axis (see Fig. 3-29). This bright zone might be a polarization effect, since the pump beam was horizontally polarized. Raman light is also observed in the pump-beam far-field images (Fig. 3-29 top, left) as a speckled ring with a diameter $\sim 3.6 \times$ the pump-beam diameter. The empty region between the central spot and the scattered light defines the perimeter of the gain-suppressed, far-field region ($\sim 200 \mu\text{rad}$).

In recent experiments, we have also observed unusual diffraction phenomena in the near field, as shown by the sequence of photographs in Fig. 3-31. Below SRRS threshold, the pump beam shows the expected circular Fresnel structure, some central-beam modulation, and fine horizontal interference fringes caused by the antireflection coating on

(a) Entire beam

(b) Raman scattered beam

the apodizer. Somewhat above SRRS threshold for a 1.2-ns pulse ($\overline{I} \approx 20.4 \text{ TW/cm}$), the SRRS appears as radial filaments along the perimeter of the beam, orthogonal to the outer one or two Fresnel rings [see Fig. 3-31(a)]. At a slightly higher intensity, $\overline{I} \approx 21.0 \text{ TW/cm}$, discrete speckling develops in the center of the beam; this phenomenon appears to be correlated with the fine horizontal diffraction structure and with the inner Fresnel rings [see Fig. 3-31(b)]. In addition, the outer filaments show some breakup into discrete speckles. The appearance of discrete speckles corresponds to the occurrence of second

Fig. 3-29. Far-field irradiance of (a) entire beam and (b) Raman scattered light at $\overline{I} \approx 24.7 \text{ TW/cm}$ for 1.2-ns, square laser pulse. Multiple images decrease a factor of 4 in intensity from image to image, top to bottom. (a) is attenuated by a factor of 10 with respect to (b).

Fig. 3-30. (a) Horizontal and (b) vertical intensity profiles of Raman scattered light in the far field. Central bright peak is an artifact from scattered pump light.

Fig. 3-31. Photographic sequence of near-field images for 1.2-ns pulses. (a) Above SRRS threshold at $\bar{I} \approx 20.4$ TW/cm. (b) Above SRRS threshold at $\bar{I} \approx 21.0$ TW/cm. (c) Far above SRRS threshold at $\bar{I} \approx 24.7$ TW/cm.

Stokes in the spectrum at $\sim 1.3 \times$ threshold for first-Stokes scattering. At $\bar{I} \approx 24.7$ TW/cm, the beam shows speckles outside the perimeter of the original beam, and both ordered and disordered discrete speckles develop throughout the beam interior [see Fig. 3-31(c)].

We have written a code for plane-wave propagation to model the steady-state SRRS/four-wave mixing interaction for beams with high Fresnel number. The code includes off-axis first-Stokes, second-Stokes, and first-anti-Stokes waves. The multiple first-Stokes rotational transitions are treated as a single transition because of the complexity of wave coupling, and the wave polarizations are assumed to be linear. Integration of the complex, coupled-wave equations is carried out with pump-wave depletion in the high conversion regime. The four-wave terms from the nonresonant optical Kerr effect and self-phase modulation are included, as is the

detuning of the first-Stokes wave from resonance (i.e., the Raman-induced Kerr effect).

The plane-wave model gives results that are in good qualitative agreement with our observations. Figure 3-32 shows results for parameters that are close to experimental conditions. Figure 3-32(a) shows the effect of SRRS gain suppression and an unexpected effect of gain "enhancement." Here we plot the ratio of the first-Stokes gain to the ideal plane-wave gain ($g_{\text{gain}}/g_{\text{L}}L$) vs the first-Stokes angle from the forward pump beam. In rotational scattering, the second Stokes radiates at close to twice the first-Stokes angle, and the first anti-Stokes radiates at an angle nearly equal to, but opposite in sign to, the first Stokes. The four-wave coupling is assumed to be planar so that momentum mismatch is minimized. At first-Stokes angles of less than $250 \mu\text{rad}$, the Stokes and anti-Stokes waves are parametrically coupled to the pump, and

Fig. 3-32. (a) Plane-wave calculations of SRRS gain and (b) and (c) converted energy fraction under conditions of parametric gain suppression and enhancement vs first-Stokes scattering angle from the forward direction.

the stimulated Stokes gain is suppressed. Beyond 250 μrad , the stimulated gain is increased by four-wave mixing and exceeds the plane-wave gain by 12%. This effect requires that the Stokes wave be blue-shifted off resonance by one-half the Raman half-width at half-maximum (HWHM) spontaneous line width.

Figure 3-32(b) plots the resulting energy-conversion curves on a logarithmic scale as a function of first-Stokes angle. Stimulated threshold (i.e., 1% conversion) is exceeded at 250 μrad . Figure 3-32(c) replots the curves from Fig. 3-32(b) on a linear scale. This figure shows the large first-Stokes conversion peak at 375 μrad and a rapid dropoff beyond 750 μrad . These predictions are in

qualitative agreement with observations, except that the energy balance between first- and second-Stokes lines disagrees with measurements of spectral energy.

The spatial dynamics of SRRS may be understood by considering the interplay of parametric gain suppression and diffraction. Diffraction and high spatial-frequency modulation at the edge of the beam correspond to plane waves in the Fourier-transform domain or rays that propagate off-angle to the forward direction. These high-angle rays are less gain-suppressed, and even enhanced [Fig. 3-32(a)], and therefore exceed first-Stokes threshold at lower intensity compared to on-axis rays. In addition, small-scale diffraction structure on the beam

from obscurations (such as optics damage and dust) will see higher stimulated gain due to high-angle plane-wave components and increased intensity modulations from linear diffraction.

We surmise that the filamentary structure of SRRS is initially radial because the four-wave parametric interactions that enhance the first- and second-Stokes scattering at the edge of the beam occur in phase-matching planes that are oriented radially. Small-scale diffraction and intensity modulation, highest around the edge of the beam, serve to "seed" the radial filament growth. At high intensity with strong second-Stokes scattering, the filaments break up into small-scale structures

that can then propagate outside the perimeter of the beam at angles approximately twice the first-Stokes angles. We are adding Fresnel diffraction and Raman-enhanced nonlinear diffraction in one transverse dimension to our SRRS propagation code. Our preliminary results substantially verify our interpretations.

Our recent SRRS spectra have shown several interesting low- J rotational anomalies that are as yet unexplained. Figures 3-33(a) and (b) show spectral observations as a function of $\overline{IL}$ for 1.2- and 2.2-ns pulses, respectively. As previously reported, the S(8) and S(10) Stokes lines in nitrogen have the lowest threshold (15.0 TW/cm) for a 1.2-ns pulse. However, slightly above threshold (15.1 TW/cm), the O(1) anti-Stokes line arises in conjunction with the S(8) and S(10) lines. This is an unexpected observation, particularly since the odd- J -rotational lines have not previously been observed in SRRS in air except at high intensity (>28 TW/cm).^{39,40} However, we are currently unable to assign any other transition to this 20-cm^{-1} shift. At $\overline{IL} \approx 15$ TW/cm, these lines are each only a fraction of one percent of the pump intensity. At higher intensity, the S(1) line appears, and the O(1) line disappears. In addition, at 1 ns, we observe the so-called 8B lines that were first observed by Averbakh et al., concurrent with the appearance of the second-Stokes lines (~ 19.2 TW/cm).^{46,47} These are Stokes-anti-Stokes line pairs generated by forced light scattering from transient Raman gratings. They are spaced at multiples of the 8B frequency (16 cm^{-1}) from the pump and correspond to the beat frequencies between adjacent rotational excitations.

For a 2.2-ns pulse, the O(1) anti-Stokes line is the first line to appear (at ~ 12.8 TW/cm). At higher intensity (~ 13.4 TW/cm), the S(8) and S(10) lines appear as expected, and the O(1) line is not observed again until much higher intensity (~ 17 TW/cm). Several theories have been proposed in the literature for understanding the spectral dynamics of Stokes⁴⁸ and anti-Stokes⁴⁹ rotational scattering; the theories do not, however, explain the $J = 1$ Stokes and anti-Stokes lines that appear in our spectra at

Fig. 3-33. Time-integrated spectral observations for (a) 1.2-ns pulse and (b) 2.2-ns pulse vs $\overline{IL}$ product.

moderate intensity. In addition, we observed both the O(1) and S(1) lines simultaneously for a 100-ps shot, far below the expected SRRS threshold. This observation indicates that another nonlinear process, such as SRS in the phosphate laser glass, could be responsible for the $\pm 20\text{-cm}^{-1}$ shift.

Figure 3-34 shows the temporal dynamics of SRRS in a streaked spectrum of a 1.2-ns square pulse with $\overline{I} \approx 24.7$ TW/cm. The first-Stokes lines S(8) and S(10) turn on at ~ 200 ps into the pulse and are very pronounced. The S(6) and S(12) lines trail slightly in time behind the S(8) and S(10) lines. At a time 100 ps later, the strong second-Stokes lines S(8) + S(10) and S(10) + S(10) appear. With the strong second-Stokes lines, we also see the start of weaker first-Stokes, first-anti-Stokes, and 8B lines. Later in time, the weaker second-Stokes and some third-Stokes lines appear. Close examination shows that the O(1) line might have some intensity near the start of the pump pulse, but the evidence is not conclusive because of background noise in the streak image.

We have described several of the significant, new phenomena observed in our latest experiments. Our future work will be directed toward the possibility of using the broadband, spatial-incoherence properties of SRRS in laser fusion research.

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Major Contributors: B. D. Cline
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Small-Aperture, Fourth-Harmonic Conversion. We continued our experiments on fourth-harmonic generation (4HG) during 1987, using an improved laser system: capable of generating up to 8 GW/cm^2 in 150-ps, Gaussian pulses at the fundamental ($1.053\text{-}\mu\text{m}$) wavelength. We apertured the output Gaussian beam to 1.7 mm to obtain an approximately flat-topped spatial profile. We achieved conversion to the second harmonic (2ω) in a separate Type II, 1.0-cm-thick KDP crystal. With this system, we achieved 80% internal 2ω -to- 4ω conversion efficiency in a Type I, 1.0-cm-thick KDP

Fig. 3-34. Time-resolved spectrum of a 1.2-ns square pulse showing transient evolution of first, second, and third Stokes and first anti-Stokes scattering ($\overline{I} \approx 24.7$ TW/cm).

crystal. We believe this value is the highest 2ω -to- 4ω conversion efficiency obtained to date in KDP and attribute our results to careful aperturing of the beam.

A computer code, which uses the plane-wave approach and incorporates previously established values for the nonlinear and two-photon absorption coefficients (β) for 4HG was written to compare with our measurements (see Fig. 3-35). The code slightly underestimates the peak conversion efficiency and also predicts a different slope for the curve after the peak. We believe that the discrepancy lies in an incomplete understanding of the nonlinear absorption at the fourth harmonic and in incorrect values of the relevant nonlinear coefficients.

We also observed an anomaly in the 4ω pulse shape (see Fig. 3-36), which may be associated with the above discrepancy. The 2ω pulse incident on the Type I quadrupler is approximately Gaussian with a full width at half maximum (FWHM) of 120 ps and a peak intensity of 3.8 GW/cm^2 . The resultant 4ω pulse is asymmetric, with its leading shoulder more intense than the trailing shoulder. The shape of the residual 2ω pulse transmitted through the quadrupler is also asymmetric, but reversed in time. We believe this asymmetry is due to phase modulation on the fundamental pulse.⁵⁰ Because the dispersion is much greater at the fourth harmonic, even a small amount of phase modulation on

Fig. 3-35. Measured, internal conversion efficiency and ratio of transmitted-to-incident energy along with code calculations for a 1.0-cm-thick KDP crystal detuned 100 μ rad.

Fig. 3-36. (a) 2ω and (b) 4ω pulse shapes obtained with a 1.0-cm quadrupler at 3.8-GW/cm² drive.

the fundamental pulse can change the wave-vector matching condition over its duration. This effect could manifest itself in different conversion efficiencies for the same drive intensity, producing converted pulses.

Another interesting observation in our experiment was the appearance of moderate, small-scale beam breakup at the fourth harmonic when none occurred in the incident 2ω beam. When we detuned the quadrupling crystal and propagated a 4-GW/cm², 2ω pulse through it, we saw no beam breakup at 2ω . However, with the crystal tuned, the 4ω beam showed moderate to heavy beam breakup at the same level of 2ω drive (see Fig. 3-37). This observation implies a higher value

for n_2 at 0.26 μ m than at either 0.53 or 1.05 μ m, but we have not yet quantified the value. We are pursuing experiments to investigate these effects more closely.

Authors: P. H. Chaffee
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Pulse Compression. Several Nova applications require that a short (~ 10 ps) pulse be generated by the master oscillator. To support these applications, we are developing a pulse-compression system to be installed in the master oscillator room (MOR). A prototype of this system has already been built and tested in the pulse-shaping lab. In the paragraphs that follow, we discuss our goals

Fig. 3-37. Beams propagated 120 cm after quadrupler. (a) and (b) 2ω beams at 4 GW/cm^2 with quadrupler detuned and tuned, respectively. (c) 4ω beam at 0.7 GW/cm^2 , 2ω drive (peak of conversion curve). (d) 4ω beam at 4.0 GW/cm^2 , 2ω drive.

in building this pulse-compression system and describe the prototype system now in use.

Experimental System. The general approach to producing a short pulse is to select a pulse from the short-pulse oscillator train,⁵¹ then compress and amplify it to produce a pulse of the required length. A fiber-grating compressor is used to compress the pulse, and the scheme used to amplify it is similar in principle to that used by Mourou et al.,⁵² but with a different set of parameters. The pulse compressor has essentially no temporal jitter from input to output. Since the starting pulse is part of the same pulse train that drives the pulse shaper and picket-fence (multiple-short-pulse) generator, the compressed pulse will be well synchronized with the main (shaped) laser pulse.

When the compressed pulse is used for backlighting, it must follow the main pulse by 20 ns. The uncompressed pulse must be selected from the high-amplitude portion of the pulse train, near the other selected pulses, so that enough energy is provided to run a pulse-energy regulator (described below) in order to minimize fluctuations in pulse energy. The pulse-compression system itself must have a minimum delay from input to output so that the input pulse does not have to be selected too early. This consideration limits certain aspects of the system design, such as fiber length, input energy, and amplifier configuration.

A high-energy, visible wavelength pulse is required to drive the high-resolution framing camera. The pulse-compression system will amplify the pulse and double its frequency; then it will be propagated along a separate beamline to the target chamber area. After it has been frequency converted, the compressed pulse must have enough energy to drive several cameras, a task that will require tens of millijoules of 1ω energy.

Prototype System. The layout of the prototype pulse-compression system is shown in Fig. 3-38. Here a 100-ps pulse selected from a Nd:YLF short-pulse oscillator is fed into the prototype system whose main components are a single-mode optical fiber, a multipass amplifier, and a pair of diffraction gratings that compress the pulse after it has been amplified.

To minimize input-output delay, the optical fiber must be short and the number of times the pulse passes through the amplifier must be small. This means the energy in the fiber will be as high as possible, limited by bulk damage or Raman scattering. After trying various fiber lengths, we chose a 10-m length of $5\text{-}\mu\text{m}$ core fiber whose Raman threshold is about equal to its damage threshold. This 10-m length provides sufficient chirp bandwidth and energy throughput with an input energy of about 100 nJ. If longer fibers are used, Raman scattering occurs at lower energies and limits the amount

Fig. 3-38. Experimental prototype pulse-compression system where oscillator-produced pulses are regulated, chirped in a fiber, amplified in a multipass amplifier, compressed by diffraction gratings, frequency converted, and analyzed by a streaked spectrometer.

of unscattered light that is passed through the fiber.

The resulting bandwidth is approximately 10 Å, but the chirp is nonlinear in time, so only a fraction of the bandwidth is optimally compressed by the linearly dispersive diffraction gratings. The output pulse has a FWHM of 10 to 12 ps, with low-level "wings." To observe the temporal and spectral characteristics of the chirped and compressed pulses, we added a 0.5-m spectrometer to our streak camera to obtain streaked spectrographic

data. Figure 3-39 shows the streak output for an uncompressed chirped pulse and a compressed pulse.

The wavelengths near the band edges represent the least linear portion of the chirp and contribute only to the wings of the compressed pulse. Thus, it is possible to eliminate the wings by spectral filtering. A pair of knife edges in the middle of the grating beam path intercept the extrema of the spectrum and cut the unwanted energy from the front and back edges of the pulse. Figure 3-40 shows compressed pulses with and without edge filtering.

Pulse width and shape are sensitive functions of the input intensity in the fiber. Because normal energy fluctuations in the actively mode-locked and Q-switched (AMQ) oscillator are too large (several percent), we developed a separate device to regulate energy. This regulating device consists of a photoconductive switch circuit that drives a Pockels cell. When the input beam enters the system, it is split to both illuminate the switch and pass through the cell. As energy increases, the cell decreases transmission and vice versa. Tests show the input energy can vary by almost 30% while the output energy remains constant to within $\pm 1\%$. When used on the compression system, this regulator has given excellent stability to the output pulse.

Operational stability is also enhanced by the use of remotely controlled motorized positioners on the input-end mount for the optical fiber. This mount must be regularly aligned to within $1\ \mu\text{m}$ for maximum transmission. Manual positioners make accurate alignment difficult because they must be handled during adjustment, causing unwanted shifts in position. A cover must also be replaced after alignment, further reducing stability. With remotely controlled positioners, however, we have observed a stable transmission of $> 50\%$ through the fiber for several days.

After it passes through the regulator and fiber, the beam is expanded and enters the multipass amplifier—a 10-mm glass amplifier (maximum gain 30) placed between two flat mirrors. A

Fig. 3-39. Spectrally dispersed streaks of a chirped pulse (left) and a compressed pulse that has gone through a grating pair having positive linear dispersion with wavelength. Time increases top to bottom, wavelength increases left to right.

Fig. 3-40. Sireak lineouts of compressed pulses with-out spectral filtering (left) and with spectral "wing" filters inserted. The two knife edges independently remove the wings before and after the compressed pulse.

Pockels cell rotates the polarization of the beam to trap the pulse in the mirror cavity for the duration of the voltage pulse on the cell. The pulse then passes through the amplifier six times for a maximum gain of one million and is ejected by the cell. At high gain, energy leakage from the cavity (via the input polarizer) results in high-energy pulses being sent back to the fiber, causing fiber damage. To prevent this, we have inserted a permanent-magnet Faraday isolator after the fiber. Another problem with this configuration is thermal lensing in the rod, which contributes to misalignment. The multipass amplifier can be aligned at one gain but be misaligned at another because of beam steering in the amplifier glass. This steering also

produces variations in beam size, sometimes causing clipping. For this reason, the system designed for the MOR uses two separate amplifiers.

The diffraction gratings are commercial optics designed for our type of application, with 1700 lines/mm and gold metal coating. They are in a double-pass configuration, so the output beam is round rather than oval as it would be in a single-pass configuration. The diffraction efficiency per bounce is almost 90%, resulting in more than 60% total transmission. Similar gratings have been damage tested to 100 mJ/cm^2 , with a safe operating range at 20 mJ/cm^2 . Our system is well within this range.

We tested the system up to 2 mJ compressed output and noticed no change in

pulse shape due to amplification. The pulse was frequency converted (35% maximum conversion efficiency without wing filtering) in a 1-cm-thick KDP doubling crystal. The conversion efficiency was low overall because of the substantial amount of energy in the wings, which were conveniently lost during conversion as a result of low efficiency at low intensities. Had the wing filter been used, an apparent loss in energy efficiency would have appeared in the grating stage, but there would have been no significant reduction in peak pulse amplitude or peak conversion efficiency. Even when the wings were present at 1ω , they were effectively rejected by the conversion process, resulting in small pre- and post-pulse amplitudes, as observed on the streak camera. At 600 μJ of 2ω energy, the pulse width was about 11 ps, close to the 1ω pulse width.

The design of the pulse-compression system to be installed in Nova is almost complete. Besides its technical requirements, it will be reliable and easy to operate. And, it will differ from the prototype system in two respects: amplification will be done in two stages (a four-pass stage and a two-pass stage), and the grating compressor will handle a larger beam and higher energies.

Author: R. B. Wilcox

Amplifier Development for the Nova Laser

Introduction

We have limited the output energy of the Nova laser system for routine operations to less than one-half of its design capability since it was first operational in 1985. Microscopic platinum inclusions ($\sim 10\ \mu\text{m}$) in the laser glass, inadvertently introduced during glass production, limited output fluences to below the damage fluence of the platinum inclusions. Eliminating this output limitation required replacing all the Nova large-aperture laser disks with platinum-free

glass. Thus, our major effort with respect to amplifier development this past year has been to retrofit Nova with platinum-free laser disks.

The 1985 and 1986 Laser Annuals described our successful research and development activities to eliminate platinum from the laser glass. These efforts resulted in new melting processes and new facilities at our two glass suppliers. Production melting of the replacement disks began in December of 1986 and will continue into 1988. The following article, "Replacement of the Nova Laser Disks," gives the production status as of the end of 1986.

A second major effort associated with the Nova replacement disks has been the development of a new edge cladding for the disks. Our new edge cladding uses an optical epoxy to bond the cladding glass to the laser disks, and it substantially reduces the production costs of the clad disks. The article, "Composite Polymer/Glass Edge Cladding," describes solutions for the last of the development problems for the cladding and the completion of its integration into production at our two disk-finishing vendors. The article, "Amplifier Gain Asymmetry Measurements on Nova," later in this section, describes Nova and off-line experiments to quantify and improve the uniformity of the Nova output beams.

Author: J. E. Murray

Replacement of the Nova Laser Disks

In the 1986 Laser Annual, we described the completion of research and development necessary for preparation of essentially platinum-inclusion-free phosphate laser glass.⁵³ During that year, we completed pilot melting runs producing about 40 laser disks to demonstrate this new technology. On the basis of our successful results, we decided to proceed with full production of replacement laser glass for Nova.

Production began in December, 1986, and has continued throughout 1987. Over 90% of the replacement laser disks have been produced and are in the process of

being clad with the newly developed composite polymer/glass cladding.⁵⁴⁻⁵⁶ Development of the cladding process is described in more detail in the next article. Here, we summarize the results from platinum damage tests on the new laser glass.

As mentioned in the 1986 Laser Annual, we are using a laser-damage test system as part of the glass production process to scan each of the large glass castings for the presence of damage-causing inclusions. A damage test system is located at each of the two glass vendors (Schott Glass Technologies, Inc., and Hoya Corporation). Details of the systems and their operation are given elsewhere.⁵⁷ In brief, the damage test systems are designed to scan an entire glass casting, using the pulsed output from a commercial neodymium-doped yttrium-aluminum-garnet (Nd:YAG) laser. The system is fully automated and operates unattended.

Following testing, each glass casting is removed from the system and visually inspected for the presence of platinum-damage sites. We routinely test polygon-shaped castings that are about 0.5 m in size, weigh approximately 30 kg, and contain about 7 L of glass. Roughly 6 to 8 hr are required to test a piece of this size. To date, the systems have been in use about 12 hr/day, up to 5 days/week for about 15 months.

Table 3-3 summarizes the platinum-damage test specifications each glass casting must pass. Our original manufacturing goal was to have no more than an average of 2 inclusions per disk. Each disk contains about 7 L of glass. We have easily surpassed this goal; the average number of inclusions per disk is less than 1 (i.e., ~ 0.5). This result is about a thousandfold improvement over the original Nova disks (the platinum-inclusion density in original Nova disks ranged from about 100 to 10 000/disk—see Fig. 3-41).

A few, small damage inclusions do not necessarily render the laser disk unusable. We can accept a certain number of damaging inclusions in each piece of glass as long as the damage size remains below the limits given in Table 3-3.

Table 3-3. Platinum-inclusion damage tests for replacement of Nova 46- and 31.5-cm glass laser disks.

Parameter	Results
Number of inclusions	<ul style="list-style-type: none"> • No more than 10 metallic particles or other types of inclusions within the clear aperture volume (volume ≈ 7 L)
Minimum distance between inclusions	<ul style="list-style-type: none"> • 25 mm
Average number of inclusions (manufacturing goal)	<ul style="list-style-type: none"> • No more than 2/disk (< 0.3 inclusions/L)
Inclusions after damage testing	<ul style="list-style-type: none"> • One damage spot ≤ 0.5 mm (largest dimension) • Remainder ≤ 0.25 mm (largest dimension)
Damage test conditions	<ul style="list-style-type: none"> • Wavelength = 1053 to 1064 nm • Fluence $> 7 \pm 1$ J/cm² • Pulse width = 8 ns • Number of shots = 100

Fig. 3-41. Platinum-inclusion densities (inclusions/L) in old and new Nova laser glass. Each Nova disk contains ~ 7 L of glass; new disks contain an average of ~ 0.5 inclusions per disk.

Fig. 3-42. Statistical distribution of platinum inclusions in Nova replacement laser disks. Over 60% of the disks have no platinum inclusions.

Figure 3-42 summarizes the statistical distribution of inclusions found in acceptable laser disks. These data represent the combined results from both vendors at approximately 90% completion of the production contract (i.e., approximately 250 laser disks produced). The results show that over 60% of the laser disks have no platinum inclusions at all, and 90% have 2 or fewer. Such results far surpass our expectations and highlight the significant progress made in producing damage-resistant laser glass.

Author: J. H. Campbell

Composite Polymer/Glass Edge Cladding

Overview. Disk amplifiers require edge claddings on the periphery of their disks to minimize the loss of stored energy to amplified spontaneous emission (ASE). Most of the spontaneous emission from stored energy in the disk undergoes total internal reflection at disk surfaces, which traps it between the parallel faces of the disk. As spontaneous emission propagates, reflecting from the two faces, it also undergoes amplification until it reaches the disk edges. This first-pass ASE gives rise to a significant, but unavoidable, reduction of gain and storage efficiency in large-disk amplifiers. The purpose of the edge cladding is to absorb enough ASE at the disk edges to minimize additional loss from more than one ASE pass through the disk.

About two years ago, we began the development of a new type of edge cladding^{58,59} for the platinum-free glass disks that are now being retrofitted on Nova. The original Nova claddings consisted of index-matched absorbing glass fused directly to the laser disks at a temperature near the melting point of the glass. Although the claddings were successful technically, they were difficult to produce and, therefore, expensive. A yield problem resulting from the high-temperature processing was a major reason for their expense; we estimate that the claddings contributed between one-third and one-half of the cost of the prefinished disks.

As described in the 1986 Laser Annual,⁵⁹ our new, composite polymer/glass cladding uses a specially formulated optical epoxy to cement the absorbing glass to the disks, thereby avoiding high-temperature processing. The cost of new claddings will be less than one-third that of the fused-glass cladding, which will result in production savings of over \$2 million for the total disk procurement. Moreover, we should be able to use the new type of cladding on future laser systems.

The articles in this section describe the successful completion of our composite polymer/glass cladding design and its successful integration into commercial production. Our work during 1987 is the culmination of a large, collaborative effort by researchers from several technical disciplines, as described in the 1986 Laser Annual.⁵⁹

Our first objective at the beginning of the year was to solve the remaining damage problem for the new cladding and to demonstrate its long-term survivability under Nova operating conditions. At the end of last year, we had demonstrated that tilting the disk sides eliminates a rapid and catastrophic cladding failure from surface parasitic modes; however, we had also discovered a second type of cladding failure. Cladding delaminations materialized and grew slowly but steadily with shot number until they became large enough to allow parasitic failure, despite the tilted disk edges. Calculations indicated that thermal stress at the bond from the single-pass ASE fluence on the cladding (i.e., no parasitic lasing within the disk volume) approached the fracture strength of the glass. We therefore concluded that such delaminations were created and enlarged by the ASE-induced stress at the bond. Our first job was to eliminate this slow delamination of the cladding with shot number.

We addressed the problem of slow delamination from two directions simultaneously: by attempting to both increase the bond strength and to reduce the stress on the bond. The most promising of the bond-strengthening techniques involved the use of a silane-coupling agent (SCA) to strengthen the epoxy-to-glass

bond. Peel tests showed a consistent twofold increase in bond strength with SCA treatment. We also found that an acid pretreatment of glass surfaces increased bond strength (see the article "Improvement in Polymer Formulation and Properties," later in this section).

Calculations performed concurrently with the work on bond strengthening indicated that reducing the absorption coefficient of the cladding from 0.75 to 0.45/mm would significantly reduce peak stresses at the bond (see the article "Modeling of Edge-Cladding Thermal Stress," later in this section). Because reducing the cladding absorption also increases the feedback of ASE from the edge cladding, we also had to demonstrate that the reduced absorption would not degrade disk performance. Back-to-back tests of a single disk with different cladding absorptions showed no performance penalty with the 0.45/mm cladding (see the article "Performance Tests of Prototype Edge Claddings," later in this section).

We tested the effect of our modifications on the slow-delamination problem with a series of life tests. In these tests, we exposed several disks to 1000 or more shots in Nova amplifiers under typical Nova operating conditions (see the next article, "Life Testing Prototype Edge Claddings"). Our life tests required an automatic gain-test facility, which allowed continuous testing for 24 hr/day to accumulate the required number of shots in a reasonable length of time (see the article "Proof-Test Facility," toward the end of this section). These tests produced no failures or visible changes to any of the modified claddings. Furthermore, one of the disks survived 4000 shots, which corresponds to about 8 years of service on Nova, with no performance change or visible change to the cladding. These successful life tests provided the final experimental proof of the viability of composite polymer/glass claddings, and they allowed us to finalize the design for Nova disk production.

We also investigated two other approaches to the slow delamination problem: increasing the glass transition temperature of the epoxy, and using a

laminated absorbing glass for the cladding. Epoxies with higher glass-transition temperatures were investigated briefly because of their higher adhesive and tensile strengths (see the article "Improvement in Polymer Formulation and Properties," later in this section). The idea behind the laminated cladding was to reduce stress at the cladding bond by distributing the thermal load more uniformly throughout the cladding thickness. Calculations showed that, not only could thermal stresses at the bond be reduced, but tensile stress could be reversed in sign to become compressive, and compressive stress would neither generate nor propagate delaminations (see the article "Modeling of Edge-Cladding Thermal Stress"). However, we chose not to complete the development of either of these two approaches in light of the above successes.

In addition to direct work on the damage problem, we undertook two efforts to better understand ASE depumping of laser disks and adhesive joints in general. In the first effort, we developed a three-dimensional (3-D), Monte-Carlo ray-tracing code to calculate the influence of cladding imperfections on gain performance in laser disks (see the article "Calculations of Effects of Imperfect Claddings," later in this section). This code showed the effect of small changes in the tilt angle of disk edges in the presence of cladding delaminations, which were totally absent in two-dimensional (2-D) models and provided the basis for picking the tilt angles for the final cladding design. The code also predicted that the tolerance on index matching could be relaxed considerably, and it showed that the 0.45/mm absorption coefficient of the final cladding design was completely adequate to prevent a reduction in disk performance.

Our investigation of adhesive joints showed the relation between bulk properties of the epoxy and shear and tensile strengths of epoxy bonds (see the article "Understanding and Measuring Adhesive Joint Strength," later in this section). It also established a firm relation between moisture uptake and glass-transition temperature of epoxies and their effect on

epoxy bond strength. This relation helped us to understand a humidity problem encountered in the early phase of cladding production, and it helped explain the variability seen in early results of peel tests when aging and humidity were not controlled.

The second phase of the development program on edge claddings focused on transferring the new edge-cladding technology to industry. This effort was a collaboration with our vendors to establish the production steps necessary for the cladding process and to initiate a quality assurance program to ensure consistent production results. The second phase started early last year, and many of the details were presented in the 1986 Laser Annual.⁵⁹ This year, we made the required changes and additions to implement the above improvements and to finalize the production process (see the last article in this section, "Cladding Quality Assurance Program and Process Engineering"). We also hired a consultant firm to review both our quality assurance program and our procedures for dealing with problems singled out by the quality assurance program.

By year end, we began full-scale production of the composite polymer/glass edge cladding on platinum-free disks, and we replaced disks in the first few Nova amplifiers. To date, we have experienced no damage or performance problems with the production version of our new edge cladding. We anticipate completing the replacement of Nova disks by the end of next year.

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Life Testing Prototype Edge Claddings. Our emphasis in the first half of 1987 was on life testing to determine if our edge-cladding design would perform satisfactorily for several years on Nova. The successful life tests of the prototype edge cladding described in this article marked the major milestone of the year and of the entire edge-cladding development program. It demonstrated beyond any doubt the viability of the polymer-based cladding, and it finalized the design for the production cladding. After we passed

this milestone, we began proof testing the amplifiers for 100 shots as they were installed on the first upgraded Nova beamline.

Our life testing concentrated primarily on the 31-cm amplifier because we had previously found that its disks were more susceptible to edge-cladding failure than the 46-cm disks.⁶⁰ The 31-cm disks are more problematic because of their longer gain path (62 cm vs 48 cm for the 46-cm disks) and because of their greater amplified spontaneous emission load (approximately 35% greater power density to the claddings). The long gain path promotes both surface- and bulk-mode parasitic oscillations, and the additional heat load tends to drive cladding/disk delaminations. As discussed in the 1986 Laser Annual,⁶⁰ we believe that the combination of these two effects causes cladding failure. The thermal-induced delamination grows with shot number until it is large enough to allow parasitic oscillations, which then produce further and rapid delamination leading to total cladding failure.

To combat the problem, we first tilted the edges of the disks to prevent surface-mode parasitic oscillation (see the article, "Calculations of Effects of Imperfect Claddings," later in this section). Second, we employed silane coupling agents to strengthen the epoxy bond to the cladding glass and disk (see the article, "Improvements in Polymer Formulation and Properties," later in this section). Finally, we reduced the 1- μ m absorption coefficient of the cladding glass, which reduced the temperature at the bond interface and thus the stress on the adhesive joint (see "Modeling of Edge-Cladding Thermal Stress," later in this section).

To test the effect of these improvements, we obtained two 31-cm disks, one whose cladding was fabricated at Zygo Corp. and one whose cladding was fabricated at Eastman Kodak. Both had 0.8° tilts oriented in our initial configuration (see "Calculations of Effects of Imperfect Claddings"), and both were fabricated with a silane surface treatment. However, a major difference between the disks was the fact that the 1- μ m absorption

coefficient of the Zygo disk was 0.45 mm^{-1} , while that of the Kodak disk was 0.75 mm^{-1} . These absorption coefficients were chosen because both values had been successfully employed with the original Nova disks made with fused-glass claddings. Both absorptions are greater than that necessary (0.3 mm^{-1} at 6-mm thickness) to prevent bulk-mode parasitics, based on a simple model where the ray is presumed to totally reflect off the outside of the cladding. Measurements in the amplifier test facility also showed that the gains of these individual disks were the values expected from their thicknesses and lifetimes (see the next article, "Performance Tests of Prototype Edge Claddings"). This result confirms that the claddings were adequately "black."

We conducted a test of these two disks using the new gain-measurement facility (see the article, "Proof Test Facility"). We accumulated 1100 amplifier firings over approximately one month at the nominal Nova operating condition (20-kV bank charge). At this point, the gain of the disk with 0.75-mm^{-1} absorbing cladding began to decline rapidly. Inspection showed that its cladding delaminations had grown large enough to allow bulk-mode oscillation to occur. Delamination strips approximately 1 mm in width had been created at both faces, thus allowing parasitics to occur despite the tilting of disk/cladding interfaces. We believe these delaminations were created and enlarged by the thermal stress produced by the higher-absorption-coefficient cladding glass. Inspection showed no change for the disk with the 0.45-mm^{-1} cladding.

We then replaced the failed disk by a second disk fabricated by Zygo with the lower absorption cladding, and we extended the test for another 1000 shots. No significant change in gain was observed for either disk. Finally a disk with 0.45-mm^{-1} cladding fabricated by Kodak was placed in the amplifier together with the first successful disk, and the test was continued for another 2000 shots. Figure 3-43 shows the measured gain performance of the amplifier over this last test of 2000 shots. The gain measurements for the plus and minus sides are for

Fig. 3-43. Gain vs shot number for a 31-cm amplifier outfitted with disks using our production cladding design.

positions displaced horizontally from the amplifier centerline by ± 6 cm. The region of higher gain around shot 400 corresponds to several shots taken at 22-kV bank charge.

The gain was extremely steady over this test period, which corresponds to about four year's use on Nova. The very small loss in gain that does occur with shot number presumably is caused by a slight degradation in the silver reflectors. Notice that the two gain channels track extremely well, as they should. The disks were removed from the amplifier for inspection, and we found no observable change in their claddings. After this inspection, the disks were put back in the amplifier and subjected to 100 shots at 24-kV charge, well above the normal Nova conditions. Another inspection again showed no change.

These tests served to qualify our cladding design on 31-cm disks as produced by our two fabricators. Notice that one of the disks was tested for a total of 4000 shots, roughly 8 years of service at Nova shot rates.

We obtained a 46-cm amplifier with four production-finished (meeting Nova flatness and polish specifications), platinum-particle-free disks for testing. Two of these disks had 0.75-mm^{-1} cladding glass, while two had 0.45-mm^{-1} absorbing glass. The amplifier was first installed on Nova at the output position of a chain (the E position), which experiences the highest laser energy, and tested for 25 system shots at currently nominal conditions (5 kJ/beamline, 1-ns pulse). Inspection after removal from

Nova revealed that there was no bulk damage in the glass and that the claddings were unchanged. This test verified the effectiveness of our off-line testing for platinum particles and proved that there are no unexpected failure modes of the cladding during actual Nova use.

After the Nova test, the amplifier was installed in the proof-test facility and subjected to another 800 bank firings. The gain was stable, and inspection again showed that the claddings were unchanged. The lack of cladding damage in these 46-cm disks with 0.75-mm^{-1} absorption coefficient claddings confirms our previous conclusion that the claddings of the 46-cm disks are less highly stressed than those of the 31-cm disks. This result also tends to confirm that our current edge-cladding design, which uses the smaller absorption coefficient cladding glass, has a considerable margin of safety.

The actual proof testing of amplifiers prior to their installation on Nova began near the end of the year. At year end, we had tested two completed 31-cm amplifiers for 100 shots each before installation on the system. It is interesting to note that the measured plus- and minus-side single-pass gains were roughly 1.90 and 1.75 at 20 kV charge, exceeding our expectations. This result is produced mainly by the uniformly long excited-state lifetimes (approximately 400 μs at 2% doping) of the Schott-produced laser glass. To date, we have experienced no

edge-cladding failures for any of the disks fabricated with our production-cladding design.

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Performance Tests of Prototype Edge Claddings. We tested 31.5-cm disks with several cladding transmissions to experimentally determine the influence of cladding absorption on amplifier gain. We specifically wanted to demonstrate that the 6-mm-thick cladding with an absorption coefficient of $0.45/\text{mm}$, which was proposed to reduce thermal stress on the epoxy bond, would not reduce gain performance of the disk. Beyond that, we wanted to know how much the cladding absorption could be reduced before it affected gain performance.

We tested a total of eight disks with six different edge claddings. The βt products (absorption coefficient at $1\text{ }\mu\text{m}$ times cladding thickness) varied from 4.5 to 1.0, giving single-pass cladding transmissions from 1 to 37%. We chose to test 31.5-cm disks because they are more heavily stressed by ASE than are the 46-cm disks, as discussed in the previous article. We tested only one disk in the 2-disk, 31.5-cm amplifier to avoid the expense and delay of fabricating a second disk.

Figure 3-44 shows gain coefficients as a function of initial bank energy for βt products of 4.5, 1.8, 1.3, and 1.0. We chose this particular set of four out of the eight tested because the same 31.5-cm disk was used for each, eliminating the need to correct the data for different disk fluorescence lifetimes and thicknesses. The disk was first fabricated with a 6-mm-thick, $0.75/\text{mm}$ absorption coefficient cladding giving a βt product of 4.5. After testing, the first cladding was removed and replaced by a 6-mm-thick, $0.3/\text{mm}$ cladding with $\beta t = 1.8$. After the second test, this cladding was ground to a thickness of 4.4 mm ($\beta t = 1.3$) for the third test, and, finally, to a thickness of 3.4 mm ($\beta t = 1.0$) for the fourth test.

Figure 3-44 shows no significant performance degradation for claddings with βt

Fig. 3-44. Measured performance of four 31.5-cm disks with different edge-cladding absorptions.

products as low as 1.3. Therefore, the proposed cladding with $\beta t = 2.7$ will clearly not cause a performance degradation. In fact, it provides a comfortable factor of 2 margin of safety in this regard.

The data spread in Fig. 3-44 at intermediate bank energies (100 to 200 kJ) is larger than expected. In this region, ASE effects are small because gain coefficients are relatively small, and we would expect all the edge claddings to perform the same. Past experience indicates that our measurement accuracy in this region is about $\pm 0.5\%$, or about one-half the size of the data symbol and too small to account for the observed differences. We attribute the spread to the fact that we had to test these four claddings in different 31.5-cm amplifiers. Our experience with off-line testing of Nova amplifiers implicates a performance variation of about $\pm 1\%$ among amplifiers of the same type, presumably due to differing transmissions and reflectivities of blastshields, flashlamp envelopes, and reflectors.

The additional four disks tested had βt products of 4.5, 2.7 (2 disks), and 1.3, but the disks themselves were different. Consequently, comparing the gain coefficients from these to the data of Fig. 3-44 required eliminating the effects of lifetime and thickness differences among the various disks. After corrections based on GAINPK, these curves each fell within the band defined by the $\beta t = 4.5$, 1.8, and 1.3 curves in Fig. 3-44 and the assumed $\pm 1\%$ error. We chose not to add them to the figure because of the added uncertainty created by their corrections.

Our results are also in good qualitative agreement with calculations given in the article, "Calculations of Effects of Imperfect Claddings," later in this section. For βt values down to 1.8, calculations indicate no change in performance, just as we observe experimentally. For smaller values of βt , the calculations become sensitive to the details of how the ASE is backscattered from the external surface of the cladding glass, which is fine-ground on all our claddings. The assumption used for the "diffuse" model of the calculation assumes all the ASE that survives one pass through the cladding glass is backscattered, clearly a worst-case

assumption because much of the incident ASE will be transmitted and not returned to the disk. Even so, the calculation shows a relatively slow onset of ASE effects with decreasing βt . It predicts performance decreases of 10 and 25% for βt products of 1.3 and 1.0, respectively, compared to the 0 and 6% observed experimentally. These discrepancies are quite consistent with the worst-case nature of our assumptions in the calculation.

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Improvements in Polymer Formulation and Properties.

Acid and Silane Surface Treatments.

A major thrust this year was to increase the strength of the epoxy-to-glass bond with the Delta epoxy formulation (described in last year's Laser Annual⁶¹) as the baseline reference material. Lifetime testing of amplifier disks bonded with this epoxy showed a slow delamination of the cladding with increasing shot number, which we related to thermally induced stress at the bond. The most obvious of the several approaches we took to solving this problem was to increase epoxy bond strength.

Silane coupling agents are frequently successful at increasing the adhesive strength of polymeric bonds to glass surfaces. Conceptually, these agents function as an interface between the adherents by forming covalent chemical bonds both between the silane and the glass surface and between the silane and the resin component.⁶¹

While we generally find SCAs to be an effective means of increasing adhesive bond strength, we found Z-6020 [3-(2-aminoethylamino)-propyltrimethoxysilane] (Ref. 62) to be a more effective coupler than either Z-6040, which contains a reactive epoxide group, or Z-6032, which contains a vinylbenzyl- and amine-organic group. We also found it more effective to apply the SCA directly to the glass surface than to add it to the bulk polymer, where migration to the surface is undoubtedly hindered by premature reaction with the matrix.⁶²

Fig. 3-45. Effect of cure temperature on peel strength.

In addition to the above, we optimized such process variables as the drying time of the SCA and the cure temperature of the SCA-treated epoxy bond lines. We found that the length of time SCA-treated glass dried before bonding to the epoxy had no effect on bond strength. A series of samples dried for periods ranging from 6 hr to one week similarly showed no noticeable effect on peel strength.

Figure 3-45 shows a brief study of the effect of cure temperature on peel strength with the SCA treatment. In each case, the sample was held at temperature until cure was complete, as judged by the disappearance of the epoxy absorption in the near-IR absorption spectrum. The exception was the 40°C cure experiment, in which no cure could be attained. A 100°C cure temperature produced the maximum peel strength but caused yellow discoloration of the sample. Acceptable results were achieved with temperatures of 60 and 80°C while the results at temperatures of 120 and 40°C showed significantly reduced peel strengths. In the final production cure cycle, the temperature of the clad disks was ramped to 80°C and held at that temperature for 4 hr.

Bond-strength measurements with 90° peel tests revealed that bond strength clearly improves with the SCA treatment. Tests showed an average peel strength of 11 ± 2 N·cm without SCA and of $24 \pm$

4 N·cm with the optimized SCA treatment—an increase of almost 120%. As part of the quality assurance program, these averages include both LLNL and vendor data.

Electron scanning microscopy for chemical analysis (ESCA) of acidified Z-6020 SCA solution-treated phosphate glass⁶³ showed that an ~15-Å-thick layer containing silicon, carbon, nitrogen, and oxygen was indeed present on the surface of the glass. Similar analysis of the glass substrate for a peel specimen showed no Si on the surface of the phosphate glass after the epoxy was peeled off, but it was detected on the peeled surface of the epoxy strip, indicating that the silane coupler was removed as the specimen was peel tested. Small amounts of phosphorus on the epoxy surface also seem to indicate that bond failure occurred at the glass-silane interface rather than at the silane-resin interface. Finally, no organic carbon could be detected on the surface of the glass. These observations indicate that Z-6020 maximizes the interfacial bonding between SCA and resin to the extent that bond failure occurs at the glass-silane interface in preference to resin-SCA debonding.

ESCA analysis by a second laboratory confirmed that the Z-6020 SCA layer is ~15 Å thick, whether deposited from acidic or alkaline solution. Surprisingly, they also reported that a conformation with a decreasing nitrogen concentration toward the glass surface is indicated [configuration (b) rather than (a) in Fig. 3-46]. This finding is inconsistent with expected acid-base interactions (at least for the alkali solution), wherein the glass surface should be basic, not interactive with the electronegative amine group, as shown in Fig. 3-46(c). Consequently, the conformation of the flexible alkyl group [Fig. 3-46(d)] is postulated to explain the ESCA data. Note that the amine groups are relatively shielded from the silane-air interface during ESCA analysis but are still available for coupling to the epoxy. This conformation is expected to be stabilized by hydrogen bonding between the amine group and the P-O bond as shown.

Although bond strength clearly improved with the SCA treatment, peel-strength measurements showed considerable

variation as production proceeded. Since quality control tests showed the material parameters to be within specifications, we initiated studies of glass surface effects in an attempt to understand this variability.

A basic tenet of adhesion science is that the bonding surface must be clean, so, from the earliest stages of the project, we took pains to ensure that the glass used had been properly cleaned and bagged according to standard clean-room procedures. (Procedures include degreasing the glass with acetone, washing it with green medical hand soap, rinsing it with distilled water, scrubbing it lightly with 50:50 ethanol:acetone, and swiping it with either acetone or ethanol, using a class 100 lint-free lens tissue.) We found, for instance, that nylon bags must be used rather than polyethylene bags because the latter release plasticizers onto the glass surfaces. These plasticizers interfere with effective wetting of the surface by the resin—a necessary condition for maximizing adhesion—and thereby cause a precipitous decrease in peel strength. A number of standard cleaning procedures were also evaluated, but none was found as effective for maximizing peel strength as freshly polishing the glass surface. Cleaning with chromic acid, acetone, methanol, or KOH/chromic acid gave similar peel strengths but only about 80% of that for a freshly polished surface.

We found acid pretreatment of the glass, either separately or in conjunction with the SCA treatment, an effective way to increase peel strength. Table 3-4 shows data for both cases (separate or with SCA) for several soak times in acidic SCA. Without the acid pretreatment, peel strengths initially increase with soak time, leveling out after about 20 min; with the acid pretreatment, they are independent of soak time.

Two possible contributors to the improved strength with acid treatment can be envisioned. One is ion exchange, which enriches the surface with respect to $-OH$ groups that are themselves more reactive toward SCA than the replaced alkali or alkaline earth ions. The other is the removal of water- or acid-soluble

surface-contamination products (e.g., metal hydroxides or carbonates) formed by the reaction of the glass with atmospheric components. The latter process, "weathering" of the glass surfaces, is

Fig. 3-46. Possible orientations of SCA molecules with respect to the glass surface.

Table 3-4. Effect of acetic acid pretreatment of phosphate glass on peel strengths^a with Delta formulation epoxy.

Time in SCA + ^b	Peel strengths (N·cm)	
	No acid	With acid
5 s	17 ± 2	24 ± 4
1 min	21 ± 3	24 ± 2
25 min	30 ± 6	28 ± 5

^a Peel strengths measured at 90°.

^b SCA + = 1% Z-6020 in 4:1 H₂O:MeOH at pH4, adjusted with acetic acid.

Fig. 3-47. Weight change in Delta epoxy upon exposure to moisture in ambient air.

known to be a problem with glasses in general, including phosphate glasses,⁶⁴ as well as a mechanism for reducing the adhesive strength of inorganic coatings. To minimize weathering from atmospheric moisture, we have instituted humidity control for the entire cladding procedure, and we bag and seal the finished disks with desiccant for storage.

Effects of Water and Humidity. We investigated the effects of water and humidity on bond strength because of a problem one vendor had with increased humidity during a few cladding operations. Qualitatively, we can surmise that samples of the Delta epoxy absorb atmospheric moisture because their flexural modulus (stiffness) decreases upon exposure to ambient lab conditions and that the absorbed moisture apparently acts as a plasticizer (see "Understanding and Measuring Adhesive Joint Strength"). Depending on the prevailing relative humidity, short-term "equilibrium" values—as gauged by thermogravimetric analysis (TGA)—of roughly 2 to 3 wt% absorption are realized within three to nine days. Figure 3-47 shows the moisture-produced change in weight for a small sample (~10 mg) of Delta epoxy. The figure also shows, at least for the small samples

used in this TGA analysis, that the effect is reversible upon heating for 30 min at 100°C.

The upper limit for water absorption seems to be considerably higher, evidenced by the fact that soaking a sample in water for six days produced an 8.3% gain in weight. Studies have shown that water absorption may, depending on chemical structure, differ by a factor of ten among different resin types.⁶⁵ For example, systems that contain significant concentrations of hydroxyl or other oxygen-containing or polar groups would be expected to absorb significant amounts of water. However, adhesion to polar surfaces such as glass also seems to be maximized in such systems.

The literature contains many references to the deleterious effects of moisture on the physical properties of epoxies, those of principal interest here being adhesion and the glass-transition temperature, T_g (the midpoint of the temperature range wherein polymers undergo a transition from glass-like to rubber-like properties). Studies⁶⁶ indicate, for adhesives, that the effects of moisture on T_g may be more important than on interfacial properties. For example, an additive such as water that lowers the T_g from a point above the test temperature to some point below it reduces system strength, which may lead to subsequent cohesive failure within the epoxy. This correlates, at least qualitatively, with our own observations, namely that T_g decreases with the absorption of atmospheric moisture and that peel strength decreases with time (presumably as a function of moisture absorption). The former point is illustrated by the data of Table 3-5, which show both the T_g and the percentage weight change of a Delta epoxy sample as being significantly affected by exposure to ambient air. If the sample is heated to 100°C for a short time, the T_g rises. The T_g remains at an elevated level if the sample is kept dry, but it decreases reversibly if the sample is exposed to moisture. Figure 3-48 illustrates the decrease in peel strength with time. Note the significant decrease in peel strength after only one week of exposure to ambient conditions.

The procedure for maintaining a dry bond line—by bagging with desiccant and operating in the dry nitrogen atmosphere of the amplifier—should be relatively straightforward once the finishing procedure has been completed. However, as shown for two epoxy adhesives of undefined composition,⁶⁷ an absorbed moisture content of more than 0.3 wt% irreversibly degrades physical properties. Therefore, stringent handling and storage procedures are necessary.

An integral part of the finishing operation for Nova amplifier disks involves grinding and polishing the clad disks. Consequently, the epoxy bond line between the disk and cladding is in contact with both water and organic grinding aids for as long as 24 hr. In addition, the two-phase glue-up operation (see "Cladding Quality Assurance Program and Process Engineering") dictates that the first-phase cured bondlines be exposed to the SCA glass treatment prior to the second gluing operation. Table 3-6 shows the effect, on polymer buttons, of exposure to these processing materials. The results are overly pessimistic both because of the relatively larger surface-area exposure in the test specimens and the fact that the outer, most moisture-rich section of the bond line is being continually removed in the grinding and polishing process. Nevertheless, it is readily apparent that the time for which bond lines are exposed to coupling and grinding solutions must be minimized during production.

The moisture-absorption rate in the assembled amplifier is expected to depend on the exposed bond-line surface area, which is quite small. In fact, 10 years has been postulated as necessary for the moisture content to reach a steady-state value in a composite 0.5 in. thick.⁶⁸ The argument has also been made, since the absorption of water can result in swelling stresses to counter those from the cure of the resin and cooling, that a certain degree of water absorption can essentially eliminate stress in a composite.⁶⁹ In sum, these studies and our amplifier tests suggest that the level of moisture absorption by the bond lines should be acceptable provided the cladding and

Table 3-5. Effect of storage conditions on T_g and weight change of Delta epoxy.

Sample ^a	Treatment	T_g (°C)	Weight change	
			Total (%)	Delta (%)
1	None ("as is")	30	—	—
	30 min at 100°C	37	—	—
	18 hr in dry N ₂	36	—	—
	7 days ambient air	28	—	—
2	30 min at 100°C	—	-2.3	-2.3
	6 days ambient air	—	-0.2	+2.1
	11 days ambient air	—	+0.7	+3.0

^a Both samples mixed from the same batch of starting materials.

◀ Fig. 3-48. Peel strength vs age of peel specimen.

Table 3-6. Fluid absorption by Delta epoxy.

Fluid	Weight gain (%)			
	1 hr	4 hr	24 hr	6 days
Water	0.7	—	3.7	8.3
0.2N acetic acid	4.3	—	Disintegrated	
1% Z-6020 (SCA, pH4, acetic acid)	6.7	—	Disintegrated	
% Z-6020 (SCA, pH10.4)	1.4	3.1	7.4	11
Surcool (grinding aid, pH9.1)	0.8	1.8	4.1	7.8
Blasocut (grinding aid, pH9.1)	0.6	1.5	3.5	7.8

finishing operations are conducted rapidly and the disks are properly packaged as soon as possible after finishing.

Higher Glass-Transition Temperatures.

Delta epoxy was specifically formulated to match the refractive index of the phosphate laser glass. This, coupled with concern about the robustness of the phosphate laser glass, initially led us to limit the temperature used in the thermal cure cycle. The result was a relatively low T_g from the Delta formulation and an incompletely cured epoxy with a correspondingly lower T_g . Early this year, concern about the resulting low T_g and the realization⁷⁰ that physical properties

such as adhesive bond strength and tensile strength are substantially reduced above T_g prompted us to institute a higher temperature cure cycle to more completely react the epoxy and increase its T_g . Even so, the typical T_g for production claddings runs about 34°C and, with an anticipated operating temperature of 23°C, there is no large margin of safety. Given that the refractive index match between the glass and adhesive is no longer so crucial with tilted disk sides (see "Calculations of Effects of Imperfect Claddings"), we considered reformulating the epoxy to produce a higher T_g material.

A higher T_g epoxy could be obtained by increasing the aromatic epoxy constituent, DER-332, in the current Delta blend (DER-332—bisphenol A diglycidal ether, by Dow Chemical Co.; RD-2—butanediol-1, 4-diglycidal ether, by Ciba Geigy Corp.; and T-403—polyetheramine, by Texaco Chemical Co.). We determined T_g for nine stoichiometrically balanced epoxy samples ranging in resin composition from 0 to 100% DER-332, using differential scanning calorimetry at a heating rate of 20°C/min. As shown in Fig. 3-49, T_g increases with DER-332 concentration to a maximum of 86°C for a system composed strictly of DER-332 and T-403. If the cured epoxy network is treated as a copolymer of DER-332 + T-403 (stoichiometric) and RD-2 + T-403 (stoichiometric), a lower bound rule of mixtures known to apply to random copolymers and miscible blends can be used to describe the variation of T_g with composition:

$$1/T_g = w_1/T_{g1} + w_2/T_{g2}, \quad (1)$$

Fig. 3-49. T_g as a function of DER-332 content in Delta-type epoxies.

where subscripts 1 and 2 refer to the weight fractions and T_g of the DER-332 and RD-2 epoxies. Figure 3-49 also shows the calculated fit of Eq. (1), with $T_{g1} = 359^\circ\text{C}$ and $T_{g2} = -9^\circ\text{C}$, determined by a separate experiment.

The expected advantages of a higher T_g formulation include faster gel and cure times, less moisture absorption, and a simplified formulation, including elimination of the accelerator, as well as higher adhesive strength. The drawbacks to using a pure DER-332/T-403 system include higher viscosity, which would increase the difficulty of filtration. To preclude crystallization of the DER-332, a small percentage of the aliphatic epoxy constituent (RD-2) would have to be retained. Other cross linkers, such as aromatic or cycloaliphatic amines, could also be used to increase the T_g .

Refractive Index Variation During Curing. As part of the quality assurance procedure, we require that vendors measure and record the refractive index of freshly mixed batches of epoxy before cementing the absorbing strips to the laser disks. Requiring the refractive index to fall within a specified range helps ensure that the constituents are properly mixed and makes possible a traceable record in case a problem with the cladding should subsequently develop. Since the vendors do not exercise fine control over measurement times or the temperature of assembly areas, the refractive index of properly formulated and mixed samples of Delta epoxy had to be determined as functions of both ambient temperature and elapsed time since mixing.

Figure 3-50 shows data for two batches of Delta starting materials, the standard batch used at LLNL and the master batch vendors use for the first production run. The isotherm lines for both batches intersect rather than parallel each other because the system cures faster at higher temperatures, thereby increasing its refractive index faster. The intersection point for the LLNL standard batch occurs considerably faster than that for the master batch, presumably indicating faster kinetics from what is possibly a more active accelerator.

Fig. 3-50. Refractive index of Delta epoxy vs time from mixing for two batches of starting materials.

Although differences exist between the two batches of materials, they are less than the tolerance on final index (± 0.003). Moreover, they probably reflect kinetic (e.g., rate of cure) differences rather than absolute differences since there are no previous indications that the refractive indexes of the cured epoxies differ significantly for the two batches of materials.

The question of how reproducible these measurements are is addressed by the 30°C plot for the vendor's master materials. In that determination, parallel readings were obtained for two separate mixtures, and the agreement was excellent.

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Modeling of Edge-Cladding Thermal Stress. The slow growth of delaminations with shot number was our major problem at the beginning of this year. To help understand the problem, we calculated the effect of delamination depth and cladding doping on stress, and we evaluated the possibility of using a laminated cladding consisting of a layer of doped glass

Fig. 3-51. Modeled temperature rise in Nova disk with ASE heating. Values shown are ΔT in °C.

fused to a layer of clear glass. In addition, we developed an analytic model of bond delamination to assess the effects of stress and bond strength on delamination rate.

Effect of Bond Delaminations. We calculated stresses with the NIKE2D finite-element code.⁷¹ Figure 3-51 shows the assumed temperature distribution. We calculated the temperature rise in the cladding for an absorption coefficient of 0.75/mm, assuming an ASE fluence of 10 J/cm² in 0.5 ms. The temperature rise is modeled as occurring instantaneously because no significant heat conduction occurs during the 0.5-ms laser pulse. Stresses are calculated at the end of the

laser pulse, which gives peak stresses since the temperature gradients are largest at this time. The bond does not significantly affect the stresses⁷² because it is so thin ($<25\text{ }\mu\text{m}$). Thus, the bond is assumed to have the same strength as the glass.

We modeled bond delaminations with slidelines in the finite-element mesh. At the slidelines, nodes can move freely parallel to the interface, but they cannot penetrate into the interface. Figure 3-52 shows contours of shear and normal stresses for the case without a delamination. With the sign convention used in our analysis, positive values of normal

stress (i.e., the stress acting to pull the disk and cladding apart) represent tension, while negative values represent compression.

In Fig. 3-52 the maximum shear stress occurs at the cladding interface, while the maximum normal stress occurs in the cladding a short distance from the bond. Peak shear and normal stresses at the bond are about 6.8 and 5.6 MPa, respectively.

Figure 3-53 shows the effect of a bond delamination. In this case, we modeled a 0.5-mm delamination at the cladding interface starting from the disk surface. The presence of the delamination causes

Fig. 3-52. Calculated (a) shear and (b) normal stress contours without delamination. Values shown are in MPa.

Fig. 3-53. Calculated (a) shear and (b) normal stress contours with 0.5-mm delamination. Values shown are in MPa.

a sharp stress concentration at the delamination edge. The peak interface normal stress remains about the same (5.5 vs 5.6 MPa); however, the maximum magnitude of shear stress increases from about 6.8 to 13.1 MPa (stress concentration factor = $13.1/6.8 = 1.9$).

We modeled additional cases with varying delamination depths, as shown in Fig. 3-54. Within the range analyzed, shear stress increases with increasing delamination depth, whereas normal stress is relatively unaffected.

Failure analysis shows that cladding failure depends most strongly on the shear-stress component (see the article "Understanding and Measuring Adhesive Joint Strength," later in this section). Consequently, the above result implies that even a small delamination of 100 μm would pose a serious threat to the bond because it would significantly increase shear stress at the bond. If a delamination grew, stress would increase more, speeding the growth rate until the disk eventually failed. Because we must assume that some delaminations may exist (due to imperfect production procedures), stresses must be kept low so that the bond is not allowed to begin to "unzip" in this manner.

Effect of Reduced Cladding Doping.

The peak temperature rise in the cladding is proportional to the absorption coefficient. We calculated the effect of a reduced absorption coefficient on interface stress. Figure 3-55 shows that the calculated peak interface shear and normal stresses decrease linearly with absorption coefficient. Consequently, an easy way to lower thermal stress is to lower the cladding absorption.

The criterion for selecting an absorption coefficient is that the cladding absorb enough ASE to prevent a degradation in disk gain. If the absorption coefficient could be reduced from 0.75 to 0.45/mm, Fig. 3-55 shows that the maximum interface shear stress would decrease from 6.8 to 4.2 MPa, and the maximum normal stress would decrease from 5.6 to 2.7 MPa.

Laminated Cladding. Another approach to our problem of thermal stresses is to move the adhesive bond away from the place where the temperature rise

Fig. 3-54. Maximum interface stress vs delamination depth.

Fig. 3-55. Maximum interface stress vs cladding doping.

is highest. A two-layer laminated cladding consisting of a layer of clear glass fused to the absorbing glass provides this possibility. By cementing the laminated cladding to the disk with the clear glass in between, the polymer is shielded from the peak temperature rise at the absorbing glass by the thickness of the clear layer.

We analyzed the two-layer laminated cladding with a 4-mm-thick layer of 0.75/mm absorbing glass. Stresses were calculated at the clear/absorbing glass interface and at distances of 1 and 2 mm from the absorbing glass to evaluate the effect of changing the thickness of the clear layer.

In the previous analyses, the polymer bond was at the point of maximum temperature rise (the clear/absorbing glass interface), and peak bond stresses occurred at the end of the laser pulse. Because this condition was no longer true with the laminated cladding, it was necessary to calculate stresses as a function of time by first calculating heat flow through the glass and then the resulting temperature gradients and stresses.

Figure 3-56 shows calculated shear stresses at distances of 0, 1, and 2 mm

Fig. 3-56. Calculated shear stress vs distance parallel to the interface for laminated cladding at (a) glass/cladding interface, (b) 1.0 mm from glass/cladding interface, and (c) 2.0 mm from glass/cladding interface.

from a clear/absorbing glass interface and at six times after the ASE pulse. The abscissa is the distance from the center of the disk to the disk surface. A sharp peak in shear stress occurs at the clear/absorbing glass interface ($\Delta x = 0$) near the disk surface at the end of the ASE pulse (0.5 ms). As the distance from the interface increases to 1 and 2 mm, the spike in stress broadens and decreases. At 2 mm, the stress is almost the same after the initial second as after the ASE pulse. (Additional results verified that a higher peak stress did not occur between the end of the ASE pulse and 1 s, and that the stresses decreased even more as the bond was moved farther away from the cladding.) Peak shear stresses at distances of 0, 1, and 2 mm from the interface are 7.0, 2.9, and 1.9, respectively. Thus, peak shear stress in the bond is reduced by 59% with a 1-mm layer of clear glass and by 73% with a 2-mm layer.

Figure 3-57 shows calculated normal stresses at distances of 0, 1, and 2 mm from the clear/absorbing glass interface. Once again, peak-magnitude stresses occur at the end of the laser pulse. Note, however, that the sign of the normal stress near the surface ($y = 21.6$ cm) changes. The normal stress is in tension (positive) at 0 mm, but it is in compression (negative) at distances of 1 and 2 mm. This difference is critically important.

For the case of a homogeneous cladding bonded directly to the disk, the polymer is at the cladding interface and is subjected to a peak tension of about

5.6 MPa. The effect is to pull the disk and cladding apart. Thus, a small delamination in the bond will tend to grow in size. With a laminated cladding, however, the bond is in an area of compression, and a small delamination will have less tendency to grow. Our analytical verification was done by inserting into the model a 0.5-mm delamination, 2 mm from the clear/absorbing glass interface. The results showed that both shear and normal stresses remained essentially unchanged from the case without delamination (i.e., no stress concentration). Thus, with respect to the possibility of a stress-induced failure, the laminated cladding is a much safer design.

In summary, a laminated cladding is a promising alternative because it

- Reduces maximum shear stress in the bond.
- Eliminates normal stress in the bond (i.e., the bond is in compression instead of tension), which reduces the likelihood of delamination growth.
- Reduces the laser-induced temperature rise in the polymer.

Laminated cladding glass is more difficult to manufacture; however, we did clad several prototype disks with laminated cladding glass obtained from Hoya Corporation, but it contained many bubbles near the interface and was quite expensive. The success of our other approaches made further development unnecessary.

Analytic Model of Delamination Growth. We derived an analytic model of delamination growth to better understand

▲ Fig. 3-57. Calculated normal stress vs distance parallel to the interface for laminated cladding at (a) glass/cladding interface, (b) 1.0 mm from glass/cladding interface, and (c) 2.0 mm from glass/cladding interface.

the factors affecting bond failures. The model combines elements of fracture mechanics, fatigue, and adhesion theory. Although the proposed model is untested experimentally, our analysis provides valuable insight into the mechanics of bond delamination. The results imply that reducing stress is more important than increasing bond strength to minimize delamination growth.

We first briefly review some principles of fracture mechanics. In this theory, it is assumed that the failure of brittle materials is caused by the growth of minor cracks and flaws, which eventually lead to large-scale fractures. At the tip of one of these cracks, locally high stresses exist. It is also assumed that, to a reasonable approximation, stress in the vicinity of the crack tip can be considered uniform and of magnitude

$$\sigma_k = k\sigma_0, \quad (2)$$

where σ_k is the local stress in the crack tip region, σ_0 is the applied (or remote) stress, and k is the stress concentration factor.

Griffith originally proposed in 1920 that the rate of crack growth is driven by the above stress.⁷³ He defined a parameter G as follows:

$$G \equiv \frac{dU}{dA} = \frac{dL \cdot dF}{dA} = (dL)(\sigma_k), \quad (3)$$

Here, G is the strain-energy release rate, dU is the energy available for crack

Fig. 3-58. Model for crack propagation.

extension, dA is the incremental area of crack growth, dF is the applied upward force, and dL is the incremental upward displacement at the crack tip. Figure 3-58 shows this model.

As previously mentioned, within the volume of material near the crack tip, stress is assumed to be uniform. The size of this region is not constant; however, we assume that it can be characterized by the length parameter L (see Fig. 3-58), and, within this region, linear elastic behavior is assumed for the material. With these assumptions, the strain ϵ caused by the upward force at the crack tip region is

$$\epsilon = \frac{dL}{L} = \frac{\sigma_k}{E}. \quad (4)$$

Here, E is a function of the Young's modulus of the material and may have different values, depending on the loading

conditions (e.g., plane stress or plane strain). Inserting Eq. (4) into Eq. (3) yields

$$G = \left(\frac{L \sigma_k}{E} \right) (\sigma_k) = \frac{L}{E} (\sigma_k^2). \quad (5)$$

The stress intensity parameter is now introduced, which characterizes the driving force for crack growth:

$$\kappa = (GE)^{1/2}. \quad (6)$$

Stress intensity has units of force divided by length to the 3/2 power. Combining Eqs. (5) and (6) and inserting Eq. (2) yields

$$\kappa = \sigma_k L^{1/2} = k L^{1/2} \sigma_0. \quad (7)$$

It has been found experimentally⁷⁴ that for metals, such as aluminum, stress intensity is proportional to the square root of crack length a . That is,

$$\kappa = a^{1/2} \sigma_0. \quad (8)$$

A comparison of Eqs. (7) and (8) shows that the characteristic length L of the crack-tip-stress field is equal to crack length divided by the square of the local stress concentration factor. Thus, the size of the crack-tip-stress field and the stress intensity increase with crack length. This result implies that the rate of crack growth also increases with crack length, in agreement with observations.⁷⁴

As stated earlier, stress intensity characterizes the driving force for crack growth. It has been found experimentally that for low values of stress intensity, a crack will not grow regardless of the number of load cycles. At a certain value of stress intensity, however, stable crack growth will initiate. The stress at this point is called the "fatigue strength," and in this region of stress, crack extension per load cycle can be related to stress intensity. Finally, at a critical value of stress intensity, crack growth becomes unstable, and large-scale fractures occur. The stress intensity at this point is called "critical stress intensity" or "fracture toughness" κ_{Ic} .

The relation between crack growth rate and stress intensity has been extensively

studied, both experimentally and analytically, with differing results. Several crack-propagation laws have been proposed, but all can be expressed in the form⁷⁵:

$$\frac{da}{dN} = \frac{\kappa^n}{M}, \quad (9)$$

where da/dN is the crack growth rate (i.e., increase in length per load cycle), and M is a material constant. Proposed values of n vary from 2 to 4, with a likely value of 2 for polymers.

We now propose a mechanics model for growth of adhesive bond delaminations based on fatigue theory. Although untested in our case, the principles of fracture mechanics have successfully been applied to crack growth in epoxy-aluminum bonds.⁷⁶ In the epoxy bond model, Young's modulus and stress intensity are replaced with the following "effective" values:

$$E_{\text{eff}} = \frac{2E_1E_2}{E_1 + E_2}, \quad (10)$$

and

$$K_{\text{eff}} = (K_I^2 + K_{II}^2)^{1/2}. \quad (11)$$

Here, E_1 and E_2 are Young's modulus values of the adhesive and adherend, respectively, and K_I and K_{II} are stress intensities for pure tension and pure shear conditions, respectively.

In our model of delamination, we make the following assertion. Assertion 1: *The theory of fatigue can be applied to bond delamination, and crack growth scaling laws remain valid.*

We now define a parameter that characterizes the relative strength of an adhesive bond. We first define the parameter dF_0 , which is the incremental upward force at a delamination edge required to extend the delamination one unit length (see Fig. 3-58). For a bond with a delamination force dF , the relative bond strength Υ is then

$$\Upsilon = \frac{dF}{dF_0}. \quad (12)$$

The work of delamination is equal to incremental force times incremental displacement. However, the work to delaminate a bond must be equal to the work of adhesion. Therefore, we make the following definition for the incremental work of adhesion:

$$dW_{\text{adh}} = dF_0 \cdot dL. \quad (13)$$

To bridge the gap between fatigue and adhesion theory, we now return to the concept of strain energy release rate. From Eq. (3), the strain energy release rate is equal to the energy available for crack extension dU divided by the area of crack extension dA . The parameter dU is the work done to extend the crack the incremental length da (see Fig. 3-58). For an adhesive bond, we assert that this is equal to the work of adhesion. Thus to extend our analysis, we make an additional assertion. Assertion 2: *The strain energy release rate for a delaminating adhesive bond is equal to the incremental work of adhesion divided by the incremental area of delamination growth.*

Based upon the above, the strain energy release rate for delamination is then equal to

$$G_d = \frac{dW_{\text{adh}}}{dA} = \frac{1}{\Gamma} \frac{dF \cdot dL}{dA} = \frac{dL}{\Gamma} \sigma_k. \quad (14)$$

Combining Eqs. (4) and (14) yields

$$G_d = \frac{1}{\Gamma} \frac{L}{E} \sigma_k^2. \quad (15)$$

Analogous to the fracture model, we define the stress intensity for delamination as

$$\kappa_d = (G_d E)^{1/2}. \quad (16)$$

Insertion of Eq. (15) into Eq. (16) yields

$$\kappa_d = \left[\left(\frac{L}{\Gamma} \right) \left(\sigma_k^2 \right) \right]^{1/2} = \left(\frac{L}{\Gamma} \right)^{1/2} \sigma_k. \quad (17)$$

Finally, combining Eqs. (17), (9), and (2) gives the following expression for the delamination rate da/dN :

$$\frac{da}{dN} = \frac{(\kappa_d)^n}{M} = M' \left(\frac{L}{\Gamma} \right)^{n/2} (k \sigma_0)^n. \quad (18)$$

Equation (18) shows that the delamination rate is proportional to the reciprocal of bond strength raised to the power $n/2$ and to the applied stress raised to the power n . We conclude that reducing stress is a much more effective way to minimize delamination growth than is improving bond strength.

The value of n in Eq. (18) is probably between 2 and 4, with a likely value of 2 for a polymer. For $n = 2$, doubling bond strength will decrease the delamination rate by a factor of 2; whereas reducing stress by one-half (e.g., a switch from an absorption coefficient of 0.75/mm to 0.45/mm) will decrease the delamination rate by a factor of 4. The benefit of lower stress would be even greater for higher values of n .

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Calculations of Effects of Imperfect

Claddings. In the 1986 Laser Annual, we described our design considerations for Nova edge claddings that led to the choice of disks with tilted edges and set cladding doping levels in both the laser glass and edge claddings.⁷⁷ Our work this year concentrated on finding a design that would avoid both parasitic oscillations in the disks and thermally induced delaminations of the polymer-bonded edge cladding. We refined our numerical calculations to look more closely at the predicted performance of polygonal Nova disks with the new composite edge claddings.

In addition to looking for catastrophic failure modes, we examined the relative performance of various designs with respect to predicted amplified spontaneous emission (ASE) depumping rates in laser disks. In this work, we used the updated Monte Carlo ASE model described in the article "Calculation of Amplified Spontaneous Emission Rates in Laser Disks," in Sec. 5. Our calculations have provided estimates of the optimum disk tilt angles, allowable refractive index mismatches and delamination sizes, and minimum cladding absorption in the design of the new composite-clad Nova disks.

The figure of merit we used in our calculations is the ASE multiplier M_{ASE} , which is the multiplicative increase in volume-averaged radiative decay rate in the disk over the spontaneous decay rate. This increase results from the stimulated emission caused by radiation that is trapped and amplified in the disk. The major consequence of the larger decay rate is a decrease in peak gain for a given pump level. For reasonable values of the multiplier (≤ 10), numerical modeling taking into account both the effects of wavelength- and temporal-averaging of the ASE multiplier indicates that the peak gain in the disks scales approximately with $M_{\text{ASE}}^{1/4}$ (see the article "Calculation of Amplified Spontaneous Emission Rates in Laser Disks," in Sec. 5). Larger values of the ASE multiplier can produce a catastrophic dropoff in disk energy storage, approaching a parasitic mode in which gain is clamped by the in-disk radiation.

Predicted ASE Decay Rates for Claddings with a Refractive Index Mismatch.

The first imperfection in edge cladding that we considered is a difference in the refractive index of the polymer from that of the laser and cladding glasses. In agreement with experimental conditions, we assumed that the indexes of the cladding and laser glasses were equal and that the refractive index of the polymer was less than that of the glass. We modeled the effect of reflection at both the laser glass/polymer and cladding glass/polymer interfaces by assuming an effective total reflectivity R_{eff} of the form

$$R_{\text{eff}} = R(2 - R), \quad (19)$$

where R is the average of the perpendicular and parallel-polarization Fresnel reflectivities at the interface. This model for total reflectivity has the proper limit at low and high reflectivity for incoherent reflection from the double interface. We neglected the coherent addition for rays with the correct wavelength to give in-phase reflections from the double interface. Subsequent propagation would take these rays to different edge positions at different angles of incidence where the

interface thickness was different, randomly changing the phase difference for each edge reflection. We also ignored the translation in space of the ray reflected from the second interface and assumed that all reflected power reentered the laser glass at the point of incidence. In our Monte Carlo calculations, a component of each ray equal to R_{eff} was returned to the disk at each intersection with the edge, and the summation for the effective gain truncated when the intensity of the ray became negligible.

The effect of a mismatch in refractive index strongly depends on whether the disk edges are perpendicular or tilted with respect to the faces. Figure 3-59 shows the predicted ASE multiplier with a refractive index mismatch Δn of 0.01 (glass index = 1.52) for a 46-cm disk with edges perpendicular to the faces. Here the ASE multiplier is plotted as a function of the product $\bar{\alpha}D$ of the volume-averaged gain coefficient and the characteristic major dimension of the gain region (long axis of the ellipse for 31.5-cm disks; short axis of the half ellipse for 46-cm disks). This figure also shows the ASE multiplier predicted for a similar disk with edges tilted at an angle of 1.5° relative to the normal (in a parallel orientation) and the multiplier predicted for a perfect edge cladding.

The enhanced ASE multiplier for untilted edges is caused by rays that stay in regions adjacent to the disk faces (surface-mode rays). For the refractive index differences considered in this example, such rays would go into parasitic oscillation at a surface gain of 4.6×10^4 . This value corresponds to an $\bar{\alpha}D$ product of 3.6 for the surface-to-average gain coefficient ratio of 3.0 predicted for a neodymium-doping density of 2%. As the gain in a disk approaches this value, we see a dramatic increase in the predicted ASE multiplier, leading to a corresponding decrease in stored energy. The existence of such surface modes requires that the index match be extremely good when edges of the disk are not tilted.

In disks with tilted edges, those rays traveling along the disk surface are directed back into the disk, where they are

amplified by the thickness-averaged gain rather than the high gain coefficient along the surface. This situation leads to an increase in the threshold for parasitic oscillation as well as a decrease in the predicted ASE multiplier, as shown in Fig. 3-59. The sensitivity of the ASE multiplier to the magnitude of index mismatch is also greatly decreased with tilted edges, as shown in Fig. 3-60. Here the calculated ASE multiplier is given for several values of Δn . Even at the extremely large index mismatch of 0.2, the model predicts that the increase in the ASE multiplier at $\bar{\alpha}D = 2.5$ (corresponding to the maximum gain observed in the 46-cm disks) is only about 20%. The gain-scaling law with respect to the magnitude of M_{ASE} would then lead us to expect a reduction of peak gain by less than 5% from that produced by a perfectly clad disk.

Predicted ASE Decay Rates for Claddings with Surface Delaminations. The current fabrication method for disks with the composite cladding requires that the final machining and polishing of disk faces be performed after all of the cladding has been attached to laser glass. We have observed small delaminations (up to about 100 μm deep) at the exposed surface of the bond line, which we believe were generated during the final machining step. The effect of such delaminations is profoundly different with and without edge tilts.

Surface delaminations present the greatest problem for disks with edges that are not tilted. In this case, we can find a closed total internal reflection (TIR) path connecting any two opposing strip delaminations on opposite edges, regardless of their position with respect to disk faces. The only loss mechanism for an internal mode of this type is diffraction. A simple two-dimensional diffraction calculation indicates that, at the average gain coefficients expected in the large Nova disks, the width of the strip delamination would have to be less than 100 μm to prevent bulk-mode parasitic oscillation. An even more severe constraint exists for facing delaminations adjacent to the same face of the disk, due

Fig. 3-59. Predicted ASE multiplier for a 46-cm disk with polymer refractive index mismatch of 0.01 (glass index = 1.52).

Fig. 3-60. Variation of calculated ASE multiplier with gain level for a 46-cm disk at several values of polymer refractive index mismatch (edge tilt = 1.5°).

to increased gain of paths along the surface. Diffraction calculations for these modes, taking into account the normal-incidence Fresnel reflections at the edges, suggest that the maximum allowable size of delaminations in this instance is less than 10 μm (Ref. 78).

Tilting the edges of a disk relative to the normal to the faces tends to remove both of the difficulties discussed above. The results of a 2-D ray analysis suggest that with the introduction of edge tilts a closed TIR path connecting two, arbitrarily placed strip delaminations exists only for certain "bad" choices of tilt angle so that parasitic modes with only diffraction losses are no longer a major consideration. In addition, the edge tilt destroys the surface parasitic modes by redirecting rays traveling along faces into the bulk of the disk, where they experience greatly reduced gain. Delaminations will, however, still produce an increase in the ASE decay rate, which we have estimated using our numerical model.

In ASE calculations for delaminated claddings, we divided the disk edges into two regions: clad and delaminated. We assumed that the clad section was

Fig. 3-61. Plan views of a 31.5-cm disk with edges in (a) parallel and (b) trapezoidal orientations. Edge views showing vertical displacement of a surface ray for (c) parallel and (d) trapezoidal orientations.

perfect, absorbing all radiation incident upon it. In the delaminated regions, we used an effective reflectivity for the air/cladding interface with the same form as Eq. (19). The ASE rays were then tracked until they were either absorbed in the clad region or until their magnitude passed below the designated threshold. In all of the cases we will discuss, we made the extremely pessimistic

assumption that the surface delamination extended completely around the disk on both faces to a depth of 1 mm.

The first result of our analysis was the discovery that not only the magnitude but the relative orientation of tilts of the disk edges can affect predicted ASE decay rates. Figures 3-61(a) and (b) show two possible arrangements of tilt directions for 31.5-cm disks, which we designate as parallel and trapezoidal, respectively. Figure 3-62 shows the predicted ASE multiplier for both of these geometries, assuming a 1-mm delamination and a tilt angle of 0.8° . As the $\bar{\alpha}D$ product increases, the ASE multiplier grows more rapidly for the trapezoidal geometry, reaching a value greater than 10^4 at $\bar{\alpha}D = 3.5$, the maximum value expected in the disk. The ASE multiplier for the parallel edge orientation grows to a maximum of about 17.0, only about 3 to 4 times greater than the value predicted for a perfect cladding.

To explain the difference in the predicted ASE multiplier for these two geometries, we had to modify our Monte Carlo code so that, in addition to calculating the predicted decay rate multiplier, it also identified those rays that gave the greatest contribution to the ASE integral. As one might expect, for both geometries at large values of $\bar{\alpha}D$, the integrand was dominated by rays, which at some point traveled along one of the disk surfaces, generally oriented along the long axis of the gain ellipse. The difference between the predicted ASE multiplier for the two edge orientations can be understood by considering the ray paths pictured in an end view in Figs. 3-61(c) and (d). In both cases the incoming and outgoing rays travel parallel to the disk surface, retroreflecting off the two 45° edges. For the parallel geometry in Fig. 3-61(c), the ray passes directly from one 45° edge to the other; whereas in the trapezoidal geometry in Fig. 3-61(d), the ray must take one TIR bounce off the disk face between reflections off the corners. If either of these rays hits a delaminated region on both of the 45° faces, it will undergo TIR off both corners and make at least two passes along the long

Fig. 3-62. Calculated ASE multipliers for a 31.5-cm disk with a 1-mm-wide cladding delamination at disk faces and edge tilt angle = 0.8° .

Fig. 3-63. Variation of predicted ASE multiplier with tilt angle for a 31.5-cm disk with 1-mm surface delamination in the cladding bond for several values of disk gain. ASE multipliers for a perfect cladding are shown on the right.

axis of the gain medium close to the surface of the disk. For larger values of $\bar{\alpha}D$, the gain experienced by such rays will be enhanced by a considerable factor ($>10^4$) over that seen by those rays that can make only one pass along the surface before they are deflected toward the center of the laser disk or are absorbed by the cladding. For the specific geometry considered here (0.8° tilt, 1-mm delamination), such two-pass TIR face paths exist for the trapezoidal geometry, but not for the parallel geometry. This is the explanation for the larger ASE multipliers predicted in the former case. In general, the maximum allowable delamination at which the ASE multiplier remains below a given level is about twice as large for the trapezoidal orientation as it is for the parallel orientation. (It is interesting to note that the relative orientation of the directly opposing sides along the short or long axis of the ellipse has no significant affect on predicted the ASE multipliers.)

Figure 3-63 shows the variation of calculated ASE multiplier as a function of tilt angle for the 31.5-cm disk in the parallel geometry with a 1-mm delamination for several values of the gain parameter $\bar{\alpha}D$. The right side of the plot shows multipliers predicted for a perfect cladding. Solid lines are "eyeball" fits to data. At

values of $\bar{\alpha}D$ smaller than those considered here, the decay rates predicted for 1-mm delamination and a perfect cladding are the same. The increase in the ASE decay rate at low and high angles is caused by rays traveling along the long axis of the gain ellipse. For small angles, the expected surface mode oscillates along the long axis of the ellipse between two, facing edge delaminations adjacent to the same face of the disk. At an angle of about 1.95° , the tilt deflects a ray traveling along one face of the disk onto the delamination on the opposite face on the other edge, after which it makes another high-gain transit. Figure 3-64 shows the results of similar calculations for the 46-cm disk, where the orientation of edges is also illustrated.

For both disk sizes, the ASE multiplier has a broad minimum in the region around 1.5° , where the maximum predicted ASE multiplier is less than twice that predicted for a perfect cladding. Such a small increase (considering the power-law scaling in the predicted peak gain discussed above) in the predicted decay rate for such a severe delamination leads us to believe that tilting the edges in the manner shown will eliminate any degradations in disk performance resulting from cladding delaminations introduced during fabrication.

Fig. 3-64. Variation of predicted ASE multiplier with tilt angle for a 46-cm disk with 1-mm surface delamination in the cladding bond (parallel edge orientation) calculated at several values of disk gain. ASE multipliers for a perfect cladding are shown on the right.

Fig. 3-65. Variation of calculated ASE multiplier at peak gain with cladding absorption level for a 31.5-cm disk (parallel orientation, edge tilt angle = 0.8°).

Based on these results, we have chosen a 1.4° tilt angle for the 31.5- and 46-cm disks using the configurations shown in Figs. 3-61(a) and 3-64.

Predicted ASE Decay Rates for Claddings with a Finite $1\text{-}\mu\text{m}$ Absorption.

Reducing the product βt of the $1\text{-}\mu\text{m}$ absorption coefficient of cladding glass and its thickness also leads to an increase in total ASE decay. In this case, the amount of radiation returned back into the glass after two transits through the cladding and reflection off the cladding glass/air interface eventually becomes non-negligible. We are presently limited in modeling this effect by the lack of either a good model or experimental data for reflection of radiation off the inner surface of a ground cladding-glass/air interface. We can, however, attempt to obtain order-of-magnitude estimates by looking at the results of calculations using simple

models of the reflectivity that mimic some aspects of the case of interest.

We have looked at two different approximations for cladding/glass reflectivity. The first approximation is a specular model, which reflects each ray according to the polarization-averaged Fresnel reflectivity for a polished interface parallel to the laser/cladding glass interface. The second approximation is a diffuse model, which assumes that each ray is totally reflected, but returns with a direction that is uncorrelated with that at which it entered. In Monte Carlo calculations for both cases, we neglected the spatial translation of the ray in the cladding and assumed that the reflected ray reentered the laser glass at the point at which the incident ray hit the laser/cladding glass interface. Both the reflectivity of the cladding/air interface and the absorption of the cladding glass along the entrance and output paths were considered in determining the fraction of the ray that was returned to the laser medium. Our calculation of effective gain for each ray was halted, as before, when the magnitude of the ray passed below a threshold.

Figure 3-65 shows the results of both models for the 31.5-cm-disk, parallel-edge geometry with a tilt angle of 0.8° . Here we plot the predicted ASE multiplier at an $\bar{\alpha}D$ of 3.5 vs the cladding glass absorption parameter βt . With the

specular reflection model, a 2-D, TIR, bulk-parasitic mode exists along the long axis of the disk with a βl threshold of 1.75. Nevertheless, the calculated ASE multiplier at a βl product as low as 1.8 is only 10% greater than that of a perfect cladding. Insensitivity to the level of absorption in the cladding is a result of edge tilt, which causes rays to "walk" out of near-parasitic modes. The effect of transforming high gain paths into those with lower gain is also seen in the results of the diffuse reflectivity calculations. In this case, the predicted ASE multiplier does not differ from that for the perfect cladding for βl values as low as 1.8. In addition, as cladding absorption is decreased below this point, the predicted ASE multiplier increases relatively slowly rather than experiencing a sudden parasitic onset, as in the specular case.

Both of these calculations suggest that, in disks with tilted edges, the two-pass absorption in the cladding required to keep ASE decay rate at the minimum level is only slightly greater than the single-pass gain in the laser medium. Experiments supporting this conclusion are described in the article "Performance Tests of Prototype Edge Claddings," earlier in this section.

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Understanding and Measuring Adhesive Joint Strength.

Introduction. The complex state of stress encountered in typical testing geometries often makes it difficult to obtain an unambiguous value for the strength of adhesively bonded joints. Consequently, adhesively bonded systems are considered sensitive and unpredictable solely on the basis of poorly characterized testing procedures. At the engineering level, such ambiguity impairs confidence in these joints and probably accounts for the current preference for conventional mechanical fastening techniques.

In the following paragraphs, we examine the premise that adhesive joints are neither mysterious nor sensitive if the

geometry of the joint is considered with regard to the dimensional constraints it imposes on the adhesive. Moreover, we consider the adhesive layer a distinct region in the laminated structure, having properties determinable from bulk samples (i.e., strength, modulus, and moisture content) and obeying relatively simple failure criteria. The strength of a properly prepared joint is then determined by those conditions that result in cohesive failure of the adhesive.

Shear Strength of Adhesive Joints.

The primary test for determining the shear strength of adhesive joints is the tensile⁷⁹ or compressive⁸⁰ lap-shear test. Shear-strength results for candidate cladding polymers obtained using lap-shear compression were reported in the 1986 Laser Annual.⁸¹ A significant drawback to using this test routinely to evaluate cladding adhesive was tensile fracture of the phosphate glass adherends at 45° to the shear plane, which inevitably occurred at or before bond failure for the high-shear-strength epoxies. Since then, we have developed a torsional testing capability and have investigated alternative testing geometries that use this mode of loading.

Our initial work focused on evaluating the cone-and-plate torsion test, which produces a uniform state of shear in the adhesive,^{82,83} even under large strain conditions,⁸⁴ if the cone angle (θ) is less than 5°. Figure 3-66(a) shows the test geometry in which a shallow-angle stainless steel cone is bonded to a phosphate glass plate with the epoxy adhesive. The measured torque M_t required to twist the cone off the plate is simply related to the shear stress in the adhesive and the cone radius R through the relationship

$$\tau = 3M_t/2\pi R^3. \quad (20)$$

Initial studies using a 1.9-cm-diam cone with $\theta = 3^\circ$ produced enough torque to cause tensile fracture of the saw-cut phosphate glass plates prior to adhesive failure. To eliminate this problem, we reduced the cone diameter to 1.27 cm and used phosphate glass plates with finely ground edges. Bond shear strengths of 47 MPa were then obtained, without glass fracture, at a shear strain rate of 1.21/min for a 100:45 ratio of DER 332/T-403.

Fig. 3-66. Specimen geometry for (a) cone-and-plate and (b) tubular-butt-torsion shear-strength tests.

Unfortunately, attempts to obtain bond strengths for the current cladding epoxy (DER 332/RD2/T403/A399), using the cone-and-plate test, were frustrated by large hydrostatic stresses developed in the adhesive during cooldown from the 65°C curing temperature, which caused premature tensile failure of the joint.

The tensile stress on cooldown was a result of the axial and lateral constraints imposed on the adhesive by the shallow annular space and the contact of the cone and plate, as well as the fact that, unlike the DER 332/T-403 epoxy, the cladding epoxy was essentially an incompressible rubber above its glass-transition temperature (T_g) of 37°C. Using a typical Poisson's ratio of $\nu = 0.495$ and a volume thermal expansion coefficient of $\beta = 6 \times 10^{-4}/\text{K}$ for the rubbery epoxy at $T > T_g$, we would expect a cooldown from 65 to 37°C to produce a tensile stress normal to the glass surface equal to the mean hydrostatic tension, $p = [E\beta\Delta T/3(1 - 2\nu)] = 42 \text{ MPa}$, with $E = 7.5 \text{ MPa}$ the measured rubbery tensile modulus of the epoxy. Finite-element modeling of the stress distribution for this geometry⁸⁵ indicates that stresses of this order occur over the inner region of the bond, which agrees with visual observation, through the glass surface, of center delamination prior to testing. The development of these large hydrostatic tensions during the cure cycle of low T_g epoxies, when bond tensile strengths are expected to be low, is a severe limitation of this test—a point that has not been discussed in previous publications.⁸²⁻⁸⁴

Another method for measuring the shear strength of adhesive bonds in

torsion has found increasing use in recent years^{82,86-89} because of the uniform state of shear produced during the test and the low variability of the results. In this tubular-butt or "napkin-ring" torsion test, adhesive is used to bond a tube or ring to the substrate of interest that is subsequently twisted off in torsion as in the cone-and-plate test. However, the method has several advantages over the lap-shear and cone-and-plate specimen geometries. Radial variation of shear strain and, consequently, shear stress is minimized if the thickness d of the tube wall is small compared to the radius R of the tube. Under these conditions, the shear stress is essentially uniform and equal to

$$\tau = M_t/2\pi d(R)^2, \quad (21)$$

with $\langle R \rangle$ the average of the inner and outer tube radii. Shear strain γ is proportional to radius and the angle of twist in radians, θ , and inversely proportional to the epoxy thickness h through the relationship

$$\gamma = r\theta/h \approx \langle R \rangle \theta/h. \quad (22)$$

Extremely thin bonds can be produced with this method. Moreover, tensile stresses present on the edges of lap-shear specimens, which can lead to premature bond failure during loading, are avoided by the torsional deformation mode. Hydrostatic tensions that appear in cone-and-plate specimens during the cure cycle are absent because the adhesive bond is not constrained normal to the glass surface and is free to contract

during cooling. Preliminary tests using the tubular-butt torsion test to determine bond shear strength demonstrated that the method was not only extremely reproducible, but relatively immune to variations in bond thickness and bonding method and simple to perform with no evidence of phosphate glass fracture. Consequently, we adopted the test to determine the shear strength of the currently employed epoxy-phosphate glass bond.

Figure 3-66(b) shows the tubular-butt torsion-test geometry we used to determine adhesive-bond shear strength. Stainless steel tubes 6.4 cm long, with an outer diameter of 12.7 mm and a wall thickness of 1.24 mm, were cemented to polished 3.8- × 5- × 0.64-cm LHG-750 phosphate glass plates using the DER 332/RD2/T403/A399 cladding epoxy. A precision fixture was made to ensure that the tubes were aligned perpendicular to within $\pm 1^\circ$ of the glass surface during the bonding procedure.

Before bonding, the stainless steel tubes were vapor-degreased with Freon TF, washed with soap and water, wiped and rinsed with reagent-grade acetone or methanol, and dried at 60°C. The phosphate glass was washed with a surgical soap solution, rinsed and wiped with methanol, and dried at ambient temperature for 2 hr in a forced-air oven to simulate Nova clean-room procedures. Silane-treated phosphate glass samples were subsequently immersed for 30 min in a 1% solution of amino silane (Z6020, Dow Corning) in 4:1 water:methanol adjusted to pH4 with glacial acetic acid. Upon removal from the silane-treatment bath, the glass pieces were rinsed with deionized water, rinsed with methanol, and forced-air dried at ambient temperature for 2 hr.

The epoxy adhesive was mixed and degassed under mild vacuum to remove any air bubbles entrained during the mixing procedure. Then a small amount of the epoxy was applied to the end of the steel tube, which was butt-joined to the face of the phosphate glass plate in the bonding fixture. Epoxy that had squeezed out from underneath the tube was carefully removed with cotton swabs to leave a uniform layer of tube-wall width. The fixture containing ten replicate

samples was placed in an oven and cured for 24 hr at 65°C, after which the samples were allowed to stand at ambient conditions (30 to 40% RH, $T = 20^\circ\text{C}$) for 24 hr until testing.

Additional samples were prepared as above, using silane-treated glass to examine the effect of moisture on bond strength. After curing and 24 hr at ambient conditions, the samples were partially immersed in 20°C deionized water, exposing only the bond perimeter. After various lengths of immersion, individual samples were removed from the water, wiped dry, and tested immediately.

All samples were tested to failure using an Instron Model 1125 Universal Testing Machine in torsional mode. The slowest rotation speed obtainable (0.0101 rpm) was used for all tests. A fixture was designed to fit into the gear-driven lower chuck of the torsion apparatus, which would clamp and rotate the glass plate during the test. The steel tube was gripped in the fixed upper chuck, which was connected to a 200-N·m torsion-load cell. Yield stress and failure stress were calculated using Eq. (21) from the torque-twist curve for each sample. A profilometer was used to measure the bond thickness of the epoxy that remained on the glass surface after failure. The average strain rate in the adhesive layer for these tests was 9.1/min, calculated with Eq. (22) from the average bond thickness, $h = 40 \pm 16 \mu\text{m}$, mean tube radius, $\langle R \rangle = 5.73 \text{ mm}$, and angular velocity, $d\theta/dt = 0.0635 \text{ rad/min}$. This very high strain rate ($\sim 910\%/min$) was a necessary consequence of the thin bond and minimum testing speed obtainable.

Figure 3-67 shows torque-twist angle data for a typical test to failure of as-received epoxy. All samples of both untreated and silane-treated glass show essentially the same behavior. The epoxy exhibits approximately linear elastic behavior up to a shear strain of about 4 to 5%, at which point pronounced yielding and a drop in stress occur. Subsequent plastic flow proceeds with some strain hardening until failure occurs. This behavior is typical of ductile polymers and metals.

Table 3-7 shows the results of the tubular-butt torsion test for samples of

Fig. 3-67. Torque-twist curve for cladding epoxy in tubular-butt torsion test.

Table 3-7. Tubular-butt torsion-test results for samples of as-received cladding epoxy bonded to untreated and silane-treated LHG-750 phosphate glass. The coefficient of variation (standard deviation/mean) is in parentheses.

Substrate	<i>n</i>	Bond thickness (μm)	Yield stress (MPa)	Failure stress (MPa)	Failure strain	Failure surface
Untreated LHG-750	13	43 (41%)	33.8 (6%)	26.8 (6%)	0.95 (28%)	Glass/steel mixed
Silane-treated LHG-750	13	37 (41%)	33.2 (3%)	27.1 (6%)	0.78 (22%)	Steel only

Table 3-8. Results of tubular-butt torsion tests for bond shear strength vs time of immersion in water for silane-treated phosphate glass.

Immersion time (hr)	<i>n</i>	Yield stress	Failure stress	Failure strain	Nominal yield strength (%)	Failure surface
0	2	34	29	0.77	100	Steel only
1	2	30	26	0.66	88	Steel only
5	1	31	30	0.86	91	Steel only
20	1	29	30	0.62	85	Glass/steel
46	1	21	19	0.73	62	Glass/steel
70	1	17	18	0.77	50	Glass/steel

as-received cladding epoxy bonded to untreated and silane-treated LHG-750 phosphate glass. Note that the maximum stress—the yield stress of the epoxy—obtained in these tests was 36% greater than the 25-MPa value reported for lap-shear strength in the 1986 Laser Annual report.⁸¹ This discrepancy supports the widely held belief that tensile stresses occurring at the edges of lap-shear test specimens lead to premature bond failure and correspondingly low estimates of bond shear strength.

Visual observation of the locus of failure in the as-received epoxy test specimens showed mixed failure in the untreated glass samples occurring randomly at both the epoxy-steel and epoxy-glass interfaces. In sharp contrast,

the silane-treated glass samples exhibited failure exclusively at the epoxy-steel interface. While this change in the locus of failure suggests that the strength of the epoxy-phosphate glass bond was improved significantly by surface treatment with silane, Table 3-7 shows that the shear stress at yield for the as-received epoxy was higher than the failure stress of the interface. Thus, it is clear that the interface did not fail by shear alone, but rather by some combined state of shear and tension resulting from second-order effects occurring at the large strains obtained in these tests. Examples from the literature suggest that epoxies generally yield in shear, but fail or fracture in tension, much like the phosphate glass. Consequently, if premature failure at the steel-epoxy interface could be avoided in these tests, tensile fracture of the epoxy at 45° to the shear plane should occur for silane-treated glass substrates. This failure mode of cohesive tensile fracture was indeed observed for the 100:45 DER 332/T403 epoxy in the cone-and-plate test with tool-steel plates. The ultimate shear strength of the joint would then approach a value corresponding to the tensile strength of the epoxy if the interfacial strength is sufficiently high.

Table 3-8 shows the results of tubular-butt torsion tests for bond shear strength vs time of immersion in water for silane-treated phosphate glass. Although the number of replicates is small, the low coefficient of variation for these tests (6%), determined for the data in Table 3-7, ensures that differences of more than 30% between single sample values are real. The effect of moisture on bond strength is apparently plasticization of the epoxy, which lowers the internal viscosity⁹⁰ and reduces both the yield stress of the affected region and, to a lesser extent, the failure stress. Visual observations of the locus of failure for these tests show that failure occurs exclusively at the epoxy-steel interface for immersion times of less than 20 hr. Immersion for 20 hr or more results in mixed failure and a noticeable softening of the epoxy layer, which becomes easier to remove from the silane-treated glass surface. If we assume a moisture penetration depth *x* at time *t* normal to the bond

thickness given by $x = \sqrt{Dt}$, we can use a diffusion coefficient for water in epoxy at 100% RH and 24°C (Ref. 91), $D = 1.2 \times 10^{-3} \text{ cm}^2/\text{day}$, to calculate a value of $x = 0.6 \text{ mm}$ for the 70-hr exposure to water. This penetration depth corresponds to about half of the bond area, which agrees with the 50% reduction in epoxy yield stress observed for these conditions (see Table 3-8). However, an increase in the solvent activity of acidified silane solutions or commercial disk-finishing emulsions could accelerate this process of epoxy strength degradation by moisture uptake, resulting in proportionately weaker apparent joint strength.

The data of Fig. 3-67 and Tables 3-7 and 3-8 demonstrate that the shear yield strength of the epoxy determines the practical shear strength of the adhesive bond. Consequently, the epoxy fails cohesively (yields) before adhesive failure at the interface occurs, even in the case of untreated phosphate glass.

Bulk Epoxy Behavior in Shear. We conducted torsion experiments on solid cylinders of the cladding epoxy to determine the effect of aging time (moisture uptake) and strain rate on the shear modulus and shear yield strength of bulk specimens. The timescale for these aging experiments was chosen to coincide with available thermogravimetric analysis (TGA) data on moisture uptake and glass-transition temperature depression vs time at ambient for the cladding epoxy over 0 to 11 days (see "Improvements in Polymer Formulation and Properties"). The results of the TGA study can be described by the semi-empirical relationship

$$w(t) = 8.35 \times 10^{-3} \sqrt{t}, \quad (23)$$

with $w(t)$ being the weight fraction of moisture absorbed by the epoxy in time t (days) at 20°C and approximately 45% relative humidity. Under more carefully controlled conditions, the slope of $w(t)$ vs $\sqrt{t}$ can be used to obtain the diffusion coefficient for moisture in the epoxy using this procedure.⁹² The effect of moisture on the glass-transition temperature of the epoxy, T_g , is roughly approximated by a linear relationship typically used to

describe the initially large effect of low molecular weight plasticizers on T_g at low concentrations:

$$T_g(w) = T_g(0) - kw = 310 \text{ K} - 450 w. \quad (24)$$

The moisture coefficient, $k = 450 \text{ K}/w$, of the glass-transition temperature depression, from the zero-moisture value of 310 K, is within the 200-to-500 range generally observed for small molecules at low concentrations in high-molecular-weight linear polymers and epoxies.^{90,93}

The epoxy samples were cast in 1-cm³ polyethylene syringes and cured for 24 hr at 65°C. This casting produced uniform 4.65-mm-diam solid cylinders that were ~80 mm in length and had very smooth, flaw-free surfaces. After curing, the ends of the epoxy cylinders were bonded into aluminum sleeves for gripping in the torsional apparatus of the Instron. A 100:45 ratio of DER 332/T403 was used as the bonding agent, necessitating an additional 18 hr of cure at 60°C. Experiments showed that the additional 18-hr cure had no effect on the mechanical behavior of the cladding epoxy, indicating that the cure was essentially complete after 24 hr at 65°C. In the first of three sets of samples prepared in this way, the liquid epoxy components were mixed and degassed under high vacuum (~70 mTorr) before curing to remove water (partial pressure 18 Torr at 20°C). Violent boiling observed at approximately 2 Torr confirmed the presence of some bound moisture in the as-received components. The degassed epoxy was then drawn into the syringe and capped to ensure an airtight seal during cure. After the end tabs were bonded, the samples were immediately transferred to a desiccator and held for 24 hr. To ensure that no water could diffuse into them, individual samples were removed at the instant of testing. The two additional sets of samples prepared were degassed under mild vacuum only to remove air bubbles entrained during mixing. These two sets of samples were cured and bonded as usual and allowed to sit at ambient conditions for one or ten days prior to testing.

Duplicate solid-cylinder samples from the 0-, 1-, and 10-day ambient aging

groups were tested in torsion on the Model 1125 Instron Universal Testing Machine at angular test velocities ranging from 0.0635 to 635 rad/min to obtain values for shear modulus G and shear yield stress τ_y over four decades of strain rate. Since strain and strain rate vary linearly from zero at the center of the cylinder to a maximum at the outer radius, the observed torque-twist curve at large deformation is a summation of linear elastic and plastic stresses, the relative importance of each being a function of the stress-strain response of the material. In general, the torque is given by

$$M_t = 2\pi \int_0^R \tau(r) r^2 dr. \quad (25)$$

Assuming a linear-elastic-perfectly-plastic response, i.e., linear elastic loading followed by a constant shear yield stress, Eq. (25) becomes, after the entire cylinder cross section has yielded,⁹⁴

$$\tau_y = 3M_t/2\pi R^3, \quad (26)$$

with R the outer radius of the solid cylinder. To test the validity of Eq. (26) for a solid cylinder of cladding epoxy that exhibits a nonconstant shear yield stress, we converted the data of Fig. 3-67 to a stress-strain curve at a particular radius, $r = \langle R \rangle$, using Eqs. (21) and (22). Then we fit the resulting shear stress-strain curve to an analytical expression, using the maximum yield stress of the epoxy as the only adjustable parameter. By integrating the resulting analytic expression numerically over the cylinder radius, via Eq. (25), we obtained an expression for the torque-twist angle curve of the solid cylinder that accurately reproduced the measured response at a comparable strain rate and aging time (one day). We found that the adjustable maximum yield stress required to superimpose the calculated and measured torque-twist curves was within a few percent of the maximum yield stress calculated from Eq. (26) using the peak torque at yield. These results confirmed two important points: 1) the epoxy in the 40- μ m-thick adhesive bond apparently exhibits mechanical

behavior identical to a bulk sample having centimeter dimensions, and 2) Eq. (26) gives an accurate estimate of the maximum yield stress for solid cylinders exhibiting a yield stress drop if the peak torque at yield is used in the calculation.

Shear moduli calculated from the linear portion of the torque-twist curves at small strain for the three sets of samples showed a strain-rate-dependent decrease over the 0- to 10-day ambient aging interval. However, we found that superposition of the moduli for all the aging times could be achieved by a shifting procedure commonly used to correlate the mechanical properties of linear viscoelastic polymers obtained at different testing rates and temperatures. This procedure of "time-temperature superposition" amounts to a horizontal shifting of the isothermal moduli data along the strain-rate axis, relative to some reference temperature, until a continuous "master curve" of modulus vs strain rate is obtained. The glass-transition temperature of the polymer, T_g , is typically chosen as the reference temperature about which all the data at higher or lower temperatures are shifted. On a linear plot, this amounts to multiplying the actual strain rate for shifted data by a dimensionless factor a_T (adding $\log a_T$ to $\log$ strain rate), the value of which is given in good approximation for a wide range of linear and crosslinked polymers by⁹⁹

$$\log a_T = -C_1(T - T_g)/(C_2 + T - T_g). \quad (27)$$

In the torsion experiments, we held the test temperature T in Eq. (27) constant at 293 K so that the shift factors required to superimpose aging-time data for the moduli did not reflect a change in the test temperature relative to the glass-transition temperature. What occurred instead was the equivalent effect of a change in the glass-transition temperature at constant test temperature, brought about by the absorption of moisture during ambient aging. By substituting Eq. (24) for glass-transition temperature vs moisture content in Eq. (27), we can obtain the complete expression for the shift factor in terms of test temperature

and moisture content w . For the conditions of the current test, $T = 293$ K and, with $C_1 = 19$, $C_2 = 125$ K determined for the cladding epoxy from dynamic mechanical experiments using a mechanical spectrometer,

$$\log a_{TW} = (323 - 8550 w) / 108 + 450 w. \quad (28)$$

Figure 3-68 shows the shifting procedure and resulting master curve for torsional shear modulus relative to the zero-moisture (0-day) reference configuration—i.e., a dry sample with $T_g = 310$ K. The shift factors obtained in constructing the shear modulus master curve are: $\log a_{TW} = 2.10$ and 0.95 for the 1- and 10-day samples, respectively, with $\log a_{TW} = 2.99$ defined by Eq. (27) or (28) for the 0-day, zero-moisture samples. Although the moisture content of these samples was not determined directly, the measured values for the shift factors, together with Eq. (28), suggest the average weight fraction of moisture in the 1- and 10-day sample sets was 0.010 and 0.024, respectively, with zero moisture assumed for the degassed, desiccated samples. These calculated moisture levels are in reasonable agreement with values of $w = 0.008$ and 0.026 estimated for 1- and 10-day ambient exposure from the TGA data via Eq. (23). The effect of moisture on epoxy mechanical properties at a given test temperature is therefore shown to be equivalent to the effect of temperature at fixed moisture content—i.e., a decrease in the viscosity of the polymer matrix. This is apparent since both effects can be accounted for by using the shift factor, Eq. (27), which is derived from viscosity considerations.

Kinetic theories of activated flow processes assume that increased temperature facilitates segmental jumps in the direction of applied stress, as evidenced by a reduction of viscosity. The introduction of moisture into the polymer matrix should likewise facilitate these flow processes if the primary effect of moisture is to lower internal viscosity. The basic theory in this regard is the Eyring equation, which has been used widely to

Fig. 3-68. Shifting procedure and resulting torsional shear modulus master curve for cladding epoxy at 0-, 1-, and 10-day ambient aging, using 0 days as reference data.

describe the effect of temperature on the shear yield stress of ductile materials.^{95,96} At low temperatures and high strain rates, the Eyring equation can be written in terms of the temperature and strain rate $\dot{\gamma}$ as

$$\tau_y = 2kT/\nu \ln \dot{\gamma}/\dot{\Gamma} + 2U_0/\nu, \quad (29)$$

with k the Boltzman constant, ν a characteristic volume, U_0 the energy barrier to flow, and $\dot{\Gamma}$ a frequency factor. The shift factor of Eq. (27) can be interpreted in molecular terms as the ratio of the polymer relaxation time ζ at temperature T to that at temperature T_g , i.e., $\dot{\gamma}a_T = \zeta(T)/\zeta(T_g)$. Expressing the frequency factor in Eq. (29) as the reciprocal relaxation time of the epoxy, $\dot{\Gamma} = 1/\zeta(T) = \dot{\Gamma}_0/a_T$, gives, with some rearranging,

$$\tau_y = \tau_y^0 + B \log \dot{\gamma}a_T, \quad (30)$$

where τ_y^0 is the shear yield stress at $\dot{\gamma}a_T = 1/\text{min}$, with $B = 2303(2kT/\nu)$.

Original data for shear yield stress, calculated using Eq. (26), vs log strain rate showed three straight lines of equal slope but decreasing intercept over the 0-, 1-, and 10-day range of exposure to ambient conditions. Superposition of the three sets of data could be achieved, however, using the same shifting procedure and a_{TW} shift factors used to construct the shear modulus master curve (Fig. 3-68), in agreement with the prediction of Eq. (30). Figure 3-69 shows the linear-log master curve for shear yield

Fig. 3-69. Shear yield-stress master curve for cladding epoxy at 0-, 1-, and 10-day ambient aging, using 0 days as reference data.

stress vs strain rate, with the 0-day (dry) samples used as the reference data. The data of Fig. 3-69 show that moisture in the range 0 to 2.5% significantly lowers the yield stress of the epoxy at a given strain rate, having the same effect as an increase in the test temperature of about 12 K for dry samples. A linear fit of the data in Fig. 3-69 gives the parameters for Eq. (30), providing the relationship between the shear yield strength of the epoxy, strain rate, moisture content, and test temperature as

$$\tau_y \text{ (MPa)} = 10.729 + 6.544 \log \dot{\gamma} a_{TW} \quad (31)$$

The slope from Eq. (31) is 6.544 MPa = $B = 2.303(2kT/v)$, which gives a value of $v = 2900 \text{ \AA}^3$. This value is in general agreement with characteristic volumes of 2700 to 6400 $\text{\AA}^3$ obtained for glassy polymers from measurements of yield stress vs log strain rate at various temperatures.^{95,96}

To test the validity of using Eqs. (28) and (31) to estimate the yield stress of the thin epoxy layer in an epoxy-phosphate glass joint for various conditions of moisture, temperature, and deformation rate, we note that the reasonable assumption of 0.5% moisture in the as-received epoxy components leads to the correct value of 34 MPa for the yield strength of the epoxy layer under the conditions of the tubular-butt torsion test.

In summary, we have found that the isothermal yield stress of the cladding epoxy increases with increasing strain rate at a particular moisture level. However, increasing moisture at a given strain rate lowers the yield stress of the epoxy, as demonstrated in the tubular-butt torsion data for immersion time in water. The combined effect of temperature and moisture on epoxy yield strength is given analytical form by Eqs. (24), (27), and (31). The propensity for moisture uptake by epoxy polymers is a result of the potential for hydrogen bonding between water molecules and the hydroxyl groups generated during the epoxy cure reaction. Studies have also shown a direct relationship between equilibrium moisture uptake and the mole fraction of backbone oxygen in the epoxy precursors.⁹⁷ It should be noted that the cladding epoxy has an unusually high mole fraction of backbone oxygen because of the polyether triamine curing agent present at almost 30% w/w. However, the same hydrogen bonding interactions that result in moisture uptake also promote adhesion to highly polar substrates such as glass, making epoxy the adhesive of choice for such applications.

Tensile Strength of Adhesive Joints.

Butt-Tensile Specimens. The strength of thin bondline joints in simple tension cannot be estimated from butt-tensile specimens because of lateral constraints imposed on the adhesive by attachment to the rigid adherends that cause the state of stress in butt-tensile joints to change from uniaxial to triaxial tension as bond thickness approaches zero. For this reason, severe bond-thickness effects are observed in thin bondline, butt-tensile tests.^{98,99} In triaxial tension, three principal stress components act on the adhesive instead of the single tensile stress that defines a state of simple tension.

To estimate the strength of a thin adhesive bond subjected to a multiaxial stress state, we need a failure criterion that provides the condition for failure (yielding) in a state of combined stress. The simplest failure criterion known to provide an accurate description of

yielding in ductile polymers and metals is the distortional-energy or Von Mises criterion.⁹⁴⁻⁹⁶ Basically, this criterion states that hydrostatic stress has no effect on failure, so that only deviatoric (distortional) stresses contribute to failure. Therefore, the condition for yielding is determined by some critical value of distortional energy. For an isotropic, linear elastic material, this failure criterion may be written in terms of the three principal stresses and the measured yield stress in simple tension, σ_y , as

$$(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_1 - \sigma_3)^2 = 2\sigma_y^2. \quad (32)$$

Recognizing that simple shear is equivalent to equal orthogonal tensions of opposite sign, inclined at 45° to the shear plane, we note that $\tau = \sigma_1 = -\sigma_2$, $\sigma_3 = 0$, and that Eq. (32) shows $\sigma_y/\tau_y = \sqrt{3}$. This agrees with the experimentally determined ratio of tensile-to-shear yield stress of 1.6 to 1.8 for the 100:45 DER 332-T-403 epoxy,¹⁰⁰ demonstrating that the epoxy obeys the simple distortional-energy yield criterion of Eq. (32). Assuming similar behavior for the lower T_g cladding epoxy, we can use Eq. (31) to estimate the tensile yield stress for various conditions of moisture, strain rate, and temperature as $\sigma_y = \sqrt{3} \tau_y$.

The strength of a glass-epoxy-glass joint tested in tension can now be determined with Eq. (32) and some simple assumptions about the state of strain in thin bondline specimens. For axisymmetric butt-tensile specimens, where the adhesive thickness h is small relative to the lateral dimensions of the adherend d (i.e., $h \ll d$), we assume that the adhesive having Young's modulus E and Poisson's ratio ν is constrained laterally by attachment to the rigid adherends having E' , ν' . Then, lateral strain in the adhesive layer for tensile loading in the axial (or 1) direction must equal the lateral strain of the adherends, so the linear strains are $\epsilon_2 = \epsilon_3 = \epsilon'_2 = \epsilon'_3 = -\nu'\sigma_1/E'$. Assuming linear elastic

behavior up to the point of failure, the stresses corresponding to this state of strain are

$$\sigma_1 = \sigma_b, \sigma_2 = \sigma_3 = \frac{\nu\sigma_b}{(1 + \nu)(1 - 2\nu)} \left[1 - \frac{\nu'}{\nu} \frac{E}{E'} \right]. \quad (33)$$

If we substitute these stresses into Eq. (32) and solve for the axial stress at break σ_b , we obtain, as h/d approaches zero,

$$\sigma_b = \sigma_y \left[\frac{1 + \lambda/E}{1 + \lambda(\nu'/\nu E')} \right], \quad (34)$$

with $\lambda = E\nu/(1 + \nu)(1 - 2\nu)$, the Lamé constant for the adhesive. For $E = E'$ and $\nu = \nu'$, Eq. (34) reduces to $\sigma_b = \sigma_y$ as required. However, for adherends that are rigid compared to the adhesive, like the phosphate glass-epoxy laminate, $E' \gg E$ and

$$\sigma_b = \sigma_y (1 + \lambda/E). \quad (35)$$

Consequently, a large hydrostatic component, which we assume has no effect on yielding, would be added to the axial stress if lateral contraction of the adhesive is prevented during tensile deformation of the joint.

We tested Eq. (34) on published data for butt-tensile strength vs bond thickness for both fractionated low-density polyethylene⁹⁸ and tin-lead eutectic solder⁹⁹ joints. Both sets of experimental data showed, for $h = d$, that the joint failed at a tensile stress equal to the yield stress, determined in simple tension, of the individual adhesive, i.e., $\sigma_b = \sigma_y$. However, at $h/d < 0.02$, breaking stress increased significantly with decreasing bond thickness. Extrapolation to zero bond thickness, where triaxial tension would be uniform over the cross section, gave $\sigma_b/\sigma_y = 2.6$ and 2.8 for the polyethylene and tin-lead-solder butt joints, respectively. These ratios are within 10% of those predicted from Eq. (34), using reported values for the individual adhesive and adherend properties. The agreement is particularly interesting in the case of the fractionated polyethylene butt joints since the

adhesion of the as-received polyethylene to the polished glass and steel adherends used was found to be poor for thin bondlines even after surface modification of the polyethylene. The author⁹⁸ of the study interpreted the discrepancy in butt tensile strength between the as-received and fractionated polyethylene as indicative of the presence of a weak boundary layer—the result of low-molecular-weight species at the interface—in the former samples. Fractional precipitation from a hot toluene solution apparently produced a pure, high-molecular-weight (fractionated) polyethylene that failed cohesively, regardless of substrate, in accordance with the simple distortional-energy failure criterion. These results for polyethylene joints indicate that the measured improvement in shear failure strength and peel strength, obtained in the present study by silane-treating the phosphate glass, may result from the elimination of a weak boundary layer at the epoxy-glass interface. Acoustic imaging studies have shown a correlation between surface treatment and boundary layer thickness in epoxy-aluminum adhesive joints.¹⁰¹

The conditions leading to Eq. (35) are fulfilled—with $h/d = 10^{-4}$ and $E'/E = 25$ —in the case of the edge cladding

prepared with silane-treated glass, where cohesive failure of the epoxy is expected. At strain rates comparable to those obtained for the tubular-butt torsion test, Eqs. (31) and (32) predict $\sigma_y = \sqrt{3} \tau_y = 59$ MPa. If a reasonable value of $\nu = 0.4$ is assumed for the epoxy, the estimated breaking stress of the cladding-epoxy bond loaded in tension at a strain rate of 9.1/min would be about 140 MPa. This is of the same order as the maximum tensile strength of phosphate glass, indicating that thin bondline, butt tensile tests simulating the cladding geometry should result in tensile fracture of the phosphate glass before adhesive failure. Limited experimental data confirm the observance of massive glass pullout in thin bondline, butt tensile specimens prepared with silane-treated phosphate glass, but indicate little evidence of interface failure. In contrast, untreated phosphate-glass butt joints exhibit predominately interfacial separation at lower stress with little or no glass fracture.

Peel Tests. To comply with the recommended ASTM procedure,¹⁰² we have adopted a 90°-angle peel test to evaluate adhesives and control the quality of Nova disk production. This test also provides a relatively simple way to study controlled tensile fracture without the boundary shear stresses present at other testing angles. Figure 3-70 shows test results obtained for 90° peeling of the cladding epoxy from untreated phosphate glass, as well as from pH10 and pH4 silane-treated phosphate glass. It is clear that treating the phosphate-glass surface with an acidic (pH4) solution of amino-silane coupling agent increases its peel strength nearly twofold over that of untreated glass at the specified crosshead testing speed of 1 mm/min. On the basis of these results, we adopted acid-silane surface treatment of the phosphate glass disks and cladding for Nova amplifier production.¹⁰³

To understand the relationship between glass surface treatment, epoxy mechanical behavior, and peel strength, we reexamined the peel test. Using linear elastic constitutive relationships for the epoxy, we performed finite-element modeling of the tensile stress distribution in the 90°-angle, quality-control peel

Fig. 3-70. Peel strength of 90° test specimens for cladding epoxy on untreated phosphate glass and on pH10 and pH4 silane-treated phosphate glass.

specimens.⁸⁵ We found that the cleavage or tensile stress in the epoxy approximates a damped harmonic wave that alternates between tension and compression away from the point of separation. This stress distribution disagrees with that reported in the 1986 Laser Annual Report,⁸¹ in which a monotonically decreasing tensile stress was assumed. However, the finite-element modeling results are in agreement with other such studies for laminated strips¹⁰⁴ and with analytic results obtained from a beam on an elastic foundation analysis.¹⁰⁵ Jouwersma¹⁰⁶ has extended the beam in an elastic foundation analysis to include the effect of the bending moment of the strip, m , on the resulting peel force F to obtain

$$F = b\sigma_b/2n(1 + nm), \quad (36)$$

where σ_b is the cleavage stress at the point the strip detaches from the substrate. The structural parameter n is given by $n = 1.32/a$ for quality-control specimens utilizing an epoxy strip with a single Young's modulus E , width $b = 25.4$ mm, and thickness $a = 0.5$ mm. An analytical expression for the bending moment of the strip,¹⁰⁷ $m = [E I (\sin\theta/\theta - \cos\theta)/F]^{1/2}$, in terms of the peel angle θ and moment of inertia of the strip $I = ba^3/12$, yields the final expression for peel strength, $P = F/b$, from Eq. (36) as

$$P + P^{1/2} \left[n^2 E I \left(\frac{\sin\theta}{\theta} - \cos\theta \right) / b \right]^{1/2} = \sigma_b / 2n. \quad (37)$$

Moment-dominated peeling of thick or rigid strips is accounted for explicitly through the bracketed term in Eq. (37). The three-dimensional state of strain corresponding to the 90° peel test is $\epsilon_1 = \epsilon_b$, $\epsilon_2 = 0$, $\epsilon_3 = -\nu\epsilon_b$ for a peel force acting in the axial (1) direction normal to the 2,3 plane of the substrate surface, with lateral contraction of the strip in the width direction, specified as zero because of the attachment to the rigid glass substrate. If the cleavage stress at failure satisfies the yield criteria of Eq. (32), the linear elastic stresses can be substituted for the assumed state of strain with $\nu = 0.4$ to get the result $\sigma_b = 3\tau_y$. Using this

value as the cleavage stress at break, one can obtain the steady-state peel strength as the root of Eq. (37).

We can now use Eqs. (24), (27), (31), and (37) to estimate the effect of moisture, temperature, and testing rate on peel strength. Figure 3-71 shows the results of these calculations for the effect of moisture on the steady-state peel strength of quality-control specimens at $\theta = 90^\circ$ and $T = 293$ K. Agreement with experimentally observed variations in isothermal peel strength with aging time (moisture uptake) appears to be quite good, highlighting the importance of using a controlled environment to conduct quality-control tests. Under such test conditions, the calculated zero-moisture value for peel strength, $P = 24$ N/cm, is in excellent agreement with values of $P = 24$ to 28 N/cm obtained experimentally for dry specimens prepared with acid-silane-treated glass, suggesting that silane treatment improves adhesion to the point where cohesive failure of the epoxy occurs in the peel test. In fact, epoxy fracture in dry specimens is observed at peel forces of about 28 N/cm. The value of $P = 11$ N/cm for untreated phosphate glass, in contrast, implies failure at or near the interface prior to bulk epoxy yielding, in agreement with the visually clean separation of the epoxy strip from the glass observed for these test specimens.

Calculated results for the effect of test temperature on peel strength indicate that increasing the test temperature from 20 to 32°C, at a constant moisture and

Fig. 3-71. Calculated 90° peel strength of quality-control specimens vs epoxy moisture fraction.

strain rate, should reduce peel strength by approximately one-half, which is reasonably consistent with measured factors of one-third for a 45° peel test. A rate-dependent peel strength is also predicted from the effect of strain rate on epoxy yield strength. The observed factor-of-3 increase in 90° peel strength over the range 0.01 to 10 mm/min crosshead speed is on the order of 3.5, which was calculated for the same strain-rate interval. The large influence of moisture, temperature, and testing rate on epoxy yield strength and, consequently, peel strength is a direct result of the proximity of the glass-transition temperature to the ambient temperature.

Edge-Cladding Failure. The successful prediction of failure in peel and butt tensile adhesive joints, which are subjected to a combined stress state implies that the distortional-energy failure criterion [Eq. (32)] can be applied to the failure of composite edge cladding. For a single shear stress τ and tensile stress σ acting on the epoxy adhesive layer in the composite cladding during amplifier firing, the failure condition becomes

$$1/3 \sigma^2 + \tau^2 = \tau_y^2, \quad (38)$$

where τ_y at zero moisture is a function of temperature and strain rate, in accordance with Eq. (31). Finite-element modeling results (see "Modeling of Edge-Cladding Thermal Stress," earlier in this section) for the peak stresses that occur in the epoxy layer at the cladding edge during amplifier firing are $\sigma = 5.6$ MPa and $\tau = 6.8$ MPa for a 0.75/mm absorptivity cladding. Modeling results for the peak stresses in a 0.45/mm absorptivity cladding are $\sigma = 2.7$ MPa and $\tau = 4.2$ MPa. Substituting these peak stresses into Eq. (38) gives the minimum epoxy shear strength for cladding survival. The results are $\tau_y = 7.5$ and 4.5 MPa for the 0.75/ and 0.45/mm absorptivity claddings, respectively.

If we assume a characteristic strain rate of $\dot{\gamma} = \alpha \Delta T_{\max} / t$ for flashlamp pulsed heating, using $\alpha = 11.4 \times 10^{-6} / \text{K}$ as the linear thermal expansion coefficient of the glass, $\Delta T_{\max}$ as the maximum temperature rise at the epoxy-cladding interface, and $t = 1$ ms as the

duration of the flashlamp pulse, we can use Eqs. (27) and (31) to evaluate the shear yield stress of the epoxy during amplifier firing. The result for a 0.75/mm absorption coefficient cladding with $\Delta T_{\max} = 37$ K is $\tau_y = 3$ MPa, which is below the minimum calculated shear strength required for bond integrity. For the 0.45/mm absorption coefficient cladding selected for Nova amplifiers with $\Delta T_{\max} = 22$ K, the calculated shear yield stress during amplifier firing is $\tau_y = 14$ MPa, which exceeds the minimum shear strength of 4.5 MPa predicted for this cladding. The low shear strength of 3 MPa for the epoxy adjacent to the 0.75/mm cladding is the result, despite the high strain rate, of the peak temperature significantly exceeding the glass-transition temperature of the epoxy, causing the shear yield stress to rapidly approach zero. If we compare the calculated values for the shear yield stress of the epoxy during amplifier firing with the critical values estimated from the peak stresses and failure criteria, we predict that edge failures should occur on silane-treated disks for the 0.75/mm cladding, while the 0.45/mm cladding will provide a safety factor of about 3.

The predicted edge failure of 0.75/mm absorption coefficient cladding appears to be in general agreement with actual amplifier tests¹⁰³ on silane-treated disks utilizing this cladding, which show progressive edge delamination with continued firing. The situation for untreated disks may be considerably worse since delamination at the epoxy-glass interface can occur at stresses lower than those predicted from a failure criterion that assumes yielding of the adhesive.

Conclusions. A distortional-energy yield criterion applied to the epoxy layer can be used to predict the failure of adhesive joints under multiaxial loading if the strength of the interface equals or exceeds the cohesive strength of the epoxy. Silane-treated phosphate glass joints appear to fulfill this condition, while untreated glass joints are suspected of premature failure at or near the epoxy-glass interface under tensile loading. We postulate that the observed increase in bond strength for silane-treated glass may be due to the elimination of a weak

boundary layer at the epoxy-glass interface, thereby ensuring cohesive failure (yielding) of the epoxy. By determining the effect of moisture, strain rate, and temperature on the yield strength of the epoxy adhesive, we obtained the information needed to predict composite edge cladding failure using the yield criterion. The results of these calculations are in reasonable agreement with experimental failure data obtained for various edge-cladding configurations.

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Proof-Test Facility. We are using a new gain-test facility to both life test the designs for our prototype polymer cladding and to proof test production disks for Nova. Life testing required that prototype claddings be subjected to thousands of shots at Nova operating levels. Proof testing checks for infant mortality as part of the quality assurance program, determines whether amplifier gain is within the expected range, provides information to compare with Nova results, and provides a basis for measuring any change in performance after an amplifier is brought off line. To meet such new needs, we constructed our new gain-test facility adjacent to the clean room that is used for assembling and maintaining Nova amplifiers.

Our new facility comprises two rooms, one in which the amplifier is tested and the other that functions as a control room. The amplifier test room was constructed as a class-10 000 clean room, allowing us to move amplifiers directly into and out of the test facility from the clean room via a pass-through in the adjoining wall. Thus, we can determine amplifier performance without leaving the clean room environment, a procedure that saves considerable effort should amplifier modifications be required.

We will test all of the large power amplifiers—fifty 31-cm amplifiers and forty 46-cm amplifiers—for at least a few shots and several for life tests of more than 1000 shots. Because the strength of the edge-cladding bond to the disk decreases with temperature, the maximum temperature of the disks is kept below 25°C throughout a test. Adequate cooling of

the disks between shots takes at least 20 min with the N₂ flow rate available (50 cfm). The combination of many shots per amplifier, many amplifiers, and an expected low shot rate suggested many months of testing. Consequently, we designed the facility to run in an automatic mode (i.e., unmanned) 24 hr/day.

The amplifier proof-test facility is comprised of three major subsystems: power conditioning, an optical setup, and diagnostics. Power conditioning provides the control, charging, and firing of the 600-kJ capacitor bank (located in the basement of Building 391), safety interlocks (for both personnel and equipment), and other control functions (for both itself and the other systems). The bank can be manually switched to accommodate either 31- or 46-cm amplifiers, but otherwise is built using standard Nova hardware. One 50-kV power supply charges both the main bank and the pulse-ionization lamp check (PILC) system. Current sensors monitor all circuits for proper current levels and detect both flashlamp-to-reflector and ground-current faults. Improper current levels detected by the control system cause the system to abort.

The optical system uses gain measurement and analysis techniques^{108,109} we developed previously for use in the amplifier development laboratory in Building 174. As shown in Fig. 3-72, the optical system incorporates a Nd:YLF probe laser and power supply along with various optical components and detectors. We measure double-pass gain G at a wavelength of 1053 nm along two beamlines symmetrically displaced ± 6.2 cm from the amplifier centerline. We selected the positions to accommodate the split-disk geometry of the 46-cm amplifier. Double passing is operationally convenient and allows us to mount all optical components at one end of the amplifier, with the exception of two mirrors held on easily replaceable kinematic mounts. The two beamlines accommodate both amplifier types with no change in diagnostic setup. By mounting the amplifier and optical system on separate tables, we minimized the effect of vibrational coupling and allowed amplifiers to be exchanged with minimal realignment.

Fig. 3-72. Diagram of the optical system for our proof-test facility used to acquire gain measurements.

The optical setup shown in Fig. 3-72 is relatively simple. We employ a reference signal to ratio out any change in the probe-laser level that may occur during the measurement time. Splitters are used to separate the input and output beams, which are nearly colinear. We discriminate between background (flashlamp light plus neodymium spontaneous emission) and probe laser light both spectrally, using gratings, and spatially, using lens-pinhole combinations. The silicon photodiode detectors are well baffled, and we reduce stray visible light by using an infrared-transmitting (RG-1000) color filter. Levels of electrical noise and background light average less than 0.5% of the total signal level on all channels. We also image the beam at the spatial-filter pinhole onto a charge-injection device (CID) camera to aid and check optical beam alignment.

The diagnostic system acquires data from several instruments and computes peak gain, which determines amplifier performance. An LSI-11/23 microcomputer with 30 Mbytes of hard-disk storage controls the system and communicates with the instruments via the general-purpose interface bus (GPB). Two Tektronix 5223 digital oscilloscopes acquire signals from the three detectors: two signal and one reference. By recording the reference signal on both instruments, we eliminated the need for precise timing between the instruments. An

optical probe measures the amplifier-disk temperature directly and is read out with a digital voltmeter. Communication to power conditioning is through a digital input/output port.

We paid careful attention to minimizing noise in the diagnostic system by isolating it from power conditioning and by preventing ground loops. With the exception of optical detectors and temperature probe, the entire diagnostic system resides in the control area.

Noise-free ac power is provided via an isolation transformer. All systems in the control and test area tie to a common point in the control area, which is then tied to ground in the capacitor-bank area in the basement. System triggers from power conditioning arrive over fiber-optic lines, while the optical detectors are isolated from the optical table. We protect the temperature-probe subsystem from laser-amplifier-induced noise by enclosing all low-level circuitry in conduit or shielded enclosures. The shielding is tied to ground at a single point in the diagnostic rack.

With the exception of safety interlocks, each system may be manually operated independently of the others. Decisions on safety, equipment performance, and test validity were included in the internal control logic of the power conditioning and diagnostic systems to allow fully automatic testing. Table 3-9 lists the system-abort criteria and associated

hazards; failure of any criterion stops the test sequence. Hazards involve risk to personnel, to equipment, and to a test device that can be damaged by continued testing beyond initial failure.

Automatic operation requires that the power conditioning and diagnostic systems be linked so that each system will perform its tasks in the proper sequence. We implemented this link as a digital yes/no handshake involving three signals: power conditioning "ready," diagnostics "ready," and diagnostics "abort." The system can also fail-safe in that a given test stops when power is lost. One cycle of this sequence that accommodates both 31- and 46-cm amplifiers consists of two basic periods: a short period during which the amplifier is checked, the diagnostic system is readied, and the amplifier is tested; and a longer period during which the amplifier is allowed to cool.

Power conditioning first checks the condition of the flashlamps. If any lamp fails the check, the test terminates immediately. If the check is successful, then power conditioning asserts a ready signal that causes the diagnostic system to prepare for acquisition of baseline data. After a predetermined delay, power conditioning generates a trigger signal that causes the diagnostic system to acquire and process baseline data.

Diagnostics then checks the temperature of the amplifier. If the temperature is too high, the test terminates; otherwise, a diagnostic ready signal is asserted. This signal, in conjunction with the power conditioning completion of the baseline sequence, allows power conditioning to open the laser shutter and begin charging the main bank. At the same time, the diagnostics system prepares the instruments for acquisition of a gain signal.

When the bank is fully charged, the power conditioning system fires the system, closes the laser shutter, and removes its ready signal. When the oscilloscopes have been triggered, data acquisition and processing begin, and the diagnostics ready signal is removed. If either gain channel is low or the continuous-wave (cw) laser level is low, an abort signal is generated, and the test terminates. If no failures are detected, the systems

Table 3-9. System-abort criteria.

Failure or hazard	Factor at risk	Responsible system
Flashlamp failure	Equipment	Power conditioning
Low N ₂ flow	Equipment	Power conditioning
Low room O ₂	Personnel	Power conditioning
High voltage	Personnel	Power conditioning
Laser hazard	Personnel	Power conditioning
Bank fire	Personnel/equipment	Power conditioning
Low amplifier gain	Equipment	Diagnostics
High amplifier temperature	Equipment	Diagnostics
Low cw laser power	Test failure	Diagnostics

are reset, and the sequence recycles after the predetermined period for amplifier cooling.

We specifically designed our software to be simple to use. The program is menu-driven, requiring operator interaction mainly for instrument and system setup. The software is written in Tektronix SPS Basic and consists of ten program modules. Two configuration files define the operating parameters of the system and the oscilloscopes.

Gain data are stored in two files: a permanent shot-summary file for each amplifier test series, and a temporary gain-waveform file for each shot. We save the last 200 waveforms in round-robin fashion, which deletes the oldest of the stored waveform files each time one is saved. This procedure provides the operator with an opportunity to review the actual processed waveforms if summary data are questioned. The summary file includes information about the specific amplifier tested, the operator, comments, and a set of data characterizing performance on each shot in the test series. Data saved for each shot include the specific gain of each channel, the temperature of the amplifier, and the time of the shot.

Our new, automated gain-test facility has been operational since April 1987. We have operated the facility successfully for many thousands of shots, including all of our life tests (see the article "Life Testing Prototype Edge Claddings," earlier in this section), and we will use it to proof test all production disks before they are installed on Nova.

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Cladding Quality Assurance Program and Process Engineering.

Quality Assurance Plan. We described a quality assurance (QA) plan for the edge cladding of Nova replacement disks in the *1986 Laser Program Annual Report*.¹¹⁰ As we gained experience during the past year, we expanded and documented our QA plan. Table 3-10 is the Table of Contents of the resultant manual, *Nova Edge Cladding Quality Assurance Plan*. This manual contains detailed technical descriptions of:

- Materials certification tests performed at LLNL and vendor facilities.
- Material preparation.
- Optical fabrication procedures.
- Packaging and handling procedures.

Table 3-10. Table of Contents of Nova Edge Cladding Quality Assurance Plan.

Section	Title
I	Quality Assurance Plan
II	Acid Etch and Silane Agent Application
III	Cementing Edge Cladding
IV	Epoxy Bottle Filling Procedure
V	Peel Sample Preparation
VI	Polymer Certification
VII	Silane Certification
VIII	Quality Assurance Samples
IX	Production Quality Assurance Tests
X	Amplifier Test Procedure
XI	Quality Assurance Report A
XII	Engineering Drawings
XIII	Cladding Information Storage
XIV	Kodak Glass Finishing Procedure
XV	Zygo Glass Finishing Procedure
XVI	Edge Cladding Inspection Report
XVII	Peel Sample Test Procedure
XVIII	Semkit Mixing Instructions ^a
XIX	Semkit Filling Procedure ^a
XX	Disk Packaging and Handling

^a Sections XVIII and XIX were superseded by Section IV October 1987.

Fig. 3-73. Maximum acceptable relative humidity during and after cladding procedure vs ambient temperature.

- Engineering drawings.
- QA reports.
- Acceptance test procedures.

The manual has been extensively reviewed with our vendors and LLNL personnel to ensure clarity and correctness. The final form has been supplied to the cladding vendors and is now the reference document for all steps of the cladding process.

Process Engineering. Throughout the year, we made changes in the cladding process to streamline operations, to reduce cost where possible, and to increase reliability. We included all of these changes in the final QA manual. Examples of such modifications include:

- Changing the epoxy-cure cycle to produce greater mechanical strength in the cured adhesive.
- Including a surface-coupling treatment to the bonded glass surfaces to maximize glass-to-epoxy adhesive strength.
- Changing the epoxy packaging and dispensing processes.
- Adding several process controls at the vendor facilities during application of the cladding and during transportation and shipping of disks to reduce the degrading effects of atmospheric water.
- Finalizing the mechanical design and operation of clamping fixtures that hold the cladding during the time required to set the adhesive.
- Finalizing specifications for the surface finish of the laser disk edge and cladding, which indicate the type of cutting wheels, polishing grit size, and cooling fluids to be used during disk shaping and finishing steps.

As an example of our efforts, we expand on one of these process improvements—the inclusion of humidity controls. Humidity control to reduce the amount of atmospheric water present during application of the cladding greatly improves reliability of the strength of the adhesive bond. The importance of humidity was first realized from QA samples at one of the vendors when a sharp drop occurred in the epoxy/glass peel strength. Review of process records and subsequent tests at LLNL indicated that poor adhesion was related to high humidity during the time of cladding application.

Experiments indicated that high humidity causes accelerated weathering of the glass surface, lowering adhesion, and that water vapor swells the cladding epoxy, reducing its glass transition temperature and mechanical strength.

We specified an acid etch of glass surfaces to be bonded and humidity control during and after the bonding to eliminate these problems. We also specified that the finished disks be bagged and sealed with a desiccant during storage and transportation. Figure 3-73 shows the allowed humidity conditions arrived at as a result of this work. The process improvements were formulated and implemented as a result of the QA program, and the potential problem of cladding failure due to reduced adhesion was solved before any disks were fabricated for Nova.

Independent Evaluation of the QA Plan and Process Development. Independent evaluation of our QA plan and rigorous process control structure was carried out in the last year with the aid of an outside consulting firm, BDM Corp., Albuquerque, N. Mex. The role of BDM Corp. was to clarify manufacturing, storage, and handling procedures so that disk fabrication could proceed smoothly. As a result of interviews conducted by BDM Corp. with LLNL personnel, we identified six QA policy areas and goals, as follows:

1. Configuration management—to maintain an accurate knowledge of the configuration and initial condition of all Nova optics.

2. Total quality control (QC) responsibility—to establish the information, tools, and responsibility for total QC of manufacturing activities at each vendor organization.

3. Continuous improvement—to ensure that each vendor organization will strive to continuously improve the processes being used for production.

4. Acceptable quality level—to establish an acceptable quality level with an attendant confidence that such quality level has been met.

5. Logistics considerations—to subject all like elements to approximately identical environmental conditions during and after manufacture.

6. Vendor-base improvement—to ensure that LLNL promotes the development of a base of profitable vendors.

To accomplish the six goals, BDM Corp. suggested the establishment of certain QA procedures to supplement those developed by LLNL. The procedures dealt primarily with communication of process changes and production information, rather than technical design. With these additions to the QA Plan, the cycle of QA and process development was closed.

A report submitted to LLNL by BDM Corp., *Strategic Action Plan to Achieve Quality Assurance for Nova Laser Operations*, contains a comprehensive account of their recommendations. Figure 3-74 is a matrix taken from that report, showing

Fig. 3-74. Matrix of recommended procedures, listed in order, to accomplish the LLNL QA policies.

LLNL QA Policies	LLNL QA Procedures					
	Configuration management	Total QC responsibility	Continuous improvement	Acceptable quality level	Logistics considerations	Vendor base improvement
Quality assurance plan	X	X	X	X	X	X
Configuration management	X					
Surveillance						X
Data base design document	X	X	X	X	X	X
Dedicated shipping program				X		
Specification reviews		X				
Vendor QA advocate		X				
Vendor motivation		X				
Training		X				X
Experimentation			X		X	
Trip reports			X	X		
Common definition of terms		X				
Visual inspection criteria		X				
Acceptance examples		X				
LLNL optics storage					X	
Source inspection				X		X
Cause/effect diagrams		X	X			
Rework constraints		X				
Variance analysis			X			
Control charting		X	X	X		
Failure documents		X	X			
Project log			X	X		

the LLNL QA procedures recommended by the consultants to accomplish the six QA policy goals listed above.

Quality Assurance Data Tracking. We have installed a data base system, which will archive inspection and assembly data on each disk. Information is gathered from glass fabricators (Schott and Hoya), glass edge-cladding vendors (Zygo and Eastman Kodak), and assembly inspections at LLNL. This information includes all aspects of cladding and finishing operations, including disk maps that show imperfections in both the laser glass and cladding. Our tracking system is designed to follow the disks throughout their lifetime in the Nova laser. The number of shots, fluence, time, and position on line are details of the laser configuration that will also be recorded. Input to the data base system can be made from the Glass Storage Facility, clean room assembly areas, the Nova Operations Room, and the Engineering Office. Over 80 information entries are made on each disk by the time it is installed in the laser. In addition, we will use the data system to track other optical elements and laser amplifiers in the Nova system.

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Amplifier Gain Asymmetry Measurements on Nova

Nova output beams, as well as those of future ICF lasers, should be as uniform as possible to maximize output and to minimize the threat of damage. Previous measurements have shown that the gain profiles of most disk amplifiers vary by several percent across their apertures,^{111,112} giving a distinct high- and low-gain side to the amplifier. In addition, we have observed considerably greater platinum-inclusion damage to the disks on one side of the Nova 46-cm output amplifiers. Consequently, we conducted gain measurements on Nova to determine the extent and size of gain asymmetries on that

system. We also measured gain profiles of individual Nova amplifiers in off-line experiments to determine how to reduce the asymmetries.

Nova Measurements. We measured the gain asymmetry on Nova by propagating first one-half, then the other half of a low-energy input beam, measuring the input- and output-pulse energies for both cases. By energizing individual amplifier stages, we isolated the active and passive asymmetries of each stage. We used three beamlines, two of which had only four 31.5-cm amplifiers instead of the usual five, and we used (or shared) a total of 25 system shots. Table 3-11 shows the amplifier orientations for each of the Nova amplifier stages and the direction of the high-gain side of that stage after propagation to the 46-cm stage. Spatial filters between stages invert the beam profile, causing the differences in high-gain directions. All amplifiers within a stage were oriented the same way.

Table 3-12 summarizes the beam asymmetries from our Nova measurements. All numbers in the table are ratios of the gain on the side of the beam away from the frame to that on the side toward the frame at the 46-cm stage. Data entries are averages where more than one measurement was taken; in that case, an integer in parenthesis precedes the entry indicating the number of shots. Errors are standard deviations, rather than errors in the mean, which are smaller by the square root of the number of measurements. Those entries in parentheses are the products of other table entries; whereas those without parentheses come from independent measurements. For example, we missed some of the required data for an independent measurement of the power-stage ratio on beamline 8, and the table entry was obtained by multiplying the ratios for the 31.5- and 46-cm stages.

All but two of the gain asymmetries in Table 3-12 show the qualitative result expected from Table 3-11. For example, gain asymmetries for the driver stage in Table 3-12 are all less than 1, indicating higher gain toward the frame at the output, as expected from Table 3-11. The exceptions are the net and total gain

asymmetries for the 46-cm stage on beamline 3. The 46-cm amplifiers are oriented with their high gain sides toward the frame, which should give ratios of less than 1 as they are defined here. Different beamlines had similar numbers of disconnected lamp circuits (because of failed 46-cm amplifier lamps)—8 for both beamlines 3 and 5, and 9 for beamline 8—and the positions of the bad circuits in beamline 3 did not obviously dominate the observed ratio. Table 3-12 also shows that passive transmissions have no asymmetries (greater than the measurement errors). Consequently, we attribute the anomalous asymmetry for the beamline-3, 46-cm stage to measurement error.

Asymmetries for the net gain of the total chain (entries in the upper right-hand corner of Table 3-12) each indicate that the beam-half away from the frame would be the higher-intensity side of the beam if the input apertures to the chains were uniformly illuminated. Further, Table 3-12 shows that this asymmetry at the output results from incomplete compensation (by the driver and 46-cm stages together) of the asymmetry in the 31.5-cm stage.

Our data indicate that the average side-to-side asymmetry for the net gain of a complete chain is given by

$$\frac{(1.15 + 1.17)1.051 + 1.06}{3} = 1.17 \pm 0.10, \quad (39)$$

The multiplicative factor of 1.051 was obtained from off-line measurements discussed below. It accounts for the asymmetry of a single, centrally located 31.5-cm amplifier, since beamlines 3 and 5 had only four 31.5-cm amplifiers. The error bar is a guess based on the shot-to-shot reproducibility of the driver stage, where data from several shots were available.

Single Amplifier Measurements. We also measured the spatial gain profiles of single 31.5- and 46-cm amplifiers in an off-line test facility¹¹³ to determine how to correct the asymmetries observed on Nova. We previously showed that disk amplifiers with even numbers of disks have a side-to-side gain asymmetry caused by end effects.^{111,112} The side of the amplifier where the end disks project into the amplifier has a greater gain than the opposite side. However, flashlamp pumping from adjacent amplifiers influences

Table 3-11. Orientation of the high-gain (+) side of the Nova disk amplifiers with respect to the space frame.

Amplifier (cm)	+ side at mounting position relative to frame	+ side propagated to 46-cm stage relative to frame
9.4 (first)	Toward	Toward
9.4 (second)	Toward, down 45° ^a	Toward, down 45° ^a
15	Away	Toward
20.8	— ^b	— ^b
31.5	Toward	Away
46	Toward	Toward

^a For beamline 10 only, the second 9.4-cm amplifier is mounted with its + side away from the frame and up at 45°.

^b The 20.8-cm amplifiers have three disks and, therefore, no significant gain asymmetry.

Table 3-12. Summary of side-to-side gain measurements on Nova.^a

Beamline	9.4- and 15-cm amplifiers	20.8-cm amplifier	Driver stage	31.5-cm amplifier	46-cm amplifier	Power stage	Total chain
Net gain asymmetry							
3	(6) 0.85 ± 0.09	(4) 0.98 ± 0.06	(8) 0.80 ± 0.10	1.35	1.10	(2) 1.35 ± 0.07	(2) 1.15 ± 0.17
5	(7) 0.89 ± 0.05	(5) 1.04 ± 0.03	(8) 0.92 ± 0.09	1.34	0.93	1.24	(2) 1.17 ± 0.04
8	(5) 0.88 ± 0.06	(3) 1.01 ± 0.03	(8) 0.78 ± 0.09	1.37	0.99	(1.36)	(1.06)
Passive transmission asymmetry							
3	0.93	(2) 0.99 ± 0.03	0.94	—	—	0.98	(0.92)
5	1.05	(2) 1.02 ± 0.05	1.06	—	—	1.03	(1.09)
8	1.06	(2) 0.97 ± 0.07	1.05	—	—	1.03	(1.08)
Total gain asymmetry							
3	0.91	1.01	0.85	1.38	1.13	1.38	(1.21)
5	0.85	1.02	0.87	1.30	0.91	1.21	(1.08)
8	0.83	1.04	0.74	1.33	0.96	(1.28)	(1.01)

^a All entries = ratio of gain of beam-half away from frame divided by gain of beam-half toward frame in 46-cm stage

this end effect, so much so that end positions in an amplifier stage can have significantly different asymmetries than central positions.

To simulate pumping from an adjacent amplifier in our off-line measurements, we used a silvered endplate mounted at a position corresponding to the mid-plane

Fig. 3-75. Measured and projected gain profiles for three positions of a 31.5-cm Nova amplifier.

Fig. 3-76. Measured and projected gain profiles for three positions of a 46-cm Nova amplifier.

between amplifiers on Nova. For end-position amplifiers, we used one silvered endplate and one black-anodized endplate (more precisely, an end-box to reduce the flashlamp fluence on the black anodization). For central-stage positions, we extrapolated from measurements with black-black and silver-black endplates, since two silvered endplates would probably have caused parasitic lasing.

Figures 3-75 and 3-76 show measured and extrapolated gain profiles for the 31.5- and 46-cm amplifiers. The vertical dashed lines indicate the 10% of $I_{\max}$ points for a perfectly aligned beam. The average measurement error is about the size of the data symbols. Extrapolated gains for the case with two silvered endplates (Ag/Ag) were obtained from the gains with silver-black (Ag/BI) and black-black (BI/BI) endplates using

$$G(\text{Ag/Ag}) = \frac{G(\text{Ag/BI})^2}{G(\text{BI/BI})}. \quad (40)$$

These figures show that the effect of adjacent amplifiers is slight in the 31.5-cm stage, but quite significant in the 46-cm stage. We expected the smaller effect for the 31.5-cm stage because the separation of end disks in adjacent amplifiers is large relative to its clear aperture. Most of the separation results from the space required for electrodes of the longitudinally orientated flashlamps. On the other hand, the vertically oriented flashlamps of the 46-cm amplifier allow a considerably smaller separation-to-clear-aperture

ratio, and the effect of pumping from adjacent amplifiers is correspondingly higher. Figure 3-76 also shows the substantial increase in spatially averaged 46-cm gain caused by pumping from adjacent amplifiers.

To quantify the effect, we spatially averaged the gains over the entire beam area (between the 10% of $I_{\max}$ points) and over each beam-half. We used the data shown in Figs. 3-75 and 3-76 for the horizontal profiles and assumed uniform gain in the vertical direction. Table 3-13 gives the spatially averaged gains and the ratios of the average + and - side gains for each of the three mounting positions. The data in Table 3-13 show that pumping from the two adjacent amplifiers in a 46-cm amplifier stage (Ag/Ag) increases the average stored energy per amplifier by 11.2 and 5.4% compared to an isolated amplifier (BI/BI) and one adjacent amplifier (Ag/BI), respectively. Corresponding increases for the 31.5-cm amplifier are only 1.4 and 0.6%, respectively.

Table 3-14 compares the measured and predicted stage-gain asymmetries from the Nova and off-line data. These asymmetries are ratios of total gain (net gain

Table 3-13. Spatially averaged gains and side-to-side gain ratios (+ side divided by - side) for the three possible amplifier positions within an amplifier stage from single-amplifier measurements.

Endplate	Simulated position in amplifier stage	31.5-cm amplifier		46-cm amplifier	
		Average gain	Ratio	Average gain	Ratio
BI/BI	Single, isolated amp	1.716	1.063	1.585	1.069
Ag/BI	Either end	1.723	1.057	1.626	1.040
Ag/Ag	Central	1.729	1.051	1.669	1.013

Table 3-14. Comparison of side-to-side gain ratios from Nova and single-amplifier measurements.^a

Stage	Beamline	Number of amplifiers	Side-to-side gain ratios	
			Nova measurements	Predictions from single-amplifier measurements
31.5 cm	3	4	1.38	1.23
	5	4	1.30	1.23
	8	5	1.33	1.30
46 cm	3	4	1.13	0.90
	5	4	0.91	0.90
	8	4	0.96	0.90
Power stage	3	8	1.38 (1.56)	1.11
	5	8	1.21 (1.17)	1.11
	8	9	— (1.28)	1.18

^a Ratio = total gain for beam-half away from frame divided by total gain for beam-half toward frame, as would be measured in the 46-cm stage on Nova. Total gain = net gain/passive transmission.

divided by transmission loss), because the off-line experiments measure only total gain. As in Table 3-12, the Nova data represent separate measurements, except for those in parentheses, which are products of other entries. We obtained the predicted stage asymmetries by multiplying the appropriate single-amplifier asymmetries of Table 3-13. Clearly, the measured Nova gain asymmetries are consistently higher than the measured single-amplifier asymmetries. We have no explanation for this result, other than the large estimated errors for the Nova measurements. Given the error of $\pm 10\%$, most of the Nova and single-amplifier measurements are consistent.

Conclusion. Our Nova measurements clearly indicate that the side of the beam away from the frame at the 46-cm stage has the higher gain. This result is qualitatively supported by the observation of considerably greater platinum-inclusion damage to the disks on that side of the 46-cm amplifier. To reduce the asymmetry, we will reverse one of the 31.5-cm amplifiers when the platinum-free glass is installed on Nova so that its high-gain side is away from the frame.

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Nova Computer Systems

Information Systems for the Nova Facility

Information Systems for Nova are composed of three major computing facilities in which we perform data management and analysis, software development, and laser system controls. The three facilities are the

- Nova Laser Facility.
- Laser Experiments Analysis Facility (LEAF).
- Unclassified Laser Experiments Analysis Facility (UNLEAF).

All three computing centers are based on Digital Equipment Corporation (DEC) VAX computers and other hardware. The systems are supported by many gigabytes of disk storage and high-performance input/output equipment for graphics, image, and archival output.

Systems management, maintenance of hardware and software, and daily operation of these systems are some of the principal functions of Information Systems. We also support diverse software development for all types of applications that are best fielded by our mid-size computing systems.

New Group Structure for Information Systems. During the multiyear construction phase of the Nova project, the computer systems and our hardware and software development efforts were divided among separate working areas. The groups were logically organized along the lines of the project work breakdown structure, and we derived priorities from project-oriented management. Construction of the facility was the major activity, with emphasis on design and implementation and on meeting external milestones.

The facility is now complete, and our experimental program has been under way for some time. Thus, our priorities have shifted toward experiments-oriented management. Operation of the facility is now a major activity, with emphasis on maintenance, improving operability of the system, timely access to experimental results, and meeting programmatic milestones.

In response to the evolution of priorities from construction to experiments, we formed the Information Systems Group. The new organization derives its members largely from two groups that were involved in computer applications development during the Nova construction phase.

The first group, Nova Controls, was responsible for the VAX computers used in Nova operations and the control system for laser alignment.¹¹⁴ The hierarchical computer network, the Novanet local area network, the Praxis language, the console graphics system, and many other control system components used in

common by other project elements were developed by Nova Controls. The group also developed software systems for laser-diagnostics data acquisition and archival.

The second group, Data Management and Analysis (DMAG), was responsible for VAX computers and software used to process and archive experimental data produced by Nova target experiments. DMAG operated a photodensitometer for digitizing film-based data and had developed a significant capability for reducing raw data from film, streak cameras, and CAMAC-based data acquisition equipment. Reduced data are archived in the electronic SHOTBOOK, which is an information retrieval and display system based on a relational data base.

The Information Systems Group directly inherits most of responsibilities of the former two organizations. However, as its new name implies, we place a high priority on providing an integrated, facility-wide approach to information management. In particular, we are integrating computer-systems management for Nova operations and LEAF VAX computers. In addition, we are coordinating our data base management on the VAX systems to reduce duplication of effort and skills. A major thrust of the Information Systems Group is to improve access to data by experimenters and operators by increasing the connections of our distributed computing assets and by developing more user-friendly tools.

Author: P. J. Van Arsdaal

Facility Upgrades. This year the Information Systems Group made many improvements and upgrades to the computer systems they maintain. Here, we provide a short description of each system and the hardware and software changes we made during 1987.

The Laser Experiments Analysis Facility is the computer system used to analyze and store data from Nova experiments. The LEAF is made up of a VAX-11/785 computer with 28 megabytes (Mbytes) of memory and 4500 Mbytes of on-line disk storage; 2 high-speed,

high-density tape drives; 64 terminal connections; and 4 high-resolution graphics systems.

Our hardware upgrades for the LEAF computer in 1987 are as follows:

- An additional unibus adapter to speed graphics output.
- A network connection to the target-fabrication building providing access to users of the target data base.
- A laser printer for text and graphics.
- A terminal authenticator to allow users to access the computer outside normal working hours.

Our software improvements consist of upgrades to the:

- Operating system software (VMS version 4.5).
- Oracle data base software (v5.1.17).
- Graphics software from Computer Associates.
- IMSL, Inc., scientific software library.

We also added TEX typesetting software to the system so that users can produce documents including mathematical equations. Our new program to back up data from disk to tape operates faster and uses less tape than the previous program. We also installed a program that automatically compacts data on the disks. If such data are not compacted, disk performance decreases significantly.

The Unclassified Laser Experiments Analysis Facility is used to process unclassified experimental data. Figure 3-77 shows the computer systems the Information Systems Group provides. This system consists of a VAX-11/780 computer with 12 Mbytes of memory and 1234 Mbytes of on-line disk storage; a low-speed, high-density tape drive; 32 terminal connections; and 3 high-resolution graphics systems.

Our hardware upgrades included the addition of 546 Mbytes of disk storage, nearly doubling the previous capacity, and a high-density tape drive. The high-density drive will use one-fourth as much tape as the older system, cutting the time to back up the disk system by a factor of 4. The fast-backup and disk-compaction programs are now installed on the UNLEAF.

Fig. 3-77. Computer system provided by the Information Systems Group.

The Nova Development System shown in Fig. 3-77 is used to develop and test programs for Nova control computers, to capture and process interferometry data for the high-average-power laser, and to run modeling programs for the free-electron laser (FEL) project. The system has a VAX-11/780 computer with 16 Mbytes of memory and 3500 Mbytes of on-line disk storage; 2 high-speed, low-density tape drives; 72 terminal connections, and 1 graphics system.

The Nova Development System was connected to the unclassified LLNL network this year. This network has over 700 nodes and allows for easy transfer of information and programs from one part of LLNL to another. A laser printer to support the development of graphics software for the charge-coupled device (CCD) camera system was added to the computer. We also added four low-resolution graphics terminals to the computer to replace obsolete equipment. The operating system software VMS was upgraded to v4.5, and the ORACLE data base software was upgraded to v5.1.17. We purchased and installed a new

version of the DISSPLA graphics software package to support the new CCD camera diagnostic system.

The Nova laser computer-control system consists of three VAX-11/780 computers connected to the 105 LSI-11/73 minicomputers. The Information Systems Group is responsible for hardware and software on the VAX computers and 36 of the LSI-11/73 minicomputers. The LSI-11/73 computers are used to control the alignment system of the Nova laser. Each of the three VAX computers has 16 Mbytes of memory, 712 Mbytes of on-line disk storage, 24 terminal connections, and 2 graphics systems that are used as operator-control consoles. The three VAX computers are connected to 2 Mbytes of shared memory for high-speed data transfer and are connected via Ethernet to the Development System for data transfer and software update capabilities, as shown in Fig. 3-77. During the past year, we expanded the memory of each computer from 4 to 16 Mbytes to support new tasks. Two laser printers were added to the VAX, supporting target diagnostics and CCD cameras to provide

hard copies of graphics displays. A new version of the DISSPLA software graphics package was installed to support the laser printers, and the operating system software VMS was upgraded to v4.5.

The MicroVAX II workstation (VAX Station II) is being used to develop new hardware and software systems to support research in the Laser Engineering Division. The computer is a MicroVAX II with 9 Mbytes of memory, 225 Mbytes of on-line disk storage, 4 terminal connections, a cartridge tape system, and an 8-plane, high-resolution color graphics display. We upgraded VAX Station II from black and white to a color display this year and added 154 Mbytes of disk capacity. The VAX Station II is connected to the Development System by an asynchronous network line, as shown in Fig. 3-77. An array processor and image digitizer were installed on the VAX Station II to support acquisition and analysis of image data.

Author: W. G. Labiak

Computing-Resources Architecture: Networking.

Introduction. Our needs for computer resources have grown in recent years as ICF projects have expanded in scope, physical size, and distribution. A key consideration in formulating an ambitious plan to increase the computer power available to ICF researchers and engineers is computer accessibility. In the modern computing environment, accessibility is accomplished through networking and resource distribution. This year, we finalized plans for implementing a project-wide computer-resource distribution system, called ICFNET.

Programmatic computing needs have traditionally fit into three distinct categories:

- Cost-effective, small-scale, real-time computing in the laboratory environment.
- Personal computers in the office.
- High-throughput mainframes with large data-storage capacity applied to data analysis and control of our large laser facilities.

Until recently, the advantages of integrating our ICF computing resources have been unrealized, with a few exceptions.^{115,116} Integration of computer systems is not a problem unique to ICF. In fact, integration technology is currently the focal point for large-scale research efforts throughout the computer industry. Because of recent developments in the computer industry in the area of integration technology, cost-effective solutions are finally becoming available.

ICFNET addresses our programmatic computer needs through development and deployment of two modern technologies. First, we are providing industry-standard, reliable, high-bandwidth data-communication links to tie together our three computing environments. Second, we are developing a software infrastructure that:

- Applies highly effective menu-driven work stations, such as the Macintosh and the VAXstation, to the laboratory and office environment.
- Integrates these work stations with mainframes to form a computing-resources architecture. Industry-standard software is used within this infrastructure, where possible.

Sharing Computing Resources. The task of integrating computing resources is conceptually simple. Computers are connected by some communication media, and data that need to be shared are transmitted from one to the other. However, in practice, an extensive spectrum of applications emerges, which greatly benefits from a more robust hierarchical implementation of the concept of data sharing. The real power of integrating computer systems is best realized when the resources of any individual system in a network can be used by any task executing within the network. Thus, we seek resource sharing rather than simple data sharing.

Examples of shareable resources are compute engines (central processing units), disk and tape storage, data bases, special codes, compilers and software-development tools, graphics displays, video digitizers, printers, and other specialized hardware, such as array

processors and the Nova laser. Specific examples of shareable ICF resources include management and analysis software for the laser and target experiment data bases; general image-processing tools, such as the Interactive Data Language from Research Systems, Inc.¹¹⁷ and the VIEW signal and image-processing system¹¹⁸; and other programs that provide scientific visualization,¹¹⁹ simulation, and modeling.

ICFNET is the backbone of our project-wide architecture that supports such resource sharing. By strategically combining the flexible and interactive environment of personal work stations with the computing speed and data-storage capacity of larger computers, the strengths of both genre can be made available at almost any location.

The most difficult aspect of such integration stems from the fact that computer manufacturers have traditionally not devoted resources to developing standards for communication among

diverse products. This situation is changing, and ICFNET uses the most widely accepted industry-standard tools available.

Ethernet Local Area Networks.

ICFNET physically connects computers with equipment conforming to the Ethernet standard. Ethernet is the IEEE 802.3 standard for data communications within local area networks (LANs). The IEEE 802.3 standardizes the physical and data-link layers of the open systems interconnection model of the International Standards Organization and, as the lowest layers of that model, specifies a mechanism for operating a reliable and accurate communications path among nodes of multivendor LANs.

The principal advantage of using the IEEE standard is that numerous computer vendors of importance to ICF supply hardware and software that are compatible with Ethernet LANs. In addition, the existing Open Labnet LLNL-wide engineering network is based on

Fig. 3-78. Unclassified ICFNET resource-sharing network. A similar network supports processing of classified data. Ethernet transfers data at rates up to 10 Mbits/s and allows our computer resources to be shared without serious degradation in performance.

Ethernet, and Ethernet is incorporated into the new Livermore Laboratories Information Exchange telecommunications system.

This year we completed plans for installing Ethernet-compatible cabling in many key areas of the ICF complex, effectively expanding both the classified LEAF and the unclassified Nova Controls networks to laboratories and offices. Once installed, the backbone will provide the base upon which inexpensive future expansion can bring reliable, high-speed data communications to any location (see Fig. 3-78).

X Windowing System. While Ethernet provides the transport mechanism for sharing data among computers, the key element of our effort to integrate computers and to share resources is the X Window System software standard.¹²⁰ Version 11 of the X Windowing System, or X11, specifies the software architecture and protocols that provide a common, network-transparent application programming environment ideally suited for integrating the ICF computing environment.

The X11 standard, developed by Massachusetts Institute of Technology (MIT) in 1987, is a cooperative effort funded by the largest computer manufacturers in the U.S. It is embraced by every major computer vendor, all of which have, or are about to, announce products based in the MIT standard.

In its simplest form, X Windows allows a work-station user to interact with application programs running on any computer in the network through multiple display areas, called windows, on a work-station screen. Using the work station mouse, windows can be resized, relocated, and even iconified to make room on the screen for other applications. Examples of application windows are an alphanumeric terminal emulator for editing documents or entering programs, a window that displays color-enhanced image output from an image-processing algorithm, and a data-base spread-sheet window.

Of greater importance is that X11 provides a standard for developing an extensible toolkit of user-interface programming objects, called widgets, with which portable applications can be written. A

widget is a program object that provides a user-interface abstraction, such as a menu button or a scroll bar, and consists of a window and procedures for interacting with the applications program. Widgets can range in complexity from a simple pushbutton to an animated clock face with buttons for setting the time, alarm, and snooze.

A hierarchy of widgets can be collected into a toolkit. A toolkit of widgets can therefore be built to support a broad range of abstractions. Applications developed with the same toolkit will automatically exhibit a consistent look and feel to the user no matter what computer system is supporting them. Toolkits can be shared by applications, thereby eliminating redundant implementations and providing a uniform user interface throughout suites of related applications. For example, we will develop a single toolkit to provide the user interface to the collection of codes that analyze data from the Nova Electronic SHOTBOOK.¹²¹

The power of X Windows stems from three related features. First, X11 is a high-level standard. It provides a platform upon which user interfaces and applications programs can be integrated across different vendors' computers and operating systems.

Second, X11 implements the connection between the user interface of a program and its computing resources in a manner that is transparent to the user and that program. That is, an application can be run on any networked computer that supports the X11 standard, whether Macintosh or VAX, for example, while the user of that application can choose to interact with it from a favorite work station on any other computer. Because X11 is a high-level standard, programs can be written so that the part of the program interacting with a user is completely transportable among work stations. X Windows provides for the development of transportable programs by forcing the separation of the function of an application program from the form of the user interface. Separation of form and function is accomplished by the application of the widget concept and object-oriented programming techniques.

In practice, the X11 widget and toolkit are powerful, indeed. At the lowest level, widgets are implemented using intrinsic functions strictly specified in the standard. A user action, such as selecting a button from a menu of buttons, is mapped into a call to an application program by the widget that describes the button. Interaction among widgets and application program functions is implemented by object-oriented programming techniques. Therefore, a generic button widget can be defined wherever a button is needed to initiate a specific function. In other words, the form and behavior of the picture of a button on the workstation screen is specified by the widget program, whereas the function to be initiated by selection of a particular button is specified entirely by the application program.

Third, X11 provides a standard for formulating toolkits of user-interface abstractions or widgets. The efficiency with which users interact with computers can be greatly improved by providing coherent, hierarchical, easy-to-master interfaces. Widget hierarchies can allow a user to naturally extrapolate program-interface behavior from simple circumstances to more complex cases, eliminating the need for in-depth training and lengthy user guides.

Software vendors provide toolkits developed to represent the style of interface they feel is best suited for their system. However, their toolkits must adhere to the standard to be truly portable and usable, and they must also provide the intrinsic widget support inherent in the standard.

Disk and Data Sharing. ICFNET supports an effective commercial technology, called the Local Area VAX Cluster (LAVC),¹²² which uses Ethernet for sharing disk storage and software among Digital Equipment Corporation computers and work stations. Other computer vendors provide similar technologies that are also compatible with the ICFNET Ethernet and that we will use to accomplish resource sharing among most of our other ICF computers.

Each LAVC consists of from 1 to 26 cluster-node VAXs, usually MicroVAX work stations, supported by one or two

boot-node VAXs. The boot node is the only computer within the cluster that must be directly connected to storage disks. Each cluster-node computer shares disks of the boot node with the other computers of the cluster. Software on each cluster node simulates direct input and output of disk data, using Ethernet to transfer data to and from actual disks (see Fig. 3-79). Each cluster-node computer typically contains no disk, yet the LAVC software allows it to be functionally identical to a computer with a local disk.

The premier feature of this architecture is that only one copy of all software need be maintained, including the extensive software comprising the VMS operating system and its utilities. By implementing the LAVC, the power of a MicroVAX is available locally to each user while the cost of maintaining and managing VMS and applications software is significantly reduced, especially for clusters containing more than a few computers.

The LAVC is especially well-suited for providing distributed processing power in the ICF classified computing environment. The LEAF VAX is located in a secured vault and is used as the boot node, with cluster-node MicroVAXs located throughout the classified office and laboratory areas wherever a classified Ethernet cable has been installed. Because disk storage is centrally located in the vault, each diskless cluster computer need only be powered off when not in use. Ethernet connections to each computer are protected between regular working hours. Without the LAVC, the disks of each local computer would have to be relocated to a vault or safe when not in use.

Another advantage of the LAVC is that clustered disks represent a cost-effective method for providing large-capacity data storage for many work stations without significant performance losses. We are now considering technology similar to the LAVC for supporting clustered disks for IBM PC and Macintosh personal computers.

Conclusion. Our ICFNET system brings together industry-standard networking and microcomputer work-station technology in an architecture that increases the effectiveness of ICF computing by

Fig. 3-79. LAVC allows VAX computers to share centralized disk storage, eliminates maintaining data structures and software on individual computers, and is a simple mechanism for passing data among concurrently executing programs. High data-transfer rates are provided by Ethernet.

improving the accessibility and flexibility of computing resources. Our system is consistently and unilaterally integrating computing resources throughout ICF projects and opens the way for the program to join the modern age of computing. The most important factors in planning the ICFNET system are compatibility with existing systems, compliance with industry standards at every level, and extendability aimed at covering the future needs of ICF projects.

Author: J. E. Krammen

Image Processing Workshop. Image processing techniques are used by various projects within Nova and the ICF program. While developing these systems, it became apparent that there is a large, although disjointed, community of researchers developing and using image processing techniques throughout the

Laboratory. In fact, there has been a remarkable upsurge of interest in image processing technology in recent years, due in part to significant increases in computer processing capabilities at reduced cost.

For example, image processing is routinely used to enhance and analyze radiographic images used for destructive and nondestructive testing of weapons devices. Other LLNL applications include analysis of ultrasound chest scans (for Hazards Control), enhancement of degraded streak-camera images, automatic analysis of diagnostic oscilloscope photographs from weapons tests, analysis of radiographs from coal-gasification experiments, line-of-sight alignment, and processing of laser fusion diagnostic data.

Further investigation showed that there is great interest in improving cooperation among Laboratory projects involving image processing. In response to

this need, an Image Processing Workshop was cohosted by members of the Nova Information Systems Group, the Materials Analysis Group of the Nuclear Energy Systems Division, and the Nondestructive Engineering Section of Mechanical Engineering's Engineering Sciences Division.

The workshop was held on April 21 and 22, 1987, at LLNL under the joint sponsorship of the Electrical and Mechanical Engineering Imaging Thrust Areas and the Weapons Agency Nondestructive Testing Organization. The short-term goal of the workshop was to provide a forum in which researchers interested in image processing could identify common needs and limit duplication of effort. The longer-term goals of the workshop were to create a community of image processing users, to stimulate the application of image processing to Laboratory programs, and to advance the state of the art of image processing.

A total of 150 people attended the workshop, including representatives from LLNL, Los Alamos National Laboratory, EG&G Measurements Group, Sandia National Laboratories, Allied Bendix Aerospace, and the Oak Ridge Y-12 Project. The 29 technical presentations encompassed almost every aspect of image processing. Each presentation was limited to 20 min, including a question-and-answer period, to maximize the breadth of the workshop. Participants enthusiastically took advantage of the breaks between sessions to exchange more detailed information and to make arrangements for future discussions.

The workshop has had a positive impact on Laboratory projects by stimulating an exchange of technical concepts and techniques among researchers. For example, an engineering work station is being developed that will perform image processing and related computing in support of digital interferometry research for the FEL and advanced laser projects. This cooperative effort between the Information Systems Group of the Laser Engineering Division and the Engineering Research Division will use the VIEW image-processing package originally

developed by the Nuclear Engineering Systems Division in support of nondestructive testing applications.

In addition, many participants expressed a need for standards for image storage and transfer; they were made aware of the LLNL Multidimensional Signal Processing Working Group, which is addressing not only standards but also other image processing issues. Partially as a result of interest at the workshop, this previously informal organization is evolving into an image processing user's group.

A special report on the workshop has been prepared and includes papers representing a few of the many ways that researchers are using image processing to solve engineering problems here at LLNL and at the other laboratories that participated in the workshop.¹²³ The workshop was a success not only because of the quality and quantity of technical information exchanged but also because it brought together those interested in image processing to form a community.

Author: J. E. Krammen

Nova Operations and Laser Diagnostics

Nova On-Line and Development Computer Systems. The Nova laser is controlled by a network of on-line computers consisting of three VAX-11/780 computers connected to a network of 105 LSI-11/73 minicomputers. The three VAX computers provide the human interface to the system using touch-sensitive screens for input and color graphics for output. Our five major subsystems controlled by the VAX computers are:

- Power conditioning.
- Laser diagnostics.
- Alignment.
- Target diagnostics.
- Charged-coupled device (CCD)

streak-camera diagnostic (a new subsystem).

The Nova computer control system has been in operation for three years and is an essential part of the Nova laser

system. The new CCD streak-camera subsystem required installation of several upgrades to the VAX computers. Each of the three VAX computers had 4 Mbytes of memory. The computing load of the alignment VAX and the laser diagnostics/power conditioning VAX was so high that we encountered frequent delays in operation. The new CCD camera system would be installed on the target diagnostics VAX to distribute the load, but we anticipated performance problems.

We replaced the 4 Mbytes of memory in each computer with 16 Mbytes of memory and a new, faster controller. Now all tasks remain in memory, improving response time, and the disk access time during peak operations has been reduced from 800 to 80 ms. A complete alignment loop can be done in 45 min, compared to 65 min before the memory upgrade. Data from post-shot laser diagnostics are available in about two-thirds the original time, and the system clock does not stall during a shot sequence.

We added CCD streak-camera software to the target diagnostics VAX with little degradation to system performance. We added two laser printers to the computers to obtain hard copy of CCD camera streaks. We also installed additional terminal lines to allow remote operation of the streak cameras.

We use the Nova development computer system to develop and maintain software and hardware used on the on-line computer system, to acquire and analyze interferometry data for the Advanced Lasers project, and to support design and analysis programs for the FEL group. We have 26 users in the Nova group, 3 users in the Advanced Lasers project, and 19 in the FEL group.

During past years, we made some hardware upgrades to the development computer. First, we installed a connection to the unclassified LLNL computer network. This network connects to more than 700 on-site computers and is a gateway to the APRANET. The network is used to transfer electronic mail, data, and programs. We added a new laser printer to the computer for developing programs related to streak-camera

software and for providing a high-quality graphics-output device for image processing. We installed four, new VT-240 graphics terminals to replace old terminals that had failed and could no longer be repaired.

We made several changes in the use of peripherals on the development computer. To reduce maintenance costs, we removed from service three RP06 disk drives, each with a capacity of 176 Mbytes, and the LP11 drum line printer. The disk capacity was made up by using two RA81 disks with a capacity of 456 Mbytes each for data that were stored on the RP06 disks. The two RA81 disks had been used as backup disks. The FEL program was given one RA81 disk, and the Nova program used the other. Some programs that had been stored on the system disk were moved to the Nova disk to provide more space for the operating system and languages. Backup procedures for the system were changed to do weekly, incremental disk backups and complete image backups once a month because the backup RA81 disks are no longer available for backup. Previously, complete image backups were done once a week. The incremental backups are as reliable as image backups; thus, the contents of the disks are not in jeopardy.

Author: W. G. Labiak

Praxis Computer Language. We have made improvements to the Praxis computer programming language, which we use throughout the Nova Control System.^{124,125} We implemented a new interface to disk files and new command input/output features, which match the conventions of the DEC VMS operating system. The new file interface provides a complete set of operations for handling sequential, relative, and indexed files, the three types of files supported under the Records Management System of VMS.¹²⁶ The command input/output software allows users to employ the DEC standard-command-line syntax, with option switches for controlling the operation of their programs.

We improved the interface between application programs written in Praxis

and the DEC VMS operating system and reorganized the appropriate software libraries. The Praxis language, consisting of compilers and all support software, was revised and substantially reorganized to meet the Nova Software Quality Assurance Plan,¹²⁷ which, among other things, requires that the entire product be able to be recompiled and linked directly from program sources under control of a command file. To this end, we removed many circular dependencies within the sources of the compiler and support routines. By doing so, each module depends only on items that were defined within previously compiled modules in a linear list of modules comprising the entire Praxis product.

We added several features to the Praxis language, including extensions to exception handling (often used for reporting errors), further definitions of the use of flexible arrays, the ability to allocate large arrays, and the ability to define floating-point constants in mathematical expressions that are evaluated at compiling time.

We used the following four categories to compare Praxis to the recently developed Ada language and several older languages:

- General appearance.
- Power.
- Freedom from errors.
- Manageability.¹²⁸

We find that both Praxis and Ada have substantially improved software-engineering practices and programming capabilities over those of previous languages, such as Fortran, Basic, Pascal, and assembly language. The fundamental Praxis and Ada languages are similar, each having small areas of relative advantage. Praxis does not have built-in multitasking operations, which are already well developed within most operating systems, and it does not allow redefinition of identifiers or operators. Such design decisions made Praxis easier to learn, usable by less-skilled individuals, feasible on smaller machines (e.g., LSI-11), and yet efficient and manageable on large projects. Ada has immense support from the Department of Defense, is now widely taught in schools, has the yet-to-be-fulfilled potential of fostering a new market in shareable software, and

many computer aided software engineering tools are now commercially available to assist in programming and software project management.

An automatic compilation system for Praxis, which is similar to one developed by DEC for the Ada language, was designed and a prototype version was tested. It employs a topological search of a special graph, which is constructed to represent dependencies of the various pieces of software to compile software modules in the correct order in a large system whenever they need to be recompiled because of source revision. Such a technique, if widely used on Nova, would reduce the possibility of accidentally using obsolete pieces of software and would make large systems of software easier to understand.

We continued to release the Praxis language in object form into the public domain through both the Argonne National Software Distribution Center and the DEC User's Society. Through this release, we have established contacts with seven other organizations in the U.S. and Europe. Our contacts are actively using the language, have been helpful to us in suggesting and making improvements, and have supplied useful software to us in several cases. The organizations are as follows:

- Air Force Weapons Laboratory.
- Case Western Reserve University.
- Centre d'Etudes de Limeil in France.
- Digitech Information Systems in Austria.
- Fermi Laboratories.
- The National Bureau of Standards.
- The University of California at Davis.

Fourteen other organizations and individuals have seriously studied Praxis and have interacted with us for three purposes: because of the position of this language in the state of the art in computer languages, the consideration of its use in their applications, and the possibility of adapting Praxis to operate on other computer systems.

Author: F. W. Holloway

Nova Alignment System. We have made several changes and additions to the Nova alignment control system. We modified the stepping motor control

(SMC) computer software to use a motor encoder as feedback for positioning devices. The use of feedback control improves the repeatability of the control system. The positions of devices can now be displayed in engineering units as well as motor steps. We installed a second SMC in the master oscillator room to provide additional capacity for new controls.

The alignment dispatcher program, which runs on the alignment VAX computer, was modified to record information about a device each time its status changes. The information that we record includes encoder positions, error conditions, device limits, device positions, and the last command sent. The information allows operators to easily identify intermittent problems with devices. The dispatcher now allows operators to program a hold command into the configuration of a device. When a hold is encountered in a configuration file, the dispatcher displays the reason for the hold request on the console. The operator can perform other operations before continuing the configuration sequence by pressing the continue button.

We also made changes to the console alignment software. We added a new

display of the status of gate valves, target positioner, aids positioner, and target-plane imager. Figure 3-80 shows this display. We modified the target alignment viewer controls to operate the new target alignment viewer installed in the polar position on the target chamber. We added controls for front lighting, reticle lighting, zoom, and magnification. We also added control and status displays of the ceiling shutters in the master oscillator room and added control and status displays of the backlighter-beam block shutters to the preamp controls. A cross hair, which is no longer used, was removed from the output-alignment status controls.

The automatic alignment system has continued to be successful at routinely aligning the Nova laser in preparation for experiments. Almost 300 automatic loops are analyzed and corrected by computer without operator supervision.

Our system was designed so that the loop performance could be adjusted by modifying parameter files. These files are loaded into the system whenever the software is restarted. We adopted this methodology to make system maintenance independent of software

Fig. 3-80. Status display for the Nova target chamber.

development. Moreover, these parameters are typically adjusted by knowledgeable alignment-system technicians who are in close contact with the laser and know it best.

We recently performed exhaustive maintenance on the system and verified that its performance was at the design specification. Our system has been helpful in detecting several instances of broken hardware within the rest of the alignment system. Typical failure items are faulty limit switches, stalled motorized actuators, and incorrect cross-coupling matrices.

Author: W. G. Lablak

Improvements to Laser Diagnostics.

We have enhanced the software for our laser diagnostic energy processor (DEP) so that our diagnostics technicians can easily measure transmission and reflectivity of the Nova focus lenses. The enhanced DEP allows the use of a new type of amplifier, called a synchronous amplifier, and provides capabilities for obtaining sensor ratios.

We use a synchronous amplifier to measure pulsed laser energy. The new sensor ratio functions allow us to display the ratio of two measured energies from any two sensors of the same type on the same DEP. In addition, an average ratio function allows us to average and display every ten ratios. Our enhancements to the laser diagnostics system provide an accurate and fast way of measuring transmission and reflectivity of the Nova focus lenses.

Author: R. T. Shelton

Spectrometer Data-Acquisition System. The spectrometer data-acquisition system (DAS) consists of an IBM PC AT computer with a GPIB interface. An AD7912 storage scope and a boxcar integrator (4 A-to-D converters) are attached to the IBM PC AT through the GPIB interface. We use the AD7912 to digitize the signal from a microchannel plate that collects electrons emitted by verified gas contained in a vacuum chamber illuminated by laser light.

The DAS software is written in Basic and is structured for software maintainability. The program allows us to edit and maintain experimental parameters and to acquire, display, analyze, store, and retrieve data. The experimental parameters include the name of the experiment, data acquisition and time, number of samples, and number of data bins. The data-acquisition functions of the program can be used to acquire histograms or raw-waveform data and can average the waveforms into bins based on the energy of a given laser pulse. The display functions of the software give us the ability to display a histogram of shot-energy values or to plot a waveform contained in any energy bin. We can use the analysis portion of the software to perform background subtraction, waveform rectification, time-to-energy-domain conversion, and to integrate waveform peaks. In terms of data storage and retrieval functions, the software stores all experimental parameters and data so that retrieving any data restores the system to the state when the data were stored.

Our spectrometer DAS functions enable operators to easily acquire data, to analyze the data in various ways, and to display data on a monitor or printer at any time during analysis. The system is now in use and has already provided useful spectrometer data on various gases.

Author: R. T. Shelton

Target-Prealignment Image Processing.

Introduction. An important component of the Nova target prealignment system, the image-processing subsystem, became operational this year. Since its activation, the image-processing subsystem of our prealignment project has demonstrated the feasibility of the most critical concepts of the project and has proven to be an invaluable tool supporting our current efforts to refine the electromechanical, optical, and video subsystems associated with target alignment on Nova. Our refinements are expected to make target-prealignment procedures practical for most target geometries.

Target alignment has traditionally been a time-consuming procedure performed

on-line prior to irradiation of a target and has relied heavily on operator judgement. The purpose of our target-prealignment effort is to improve reliability, speed, and repeatability of target alignment in the Nova facility.

The prealignment system provides researchers, target designers, and alignment operators with the opportunity to manipulate, view, characterize, and align targets off-line at an optical bench, where Nova target-alignment mechanisms have been duplicated. The prealignment system uses a technology for transferring target-alignment parameters from the optical bench to the Nova chamber, which greatly improves the efficiency of on-line alignment. This technology is image processing. Image processing is intrinsic to the functionality of the prealignment system.

Prealignment Process. In the simplest scenario, a target is first aligned on the optical bench by conventional techniques. As shown in Fig. 3-81, the target is viewed

through three video cameras positioned at precise locations about the center point of the bench, which represents the center of the Nova target chamber. Each camera is outfitted with relay optics and an in-line reticle, which defines the line of alignment from that viewer. All vital aspects of the viewing system are calibrated with respect to a nearly identical viewing system on the Nova target chamber.

Once a target is aligned to the three line-of-site reticles, video signals from the cameras are digitized and stored on computer disk by the image-processing subsystem. Each digitized image typically appears as a backlighted target, obscuring portions of the full-field-of-view alignment reticle, depending on magnification. Images can be stored days or weeks before the target is used in an experiment at Nova.

The same computer disk accompanies the target to Nova. Here, a duplicate image-processing system reads the

Fig. 3-81. Sequence to realign a Nova target for a target shot following off-line prealignment.

images into memory and compares aligned images with live target images provided by the chamber-viewing system as the target is manipulated in the target chamber. Each of three color TV monitors displays the instantaneous difference between the live image of the target and the stored image for each of three viewing angles.

Before target silhouettes can actually be compared, however, the two image planes must be registered with each other for each view because alignment of a target in the stored image is relative to the reticle. Therefore, for each view, the image-processing system is used to shift the stored image on the monitor screen with respect to the live reticle image until the reticle images exactly coincide. Now the target can be positioned in the chamber so that its image matches the position of the stored image. Monitors display a vivid picture of the alignment differences between the sorted image and the target. The target is realigned with the stored image when the difference image suddenly becomes blank.

Once target images coincide for all three views simultaneously, the target is realigned.

Image-Processing Subsystem. Figure 3-82 shows the image-processing subsystem, which consists of two, nearly identical work stations. One station is attached to the optical bench in the prealignment laboratory; the other is integrated with the Nova target-alignment system. Each station uses an IBM PC AT, which includes a keyboard, color terminal display, two removable hard disks, each capable of storing 10 Mbytes of data, and two floppy-disk drives.

Each computer is interfaced to the video camera system with commercial hardware. The hardware, or video processor, includes onboard memory and is capable of digitizing video signals at a resolution of 512 pixels horizontally by 512 pixels vertically, with 8 bits of intensity information. Memory in the hardware can store $512 \times 512 \times 12$ bits of data. Therefore, one $512 \times 512 \times 8$ image can be stored, or two 512×512

Fig. 3-82. Two PC-based work stations perform image processing as part of the prealignment system.

images can be stored, each with only 6 bits of intensity data. The video processor is capable of manipulating portions of stored images and performing simple image-processing functions on the entire set of data, such as logical OR and subtraction, at TV frame rates of 30 frames/s.

The video processor provides output video signals in three colors, although the input digitization process is only sensitive to levels of intensity. Therefore, digitized black-and-white TV signals can be redisplayed as color-enhanced images containing combinations of red, green, and blue. The output of each video processor is displayed by a color monitor.

The image-processing station at Nova contains three sets of video-processing hardware so that three, simultaneous, color-enhanced image-difference displays can be provided on three color monitors. The station at the prealignment bench contains only one video processor and one color TV monitor, providing only one view at a time. This is, in fact, the only difference between our two image-processing stations.

Three video-input channels are available on each video processor, although only one input may be selected at a time for input to the digitization logic. The video processor in the prealignment bench station has a camera attached to each of the three inputs. The Nova station has three video processors, with one camera attached to each. Video-processing hardware can be controlled by software running in the PC AT, and digitized images can be transferred between video-processor memory and computer memory, and from there to disk under program control.

We wrote an extensive software package to provide an operator of the image-processing station with a hierarchy of menus on the terminal screen, from which may be chosen commands for performing over 40 functions. We developed only one software package so that the software running on each work station is identical. Menus are arranged in hierarchies so that an operator experiences a logical transition from general functions with higher-level menus to more specific control at lower-level menus. When

selected, many menu items display another menu containing more specific options pertaining to that general function.

Menu-selectable functions include manipulation of over 20 settings that control the behavior of one or more video processors; the use of controls that select the flow of data among video-processor memory, computer memory, and disk; and online help describing the system and function of each menu item. A menu of special note provides the operator with a choice of image to be output to the color monitor. Four images are currently available. For each view at the Nova station, the operator may choose to display the stored image, the live image, or the actual black-and-white difference between the live and stored images and a color-enhanced difference between the live and stored images, which shows each image in a separate color and the difference in a third color. Another menu of note allows the operator to quickly specify which of three video-input channels is selected for digitizing and displaying data. This menu is important at the bench station, where three views must be multiplexed through one video processor.

Simple schematic graphics on the terminal screen represent the current status of video processors, the currently selected flow of digitized data through the system, the amount that stored images are shifted during reticle alignment, and the type of image currently selected for display. Limited text can be appended to image data when stored on disk, and the text becomes available when the image is restored.

Our software also controls the way image data are stored and combined in the video processors. The difference between a stored image and a live image is computed entirely within the video processor. However, the PC AT program configures the video-processor memory so that the live image is stored in one-half of onboard image memory and is continually updated from the video camera input (see Fig. 3-83). The stored image is located in the other half of image memory. Both images contain only 6 bits of intensity information. The PC AT software then loads video look-up table memory in the video processor with bit

Fig. 3-83. Video signal subtraction is accomplished at TV frame rates by passing digitized images through a look-up table.

patterns and configures the processor to access data from both portions of image memory such that the difference of each pixel value is displayed on the color monitor.

Applications of the Image-Processing Subsystem. Our image-processing subsystem has been useful in preparing other components of the prealignment system for regular use. For example, images of Nova reticles were digitized and stored at four different magnifications for each view. These images were then taken to the prealignment bench, where the reticles, optics, and video system of the bench were aligned with those of Nova. A simple target was then used, first on Nova and then at the bench, to ensure that the systems were totally aligned with each other.

The video and optical systems of Nova are an older design than those of the optical bench. Backlight intensities and magnifications are not adjustable on the Nova system. We adjusted these variables at the prealignment station, again using the image-comparison features of the image-processing subsystem.

We are also giving serious consideration to upgrading Nova target-alignment systems as a result of such comparisons, and we have already made many refinements to the optical portions of the prealignment system to improve alignment precision and image quality.

The prealignment image-processing subsystem has proven to be extremely useful for vital tasks other than target alignment. We use the system to determine the accuracy and repeatability of inserting certain diagnostics into chamber center and to characterize motions of the target-insertion mechanism.

Conclusion. The dramatic visual feedback provided by the image-processing capabilities of our prealignment system has improved the efficiency of our target-alignment procedure. The system has demonstrated that, for certain target geometries, repeatable realignment can be performed within the limits of optical and digital resolution of the system, which are typically well within specified target-alignment tolerances. The image-processing subsystem has

been instrumental in assisting our efforts to refine components of the prealignment system.

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and G. D. Rambach

Analysis and Data Management

The LEAF/UNLEAF System.

Upgrades. During 1987 the Laser Experiments and Analysis Facility saw several modifications in hardware and software. The upgrades include a fiber-optic communications link, laser printer output, round-the-clock terminal access, improved versions of the operating system, and an upgrade to the ORACLE data base.

We installed a fiber-optic Ethernet cable to connect the LEAF computer to the Target Fabrication building. The network is connected to the terminal server in the Target Fabrication building and is driven by the Local Area Transport Protocol software on the LEAF computer. High-speed terminal connections are now available in the Target Fabrication building; in the future, additional terminals and computers could be connected to this high-speed network.

Another substantial hardware change was the addition of the Optimum Electronics Terminal Authenticator System. This addition to LEAF gives users a convenient means of terminal access 24 h per day. In the past, users had to physically connect their terminals through a connect/disconnect box inside the LEAF vault. This method was not only inconvenient but also required all frequent after-hour users to be on the vault-access list, thereby reducing security.

Users can now access the computer after hours by using one of the 12 terminals connected to the Authenticator. The security system requires a special password before electronically connecting the terminal to the computer. Once the password has been entered and verified, the user receives a continue

prompt and may log on as normal. The system has proved to be successful, and we have plans to add 12 additional ports.

Two additional hardware upgrades took place this year. The first is a unibus to separate the Ramtek from other peripherals, reducing bus contention and improving throughput. The second is a laser printer for quality text and graphics output.

The VAX operating system went through two software upgrades, from version 4.3 to 4.5. Version 4.4 was a major upgrade with new features, such as the capability of callable DCL subroutines. Shortly after we installed version 4.4, we added version 4.5, which primarily fixed bugs found in version 4.4.

Another major software upgrade was that of the ORACLE data base management system. The latest version incorporates substantial improvements and changes throughout the entire data base facility.

We installed a new software package by RAXCO, called RABBIT5, on the VAX to allow for improved backup performance. The RABBIT5 backup is 2 to 2.7 times as fast as the VMS backup facility, and the CPU time required to execute the VMS backup utility is 8.6 times greater than that of RABBIT5. The impact on users is that they run the RABBIT5 backup program much less than the VMS backup program.

Additional software upgrades include:

- The addition of DOPTEP, a disk optimizer package produced by RAXCO for disk defragmentation. (Disk accesses are faster when files are not fragmented.)
- The addition of the TEX typesetting package, which is capable of producing either simple documents or those with complicated mathematical and scientific figures.
- An upgrade of the IMSL Math and Statistics Library to include all previous capabilities and many new ones.
- An upgrade of the Computer Associates TELLAGRAF graphics package to version 6.1.
- An upgrade of the Computer Associates DISSPLA graphics package to

version 10.5. This version includes an LN03 driver to enable DISSPLA graphics output on the laser printer.

During 1987, we upgraded the Unclassified Laser Experiments and Analysis Facility hardware to improve its input and output capabilities. We enhanced the UNLEAF disk structure by adding a System Industries 9722 Winchester disk, which is capable of storing 546 Mbytes of data on 8-in. platters.

Another hardware addition to UNLEAF was a System Industries 9621 dual-density tape system. This tape drive replaced the older and less reliable DEC TU-45 tape system. The new drive has 1600 or 6250 bpi for storing 180 Mbytes of data on a single 2400-ft reel of tape. In the past, we could store only 70 Mbytes on a tape.

Author: V. A. Martin

Film and Data Processing. Nova target-shot experiments produce two types of data that are stored in the LEAF

computer system. The first type is electronic data, which is recorded in the on-line control computers that operate Nova. These data are transmitted to LEAF via a one-way fiber-optic link. The second type is recorded on film and is digitized for computer storage by a PDS microdensitometer. All data are stored in a data base controlled by software from Oracle Corporation.

The major diagnostics that produce film that is digitized are as follows:

- McPIGS (40%).
- Streak cameras (10%).
- Optical/x-ray streak camera (8%).
- Streaked optical spectrograph (5%).
- 8 × microscope (4%).
- 22 × microscope (2%).
- Henway x-ray spectrograph (2%).
- Others (29%).

Some data, such as McPIGS, are transferred to the UNLEAF via magnetic tape for further analysis. The transfer allows visitors and employees who do not have access to the LEAF computer to process and see their data.

Fig. 3-84. Total film images digitized by the LEAF PDS system during 1987.

Each week we archive both the film and shot data files from the shot data disk to magnetic tape. A program has been written to keep track of which data are on which archive tape so that users can easily retrieve old data. As shown in Fig. 3-84, we digitized a total of 4326 film images for Nova, Phoenix, Phebus, and special projects during 1987.

Author: B. L. Stewart

LEAF Analysis and Data Base Software.

Introduction. In this article, we describe developments related to interactive software that support the Nova staff in defining and reviewing Nova experiments and Nova management in defining and tracking material charges, personnel information, and expenses. All data handled by this software are included in or referenced by the ORACLE relational data base tables.¹²⁹

The PRESOT module, which we use to define a laser target experiment (or shot), is now operational on both the LEAF and Nova Development System VAX computers. We are currently implementing the laser science (LASSCI) module, which we use to define laser bench experiments on Nova Development System VAX computers. We defined a target fabrication data base that is now in use and is linked to the LEAF VAX. We also developed software to place each post-shot experiment definition into the electronic SHOTBOOK data base on the LEAF VAX. With these software-interface systems in place, we will have a consistent level of communication among all aspects of the laser experiments program.

The definition of each target experiment is archived in our PRESOT data base by a shot physicist using the PRESOT module software. These data form the basis for setting up the Nova laser for a given shot. The definition of each target type and its design, availability, and status are archived in our target fabrication (TFAB) data base on the LEAF VAX. These data are maintained by Nova target fabrication staff.

The results of processed shot data are archived in the electronic SHOTBOOK data base; these results are available to the physics staff through the electronic

SHOTBOOK software located on the LEAF VAX. We regularly develop and upgrade specialized data-processing codes to further support the needs of our physics staff.

We define and maintain material requisitions in the FINANCE data base of the LEAF VAX. These data are used to provide the Purchasing Department with a hard copy requisition form and to track the status of orders. We also maintain complete general-ledger accounting data and time charges for personnel on a monthly basis in the FINANCE data base. This data base also includes general information on personnel.

As described earlier in this section, the LEAF consists of a VAX-11/785 computer; the Nova Development System consists of a VAX-11/780 computer. Both the electronic SHOTBOOK and PRESOT modules use the Forms Management System menus.¹³⁰ All software is designed around the ORACLE relational base management system from which data are received. We have installed the latest version of the commercial graphics software package DISSPLA¹³¹ on the LEAF VAX and now use this package to provide the necessary graphics at a terminal or any hard copies that are desired.

LEAF Analysis Software. We have written or upgraded the following specialized data-processing codes to keep pace with evolving diagnostics on Nova:

- For Dante diagnostic instruments, we implemented a common data base and software system on the UNLEAF and LEAF VAX computers to handle filter and foil inventories and calibrations. Information about material received in the UNLEAF area can now be assessed and analyzed by the diagnostician in the LEAF area.

- For the new photoconductive detector (PCD), we designed and implemented a data base with supporting software to provide analysis of response functions.

- For NTOF instrumentation, we extensively revised existing software to provide preprocessing of selected oscilloscope data. The results are a reduction in time for post-processing analysis from about 5 min to about 10 s.

- We implemented the first form of the code TOTAL SPECTRA, which displays as a continuous spectrum the separate

Fig. 3-85. Typical results for separate Dante, Henway, and FFLEX spectra.

spectra obtained from up to four, separate diagnostics [TGSS, Dante, Henway, and the filter-fluorescer x-ray spectrometer (FFLEX)]. Figure 3-85 shows typical results for separate Dante, Henway, and FFLEX spectra. The end product will incorporate these data for comparison to and analysis of the PCD response.

- We completed the data base design for the large neutron-scintillator array (LaNSA) diagnostic, which will be implemented in 1988.

Electronic SHOTBOOK. The electronic SHOTBOOK continues to be a fully operational tool used to review and analyze the results of Nova laser experimentals. We incorporated new software to keep pace with six new types of diagnostics that were added to the Nova laser in 1987. The software provides the related displays, menus, and ORACLE update forms required to access and annotate processed data from the new instruments. We further modified the software to announce "No data available" in the table of contents to obviate the need for proceeding to an empty display; we also added a binary-read routine for ~60% faster reading of PDS data. Additional information about the electronic SHOTBOOK is contained in earlier Laser Annual reports.

We extended access to the electronic SHOTBOOK, which is used by about 40 staff physicists and data-processing personnel, by means of a two-way data link between the LEAF VAX and the remote Target Fabrication Facility. We will install a similar two-way link between the LEAF VAX and the Nova Operations control room so that full access to the LEAF software and data base is available to our staff at all locations.

PRESHOT Definition. The purpose of the PRESHOT module is to provide a convenient method to define, archive, display, and obtain a hard copy of requested Nova experiments. Using this program:

- The shot physicist can define all aspects of a target shot request well ahead of the actual shot date. At definition time, the PRESHOT number and the related target identification are uniquely and automatically generated.

- All staff members can obtain hard copies of any experiment request.

- All staff members can readily review, either in general or in detail, the experiment plans of the physics staff.

- The related data base tables of the electronic SHOTBOOK can be automatically updated when a requested experiment, as defined by a PRESHOT, is completed.

PRESHOT module software was originally placed on the LEAF VAX. The module has now been placed on the Nova Development VAX for use by the operations staff in modifying an original PRESHOT request to represent the actual shot conditions and to specify the related shot number when the experiment is completed. After a shot, these data are transferred to the LEAF VAX for final archiving, and the Nova Development VAX data base records are "locked" to prevent change. Further information about the PRESHOT module is contained in the 1986 Laser Annual.

We have completed the design of a LASSCI module, which is similar in structure to the PRESHOT module, but specific to the various laser bench experiments conducted on beamlines outside of a target chamber. Implementation of this software is now in progress for use by the Nova operations staff at the Nova Development System VAX.

As with the PRESHOT module, the writing of a detailed pre-LASSCI shot request is simplified by adding the option for a user to begin with the definition from an existing pre-LASSCI experiment request. The request includes parameter definition, laser diagnostic requirements, and other special instructions for setup. Copies of the parameter and diagnostic section are done by ORACLE insert

instructions to the data base, and the new instruction file is simply copied from the old file.

Writing a new instruction file or modifying from a copy is done within the code using the VAX on-line editor, EDT. The instructions file can be either a single page or, as is more often the case, a structured and lengthy document that can be quite similar in form from one shot to the next.

Target Fabrication. With the data link from the Target Fabrication Facility to the LEAF VAX in place, we designed and implemented a comprehensive target fabrication (TFAB) data base, with related forms, for archiving both target design and status. This data base also serves to inform shot physicists of target availability. To complete the package, we added a report writer to provide lists of targets sorted by builder, kind, build date, and status. We maintain data base consistency by controlling uniqueness of the target identification specification number and of the target kind. Data base integrity is controlled through the ORACLE software. Further effort is pending to archive as-manufactured target data in the TFAB data base so that detailed information about the kinds of laser targets used to date will also be available via the electronic SHOTBOOK.

Financial Data System. We added software to the requisition-control section on the LEAF VAX to provide direct printing of multipage, fanfold requisitions using the data base information. In addition, we expanded the data base to handle split accounts, where several account/subaccount pairs are charged to one requisition. As for a single-account requisition, costing is apportioned to each account/subaccount for each item, and each of the split accounts are also maintained in the data base for tracking purposes by individual project managers. Either the requester or the recipient of each order can be identified. We restructured the data base to maintain consistency with the Paris system, which will soon be available in the LEAF area.

Conclusions. We have demonstrated the quality and integrity of our software designs, coupled to a relational data

base, for defining and archiving Nova experimental data and for providing ready access to that data for later review. Thus, our design methods are providing a useful basis for continuing software developments. In 1988 we expect to apply the same philosophy for software design to the solution of other communication needs in assisting LEAD, Nova operations, and target-fabrication activities.

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Multidimensional Signal Processing.

Wave-Front Sensing. Signal and image processing is a major area of emphasis for Information Systems. This technology is critical to many of the data-analysis codes we use to process experimental data. In addition, the automatic alignment system in the Nova laser facility is based on image processing of video-sensor images for extraction of the position of the laser beam and the location of alignment references. These systems are described elsewhere in this and past Laser Annuals.

Wave-front sensing of laser beams is an emerging application of multidimensional signal processing. This measurement is becoming increasingly important as we strive to improve the performance of our laser components and to better understand the effects that the quality of laser light has on our target experiments. Interferometry has long been used to qualify our optics; we have now extended the techniques so that we can measure wave fronts of pulsed laser beams.

We developed an interactive analysis system for the signal processing of images digitized from interferometric wave-front sensors. The technique is based on angle modulation theory in which a large amount of tilt is introduced into one arm of the interferometer. The tilt component then serves as a spatial carrier to the modulating wave-front phase. Fourier-domain bandpass processing of the interference fringes effectively removes the adverse effects of irradiance variations and camera background noise.

An aberration consisting of pure tilt appears in the interferogram as many straight, sinusoidal intensity fringes. The addition of tilt acts to straighten out the patterns normally observed in an interferogram and provides high resolution of small aberrations.

Fringe intensity is given by an equation of the form:

$$I(x,y) = A(x,y) + B(x,y) \cos[2\pi^*f_c^*x + \varphi(x,y)], \quad (41)$$

where $A(x,y)$ and $B(x,y)$ are contributions from unwanted irradiance variations and the video camera background signal. The linear phase term $2\pi^*f_c^*x$ is the wave-front tilt, which is removed in the processing. The desired wave-front surface is then given by the remaining phase term $\varphi(x,y)$.

Figure 3-86 shows the Fourier components of the interferogram of a deformable mirror produced by the Zygo Mark III AC heterodyne interferometer. Interferograms prepared on this instrument were used to validate the analysis software. The large spectral mode at the lowest spatial frequencies is the background

signal resulting from the sum of the two interfering beams. The tilt introduced into the interferometer acts to separate this component in frequency space from that of the desired phase signal. A digital bandpass filter is then employed to pass the spatial frequencies around the carrier frequency while rejecting the others.

The analysis is performed in the following manner. The 2-D Fourier transform of the interferogram is computed and complex multiplied with the transfer function of the bandpass filter. This so-called fast convolution is much quicker than the comparable spatial-domain operation and can be carried out in a few seconds on an array processor. Next, one-half of the spectrum is selected, converting the original bandpass system to a low-pass system, which is easier to process. An inverse Fourier transform yields a complex image since the low-pass form of the frequency-domain signal no longer has Hermitian symmetry.

The wave front is retrieved by computing the phase of the complex image [Fig. 3-87(a)]. The phase values are indeterminate to a factor of 2π because an arctangent function is used. Discontinuities occur at $+\pi$ or $-\pi$ when the arctangent

Fig. 3-86. (a) Interferogram of a modal deformable mirror. (b) Log magnitude of the Fourier transform of (a).

Fig. 3-87. (a) De-modulated phase surface of the interferogram in Fig. 3-86 containing 2π discontinuities. (b) Phase-unwrapped and de-tilted wave front from (a).

Fig. 3-88. Processed wave front measured on a high-average-power slab amplifier at increasing pump power.

function "wraps" to the opposite value. These phase values are then unwrapped to produce a smooth function of phase.

The linear phase term is removed by subtracting the best least-squares-fitted plane from the phase surface, shown in Fig. 3-87(b). With this method, the wave-front scasor is blind to low-order tilt (pointing) phase errors. If we wish to retain this capability, then the spatial carrier term must be measured independently and subtracted from the unwrapped phase image.

Our High Average Power Laser Program is involved in designing advanced, solid-state laser-amplifier components that can be used in future ICF applications. Our analysis system is being used to investigate the effects of laser amplifier heating on wave-front distortion. The measurements are made by placing the amplifier within the sampling cavity of a Fizeau interferometer.

Figure 3-88 shows isometric plots of the laser wave fronts taken from one of these amplifiers at five, linearly increasing flashlamp-pump-power levels. In the quiescent state, there is less than 1/20 wave rms of phase distortion. As pump power increases, a fairly linear increase in rms wave-front distortion is evident.

The software and signal processing for this project has been implemented using one of the Nova VAX-11/780 computers with an attached array processor. Interferograms are processed at a 128×128 resolution, a spatial sampling that allows processing of aberrations up to approximately 2 waves in magnitude.

Our system is in frequent use for analyzing wave fronts in several laboratories in the Laser Program. The analysis

package has made possible for the first time a practical way to measure the aberrations of pulsed optical systems.

A portable unit with an improved operator interface would be beneficial for use in the laboratory environment. The need also exists for increasing the spatial resolution and maximum aberrations we can process. In response to such needs, we are now developing a second-generation system based on a microVAX GPX workstation. The image spatial resolution and array processing specifications are being increased to 512×512 pixels to allow for the required wider bandwidths. This work is being cofunded as a development project for the Remote Image and Signal Engineering Thrust Area of the Engineering Research Division of the EE Department.

X-Ray Holography Modeling. As we reported in the 1986 Laser Annual, we have been successful in building special targets, which, when irradiated with specially focused Nova laser beams, re-emit coherent light in the soft x-ray regime. We have begun to investigate the possible uses of coherent x rays for biological imaging applications. Gabor holography is the principal configuration under study.

We formed a working group to bring together the disciplines of physics, biology, and multidimensional signal processing to quantify the types of problems that could be addressed. The function of the Information Systems Group is to provide computational support for the numerical simulation and analysis of x-ray holograms of biological objects. These objects are primarily characterized as wet protein structures that are

semitransparent to soft x rays. The team members are also implementing the reconstruction software that deconvolves the hologram recording into a 3-D image of the original object.

To date, the Group has written Fresnel diffraction codes, which can simulate Gabor holography. Holograms of clusters of simple proteins have been synthesized. At present, reconstruction has been implemented with linear deconvolution, and we are now progressing to a more robust, iterative approach to phase retrieval that separates the multiple-image planes that are encoded in the hologram. The algorithms have been integrated into the VIEW multidimensional signal-processing workstation project.

We are designing experiments using optical lasers and scaled biological specimens to test and further develop the algorithms. Our experiments are expected to provide inexpensive estimates of the 3-D image quality obtainable by x-ray holography of semitransparent biological objects.

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Laser Experiments and Advanced Diagnostics

Introduction

We have completed the second full year of operation of Nova as a target-experiment facility. Our principal activities in target experiments and diagnostics were related to understanding the physics of indirect-drive fusion using Nova.

During 1987 we greatly enhanced our diagnostic capabilities, allowing for more sophisticated experiments with multiple diagnostics. We continued to develop capabilities for time-resolved x-ray imaging on Nova and activated the 22 × Wölter x-ray microscope that is coupled to an

x-ray streak camera. We also enhanced our framing capabilities by developing a four-frame, x-ray pinhole camera. We continued our advances in implosion and drive diagnostic capabilities. Our experimental efforts were directed toward understanding several physics issues rather than attaining specific milestones. Some of our major accomplishments are summarized in this article.

One of our major areas of research continues to be the understanding of high-Z laser-target coupling and x-ray conversion. We conducted an important set of experiments to better understand in detail the processes of laser absorption, energy transport, and x-ray conversion in the laser interaction with high-Z matter (see the articles under the next heading, "Experiments on Multikilojoule Laser-Plasma Interactions"). The emphasis of these experiments was to isolate the physical processes involved to better understand their role in laser interaction and x-ray conversion processes. We used sophisticated targets, such as low concentrations of gold diluted in beryllium and small dots of gold imbedded in plastic as well as other high-Z materials. The results have improved our modeling and increased our understanding of high-Z plasmas.

We also performed extensive experiments to investigate the hydrodynamics of radiatively accelerated materials. Some of the results are presented in articles under the heading, "Implosion and Hydrodynamics Experiments," later in this section. We demonstrated the ability to study the stability of an interface between two accelerating materials using point-projection spectroscopy; here, we can quantitatively measure the amount and extent of the mixed region. We also demonstrated that we can measure no mixing in stable interfaces, thus giving confidence to our experimental technique. We measured the growth of Rayleigh-Taylor instabilities in accelerated materials having initial perturbations. These experiments were the first application of the 22 × Wölter x-ray microscope, and they established its operation on the Nova target chamber.

For the first time, we have directly imaged neutrons from a burning core (see the article, "Neutron Penumbra Imaging"). The images were obtained by a coded aperture technique. We have developed two systems for measuring the time for neutron production relative to the incident drive. One system uses a microchannel-plate detector for neutrons (see "Neutron Emission-Time Measurements"), while the other uses photoconductor detectors (see "Measurement of Single, 14-MeV Neutron Pulses with 100-ps Resolution"). The photoconductor detectors have better time resolution and can resolve the actual burn widths to less than 130 ps, but they are less sensitive than the microchannel plate. In addition, developmental work has begun on the large neutron-scintillator array (LaNSA) (see "Large Neutron-Scintillator Arrays," later in this section). When completed, this system will allow us to measure fuel ρr up to 1 g/cm^2 using the tertiary neutron technique.

We also made substantial advancements in fielding new diagnostics on Nova and in better understanding the performance of existing diagnostics. Major advances have been made in our streak and framing camera capabilities (see the articles under the heading "Streak and Framing Camera Technology"). We have developed four-frame gating systems, which are now routinely operating on Nova to obtain x-ray images and x-ray spectra with 100-ps resolution. Several research efforts continued to better quantify the output of optical and x-ray streak cameras.

In addition to the ICF efforts, steady progress continued in the laboratory x-ray laser experiments. We observed gain in nickel-like transitions for the first time (see "Progress in Nickel-Like Lasers"). Progress also continued on producing a lasing cavity. For the first time, we used a mirror and beam splitter to demonstrate a triple-pass system in an x-ray laser medium. This work demonstrates advances in our microstructure capabilities (see "X-Ray Optics and Technology"). Several experiments demonstrated the potential applications of an

x-ray laser, and we produced the first x-ray hologram using an x-ray laser (see "Demonstration of X-Ray Holography with an X-Ray Laser"). The output at the x-ray laser was used to nonresonantly excite another medium that can potentially produce lasing.

Under the heading "Facilities," we discuss several improvements that have increased our operational effectiveness and safety. In the article, "CADD System for Nova Targets," we discuss how this device will allow new instruments to be designed without conflicts among beam paths, existing instruments, or observation lines of sight. Another safety feature on the ten-beam chamber is discussed in the article, "Chamber Collision-Avoidance System." This system controls the insertion of instruments near the center of the target chamber to avoid damage by colliding with other incompatible instruments. The final article concerned with laser facilities is "Upgrade of the Short-Pulse Laser."

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Target Experiments

Experiments on Multikilojoule Laser-Plasma Interactions. For laser fusion to succeed, interaction processes in the laser-produced plasma must absorb laser light efficiently and produce few suprathermal electrons. As the energy and duration of laser irradiations increase, the plasmas they produce develop longer scalelengths and become more planar.

During the 1980s, multikilojoule lasers have allowed the study of laser-plasma interaction processes in plasmas that could not have been produced with smaller lasers. Here, we summarize some experiments on laser-plasma interactions performed at LLNL in recent years, with emphasis on results that directly affect the prospects for laser fusion.

Consider what conditions can be produced by a high-power laser that is able to irradiate a target with more than 1 kJ of energy. The irradiation can satisfy three conditions that cannot be simultaneously

met using smaller lasers. First, the intensity I_L can exceed 10^{15} W/cm². This value is near the upper limit envisioned for laser fusion. Second, the pulse duration τ can equal or exceed 1 ns. This duration is long enough to allow a large corona to develop in front of a target (although it does not equal the 5- to 10-ns pulse durations that will be needed for high-gain laser fusion). Third, the spot diameter can exceed 300 μ m, with effects described below.

Given such irradiation conditions, the plasma produced will have the following properties. First, it will have an electron temperature greater than 1 keV. Such temperatures are comparable to those anticipated in high-gain targets. Second, the plasma will remain relatively planar throughout a 1-ns laser pulse because the spot size is larger than $c_s \tau$, where c_s is the ion sound speed in the plasma. High-gain targets are expected to produce relatively planar plasmas. Third, the plasma will reach density-gradient scalelengths L of several hundred laser wavelengths λ or more, where $L = [(1/n) dn/dx]^{-1}$. These scalelengths are smaller than those expected in high-gain targets, where L/λ will reach several thousand. However, they can be used for significant experiments.

By producing the conditions just discussed, multikilojoule lasers can perform two useful types of experiments. First, they can identify mechanisms of laser-plasma interaction that appear in planar plasmas at intensities and temperatures of interest to laser fusion. The most obvious example is sidescattering processes, which are not strongly favored in spherical plasmas produced at relevant intensities by smaller lasers. Second, multikilojoule lasers can be used to study the dependence of any interaction mechanism on scalelength L . Through such experiments, we hope to learn enough to extrapolate with confidence to the scalelengths anticipated in high-gain targets. We must reach values of L/λ that significantly exceed 100 to exceed the thresholds or to achieve significant gain for several interaction mechanisms of interest.¹³² In addition, by performing such

experiments using several laser wavelengths, we can study wavelength scaling of important mechanisms.

In the following articles, we discuss measurements of light scattered out of the target plasma in two ranges of frequency. After describing the experiments themselves, we discuss measurements of light scattered with frequencies near the fundamental frequency of the laser light (ω_0). Such light may be reflected from critical density n_c or may be scattered directly by stimulated Brillouin scattering (SBS), in which the incident light wave decays into a scattered light wave and an ion-acoustic wave.¹³³⁻¹³⁵ It may be indirectly produced as a result of resonance absorption,^{136,137} in which resonantly driven plasma waves near the critical density absorb (and may reemit) light, or by parametric decay,¹³⁸ in which the incident light wave decays into an ion-acoustic wave and an electron-plasma wave.

Following our discussion of scattering at frequencies near ω_0 , we discuss scattering at frequencies below ω_0 , but above $\omega_0/2$. Light is scattered into such frequencies by stimulated Raman scattering (SRS), in which the incident light wave decays into a scattered light wave and an electron-plasma wave.¹³⁴ We ignore light scattered at $\omega_0/2$ by two-plasmon decay^{139,140} or by Raman scattering at $0.25n_c$ because the evidence indicates that in large plasmas, scattering by SRS at densities below $0.25n_c$ overwhelms such scattering both as a source of hot electrons and of scattered light, and because there is no reason to expect the trend to change in larger plasmas of high-gain targets. In our conclusions, we summarize the results and consider their implications for laser fusion.

Review of Experiments. The laser facilities for our experiments are summarized in Table 3-15. We used the Argus¹⁴¹ and Shiva^{142,143} lasers to produce results with 1.06- μ m laser light. We used the Novette facility to irradiate targets with 0.53- μ m light^{144,145} and 0.26- μ m light.¹⁴⁶ We used the Nova facility for our results on 0.35- μ m light. As the table shows, the energy on target was typically 2 kJ, but varied from less than 1 kJ to about 4 kJ.

When possible, our experiments involved only one laser beam so that the results could be interpreted most clearly. However, experiments using Shiva required ten beams.

All the lasers used neodymium-doped glass to amplify a low-energy pulse of light, and they all operated with narrow bandwidth. Wavelengths shorter than $1\text{ }\mu\text{m}$ were produced by harmonic conversion of $1\text{-}\mu\text{m}$ light. In every case, the intensity at the target plane was strongly modulated, with peak-to-valley fluctuations on the order of 5 to 1.

Laser pulses were usually Gaussian in shape and varied from 700 ps to 2 ns in full width at half maximum. Nova pulses for our results were square, 1-ns pulses. A few experiments found no strong dependence on pulse shape or duration over this range; thus, we ignore differences in pulse shape during our discussions.

Data were obtained by irradiating two types of targets. Some targets were thick enough that they did not burn through during the laser pulse. We refer to these as disk targets. Most of the disk targets were gold, although we used some low- Z disk targets. The experiments using disk targets produced local-gradient scale-lengths L of several hundred laser wavelengths. We refer to the second type of target as a foil target. Such targets are thin enough to burn through during the laser pulse. The expanding plasma that results has a density maximum and may have a significant volume over which L exceeds 1000 laser wavelengths. We obtained extensive data from CH foils and some data from gold foils.

Results from foil or disk targets irradiated with about 2 kJ of energy are compared to results from disk targets irradiated with about 100 J to identify the effects of scalelength and planarity on laser-plasma interactions. Our results were obtained with laser intensities on the order of $2 \times 10^{15}\text{ W/cm}^2$. We performed some experiments to study the scaling of laser-plasma interaction processes with laser intensity, although much of this work is beyond the scope of this discussion.

Table 3-15. Laser facilities used for laser-plasma interactions.

Laser	Number of beams	Energy (kJ)	Wavelength (μm)
Argus	1	0.5 to 1	1.06
Shiva	10	3 to 4	1.06
Novette	1	3 to 4	0.53
Nova	1	1.5 to 2	0.35
Novette	1	1 to 1.5	0.26

The many diagnostics used to measure the scattered light emitted by plasmas and to measure the hard x rays produced by suprathermal electrons are described in detail in the reports cited above. In general, diagnostics included absolutely calibrated photodiodes to measure the fluence and angular distribution of scattered light, spectrometers coupled to streak cameras to measure the time-resolved spectra of scattered light, and absolutely calibrated x-ray spectrometers to measure hard x rays.

Scattering Near ω_0 . We now discuss measurements of scattered light with frequencies near the frequency of the light incident on target, ω_0 . The results of long-scalelength and wavelength-scaling experiments might have been either favorable or unfavorable for laser fusion. On the one hand, longer-scalelength plasmas absorb a larger fraction of laser light before it reaches the critical surface. Such absorption reduces the reflection of laser light from the critical surface and hot-electron production by resonance absorption. In addition, other interaction processes may be collisionally damped (or simply overwhelmed by inverse-bremsstrahlung absorption) at shorter laser wavelengths. On the other hand, long-scalelength, planar plasmas should have significant gain for SBS backscatter and sidescattering so that the absorption of laser light could become much less efficient. Of particular concern is any convective process that scales as $\exp(\beta)$, where $\beta \propto I_L L$. Any such process that is detectable in multikilojoule experiments has the potential to be devastating at the megajoule level.

As shown in Fig. 3-89, long-scalelength, planar plasmas have produced SBS side-scattering. Here, the fluence of scattered light in J/sr is plotted as a function of azimuthal angle about the axis of

Fig. 3-89. Fluence of scattered ω_0 light vs azimuthal angle with respect to direction of propagation of laser beam for two polar angles. Observed scattering is much stronger in the out-of-plane direction.

Table 3-16. Percent scattered light near the fundamental frequency of incident laser light.

Incident wavelength (μm)	100-J lasers (gold disks)	2-kJ lasers (gold disks)
1.06	60	60
0.53	30	25
0.35	Not measured	15

propagation of the laser beam. Data are shown for polar angles of 120° and 135° with respect to the direction of propagation. In this experiment, we used 2 kJ of $0.53\text{-}\mu\text{m}$ light to irradiate a $660\text{-}\mu\text{m}$ spot on a gold disk for 1 ns to produce an average intensity of $6 \times 10^{14} \text{ W/cm}^2$. The laser beam was incident along the target normal. Scattering perpendicular to the electric-field vector of the incident light (out of plane) is many times stronger than the in-plane scattering. This dependence is expected for SRS sidescattering and is not expected for any other mechanism at these frequencies.

We also observed SRS sidescattering from plasmas produced with 1-, 0.53-, and $0.35\text{-}\mu\text{m}$ light (but made no observations with $0.26\text{-}\mu\text{m}$ light). Experiments at similar laser intensities and pulse lengths, but using smaller spots, have not seen this process. Thus, it is a scattering mechanism that appears in relatively long-scalelength, planar plasmas. The magnitude of SRS sidescattering, integrated over polar angle from 90° to 150° , has been as large as 10% of the laser energy. Thus, SRS sidescattering can be a

significant energy-loss mechanism, and its scaling to high-gain conditions is of concern to laser fusion. Fortunately, SRS sidescattering has been observed to saturate quite strongly as a function of laser intensity. As a result, the best projection is that it will not substantially exceed 10% of the laser energy even under high-gain conditions.

Backscattered light with frequencies near ω_0 may result from SRS backscatter, reflection from critical, or other more complex processes. We have not carried out sufficiently detailed spectroscopic studies to determine which mechanisms dominate under what conditions. We have measured the total fraction of incident laser energy that is backscattered, and it is smaller from 2-kJ experiments than it is from 100-J experiments.

The net effect of smaller backscatter and the advent of sidescattering is that the total amount of scattering at frequencies near ω_0 changes little as scalelength increases, as is illustrated in Table 3-16. (Results for the 100-J laser were obtained using Argus.¹⁴⁷) At the two wavelengths for which data are available, the difference between experiments with different scalelengths is less than the relevant uncertainties. We view with cautious optimism this scaling of total scattered light as scalelength increases. It leads us to hope that the total scattering will not increase significantly in high-gain targets with larger plasmas.

The scaling of scattered light with laser wavelength is much more favorable. The total scattered light decreases as wavelength decreases, as Table 3-16 also shows. In addition, both the backscattered energy and SRS sidescattering are observed to decrease. Inverse-bremsstrahlung absorption definitely is larger in the denser, cooler plasmas produced by the shorter-wavelength lasers, and more absorption takes place at lower densities. Furthermore, any light that is scattered is more strongly absorbed as it leaves the plasma. Moreover, other collisional affects might also act to decrease the scattering. Thus, these wavelength-scaling data are both sensible and encouraging. If the mechanisms were more clearly understood, we would expect to conclude

that scattered light at frequencies near ω_0 would not prevent high-gain laser fusion, particularly for laser wavelengths of $0.53 \mu\text{m}$ or less.

We excluded data for foil targets from Table 3-16 for two reasons. First, the foil targets produce long density-gradient scalelengths but relatively short velocity-gradient scalelengths. Because SRS requires long scalelengths of both density and velocity gradients, such targets should produce little SRS. Second, as expected, we observe very little scattering at frequencies near ω_0 after such targets burn through. The velocity-gradient scalelengths of such targets at very low densities ($n \leq 0.05n_c$) are, however, quite large after burnthrough. Thus, our result shows that the dominant region of SRS from such plasmas is at densities above $0.05n_c$, a result that is also encouraging.

Stimulated Raman Scattering. The possibilities for SRS are similar to those for SRS. First, increasingly planar, long-scalelength plasmas may allow more growth of SRS backscatter and sidescattering. Second, collisional damping may quench SRS. Finally, it is important to examine the saturation behavior of SRS.

We have observed SRS sidescattering from targets irradiated at all the laser wavelengths included in Table 3-15. High- and low-Z disks and high- and low-Z foil targets have all produced such scattering. The expected strong difference between out-of-plane scattering and in-plane scattering has been observed for 0.53 - and $0.35\text{-}\mu\text{m}$ irradiations. (Sufficiently detailed measurements were not carried out for 1.06 - and $0.26\text{-}\mu\text{m}$ light).

Figure 3-90 shows some data from irradiation of a gold disk target with $0.53\text{-}\mu\text{m}$ light and the same experimental geometry as that described for Fig. 3-89. The figure shows the strongly out-of-plane scattering observed at 120° and 135° from the direction of propagation of the laser beam. Based on the measured spectrum, this light was probably scattered at angles near 90° within the plasma, after which refraction steered it to the observed angle.

We have observed increasing SRS backscatter from longer-scalelength plasmas.

Figure 3-91 shows the increase of the total amount of SRS for gold targets irradiated with $0.53\text{-}\mu\text{m}$ light at an intensity of about $2 \times 10^{15} \text{ W/cm}^2$. This figure shows the fraction of laser energy scattered as SRS light vs normalized gradient scalelength L/λ . Experiments on Argus with small scalelengths and relatively

Fig. 3-90. Fluence of SRS light vs azimuthal angle with respect to direction of propagation of laser beam for two polar angles. Observed scattering is much stronger in the out-of-plane direction.

Fig. 3-91. Fraction of laser energy scattered as SRS light vs scalelength of electron-density gradient. SRS increases strongly with scalelength.

Fig. 3-92. Fraction of laser energy scattered as SRS light vs collisional-damping parameter. SRS is quenched as the damping parameter approaches 1.

Table 3-17. Percent of Raman-scattered light.

Incident wavelength (μm)	100-J lasers (gold disks)	2-kJ lasers (gold disks)	2-kJ lasers (CH foils)
1.06	0.02	1	10
0.53	0.01	1	10
0.35	0.01	1	3
0.26	Not measured	0.07	2

spherical plasmas produced SRS fractions of about 10^{-4} . The relatively planar and larger-scalelength plasmas of the Novette experiments produced SRS fractions on the order of 10^{-2} for disk targets and 10^{-1} for foil targets. It is unclear from existing data at what level SRS saturates as scalelength increases. Given that high-gain targets will produce plasmas with scalelengths significantly larger than those shown in the figure, this question is of some concern for laser fusion.

Fortunately, it is possible to control SRS by means of collisional damping. Figure 3-92 shows the SRS fraction as a function of the collisional-damping parameter ν_{ei}/γ_0 , where ν_{ei} is the electron collision frequency, and γ_0 is the homogeneous growth rate for SRS. The data are from foil targets irradiated at an intensity of about 10^{15} W/cm². This figure includes data from CH and gold foils irradiated at various wavelengths. Experiments near or below the collisional-damping threshold, $\nu_{ei}/\gamma_0 \approx 1$, produced negligible amounts of SRS.¹⁴⁸ We also

observed a strong dependence of SRS fraction on intensity in experiments near the collisional-damping threshold.

Table 3-17 shows the wavelength- and scalelength-dependence of SRS. (Results for the 100-J laser are from high-Z targets irradiated with 1.06-μm light¹⁴³ or 0.53-μm light¹⁴⁹ using Argus, and from low-Z targets irradiated with 0.35-μm light¹⁵⁰ using the GDL laser at the Laboratory for Laser Energetics at the University of Rochester. The SRS fraction depends quite dramatically on scalelength, as discussed earlier. However, at all wavelengths longer than 0.26 μm, the SRS fraction is independent of wavelength within experimental uncertainties.

Three observations are relevant here. First, effects of the decrease in L/λ^2 from 1.06 to 0.35 μm are apparently offset by effects of the increase in L/λ . As a result, there is no substantial change in the SRS fraction. Second, SRS responds much less to changes in wavelength (and consequent increases in inverse-bremsstrahlung absorption) than does SBS, suggesting that SBS occurs at higher densities than SRS. Third, the decrease in the amount of SRS from the CH foil at 0.26 μm might be an effect of inverse-bremsstrahlung absorption.

We also observed hard x rays produced as a result of SRS by means of the bremsstrahlung x rays emitted by such electrons when they slow down in a target. The amount, temperature, and timing of these hot electrons were in quantitative agreement with the expected production of hot electrons by the plasma waves that grow during SRS.^{144,145}

In summary, we observed the following trends from SRS data:

- SRS sidescattering develops in large plasmas.
- The total amount of SRS increases as the gradient scalelength L increases.
- The amount of SRS is not a strong function of irradiating wavelength.
- SRS can be quenched by collisional damping.
- The saturation level of SRS in the absence of collisional damping is not known.

Conclusions. During the 1980s, we have produced relatively large, warm, planar plasmas using multikilojoule

lasers operating at four different wavelengths. Although the pulse lengths and scalelengths are not as large as those that will exist in a high-gain laser-fusion target, they are large enough to exceed thresholds or to provide substantial gain for several interaction processes that are sensitive to gradients in the plasma. Experiments using such plasmas may well allow us to extrapolate with confidence to high-gain conditions.

Our experiments have, indeed, produced new effects and some clear trends. Backscatter and sidescattering at frequencies near ω_0 are present, but their strong saturation with laser intensity and their significant reduction as laser wavelength decreases lead us to believe that, at some laser wavelength from 0.26 to 0.53 μm , SBS will not prevent laser fusion from working. One minor mystery that remains is that it appears that SBS does not take place at densities well below $0.25n_c$.

It is now clear that SRS and the consequent hot-electron production can be controlled by collisional damping, although it may necessitate some undesired design tradeoffs in some types of laser-fusion targets. It is not yet clear at what level the SRS will saturate in the absence of collisional damping. The latter issue remains a fruitful topic for further experimentation.

We can draw several conclusions with regard to the effect of interaction mechanisms in underdense plasma on the prospects for laser fusion. First, the plasmas produced and studied using multikilojoule lasers are large enough and planar enough that we have found evidence of every process that is expected, from theoretical considerations, to be present in a high-gain target (except for filamentation, which is probably present and undetected). Second, available data indicate that no known process (including two-plasmon decay and forward-Raman and resonance absorption, which were not discussed here) scales in such a way that it definitely prevents laser fusion from working for laser wavelengths well below 1 μm . Third, based on present knowledge, the effects of interaction mechanisms in underdense plasma are not negligible. They appear likely to affect the choice of driver wavelength, laser

intensity, and laser-beam smoothness or bandwidth, as well as the details of target design.

The case for the viability of laser fusion is much more complete than it was in 1980. However, the case for any specific driver and target design is not yet complete. To have maximum impact on ICF, future research should focus on the issues now remaining to place more definite constraints on the properties of the driver and target.

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Brillouin Scattering from Exploding Foils. Stimulated Brillouin scattering is a parametric instability in which an electromagnetic wave decays into another electromagnetic wave and an ion wave. The instability has long been of concern in ICF studies because of its potential for scattering large amounts of the incident laser light. Experiments with 1.06- μm light incident on CH targets have shown that 20 to 40% of the incident light energy is scattered.¹⁵¹ Although recent experiments¹⁵² at shorter wavelengths (0.35 μm) have observed tolerable levels of SBS (<2%), this observation could be due to relatively short plasma-density scalelengths, which limit convective SBS growth. In addition, the observed wavelength shifts in backscattering are not well understood, implying that the sources of SBS and their locations in the plasma—the factors we need to know to extrapolate SBS behavior to fusion targets—are not well characterized.

In our experiment, we used one arm of the Nova laser to irradiate 3- μm -thick CH (polyethylene) foil targets and 0.4- μm -thick gold targets with 0.35- μm laser light. We varied the intensities from 3×10^{13} to 4×10^{15} W/cm², the square-pulse lengths from 4.4 to 1.0 ns over the intensity range, and the spot size from 1100 to

Fig. 3-93. Contour plots of digitized, spectrally resolved streak data for incident intensities of (a) 1×10^{15} W/cm², (b) 1×10^{14} W/cm², and (c) 3×10^{13} W/cm².

250 μm . These values were used to produce (after the foils burned through) an underdense plasma with a plasma density scalelength L ($L/\lambda_0 \approx 1000$) and electron temperature T_e (from 1 to 3 keV) that were nearly independent of the incident laser intensity.¹⁵³ Because each Nova laser arm can provide up to 2.1 kJ of 0.35- μm light, we are able to produce plasmas that are nearly 1-D ($L > c_s \tau$). In addition,

once the foil burns through, the electron temperature is nearly independent of time until the end of the laser pulse.

We primarily discuss spectrally and temporally resolved measurements of light backscattered into the focusing lens.¹⁵⁴ This light is optically relayed to a 0.5-m Czerny-Turner spectrometer (0.5- $\text{\AA}$ resolution) with an S-20 streak camera attached to provide 30-ps temporal

resolution. A photodiode array provided time-integrated, angular-resolved data at 351 nm (bandwidth = 10 nm), and a calorimeter measured the total backscattered energy near the incident wavelength.

Figures 3-93(a) through (c) summarize the time-resolved data from CH foils and show streak data for incident intensities of 1×10^{15} , 1×10^{14} , and 3×10^{13} W/cm², respectively. Each contour level is a constant relative intensity; the intensity interval between contours is constant and identical for all three plots. The plots of relative intensity vs time are taken along the wavelength of maximum backscatter.

We note three interesting features from these data: First, a measurable backscatter signal is observed for the entire duration of the laser pulse. Second, the wavelength shift is nearly constant throughout the laser pulse. Third, the backscattered signal is brightest during the early part of the pulse, the duration of the brightest portion increases as beam intensity decreases, and the duration of the brightest portion correlates well with the burn-through time calculated by the 1-D LASNEX code. The wavelength shift is 5 Å towards the blue for the highest intensity and is unshifted for the lowest intensity.

By comparing the wavelength shifts from targets at two different angles, we can infer both the sound speed and the Mach number at which SBS occurs, as described in Ref. 155. The measured shifts of -5.1 Å for normal incidence and 0.2 Å for 50° incidence (before the foils burned through) give a sound speed of 3.8×10^7 cm/s, assuming the scattered light comes from a density of $0.2 n_c$. This value is close to the sound speed predicted by LASNEX (4.8×10^7 cm/s). We also calculate a Mach number of 1.5; comparison with LASNEX velocity and density profiles (see Fig. 3-94) shows that the SBS is probably coming from densities less than $0.25 n_c$.

The backscattered signal before the foil burns through is probably being driven by light reflected from the critical surface. Note that the reflected light does not have to be Doppler shifted to match the wavelength of the backscattered light in order to enhance SBS; there is both theoretical^{156,157} and experimental¹⁵⁸

Fig. 3-94. LASNEX-calculated values of (a) expansion velocity and sound speed and (b) electron density vs axial distance from the initial target surface before foil has burned through ($t = 230$ ps).

evidence to support this argument. After the foil burns through and the critical surface goes away, the ion fluctuations formed in the early part of the pulse are probably large enough to be maintained by the incident light alone. Because the SBS is coming from low densities and persists after the critical surface is gone, it is unlikely that a suggested model¹⁵² involving filament collapse at the critical surface is responsible for the blue-shifted SBS. Note also that SBS at densities less than $0.25 n_c$ can interact with and suppress stimulated Raman scattering in these regions.¹⁵⁸

The gold-foil data are different from those for CH targets. Scattered light was observed for the first 300 ps of the laser pulse regardless of the laser intensity, and the scattered light wavelength

Fig. 3-95. Scattered signal from a gold target.

shifted towards the red with time (see Fig. 3-95). The time-dependent wavelength shift is most likely due to the change in the electron temperature as a function of time. The duration of the backscattered pulse is possibly a result of a collisional absorption mechanism since this phenomenon is also observed in thick gold targets (where the scattered signal turns on again at later times, but at a longer wavelength). Simultaneous measurement of the backscattered light at two different angles shows that the backscattered light near the start of the laser pulse is coming from a region of very small Mach number. This result implies that SBS originates close to the critical surface. Thus, in contrast to the CH foil case, we do not expect to see much interaction between SBS and SRS in these targets.

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Experiments on $3\omega/2$ Emission. The two-plasmon decay (TPD) instability¹⁵⁹⁻¹⁶² is the parametric decay of an electromagnetic wave into two electrostatic waves in the region of plasma where the electron plasma density n_e is one-quarter the critical electron density n_c of the incident laser light. The instability has long been of interest both theoretically and experimentally,¹⁶³⁻¹⁶⁶ in part because the plasma waves generated by TPD can, in turn, produce suprathermal electrons¹⁶⁷ that can degrade the

performance of ICF targets. The TPD plasma waves can recombine with incident laser photons to produce photons near half-harmonics of the incident laser frequency. This emission usually appears spectroscopically as a pair of peaks, one upshifted and the other downshifted, with respect to the half-harmonic of the laser frequency. The amount of splitting between the two peaks has been investigated as a possible electron-temperature diagnostic.¹⁶⁴

Previous investigations of three-halves harmonic emission from plasmas with incident laser wavelength $< 1 \mu\text{m}$ have been time resolved^{163,167} or angularly resolved^{164,165} while simultaneously spectrally resolved. Detailed Thomson scattering measurements in CO_2 laser experiments^{168,169} have been conducted, which directly measured the electron plasma waves from TPD. A reoccurring problem in the interpretation of experimental data has been the lack of agreement with theoretically predicted angular distributions of the scattered light.

In our experiment, the emission was simultaneously spatially and spectrally resolved, with the result that the blue-shifted and red-shifted features are seen to be originating from spatially different regions of the plasma. Although observation of transmitted $3\omega/2$ through thin foils^{163,165,167} that saw only the blue-shifted component supports the notion of forward-propagating blue-shifted plasmas, our data represent the first direct confirmation of this process.

Our diagnostic technique consisted of imaging the target through a 0.5-m Czerny-Turner spectrometer onto a 2-D optical multichannel analyzer (OMA). The entrance slit of the spectrometer was aligned with the laser axis so that one dimension of the detector is space z and the other dimension is wavelength λ (see Fig. 3-96). The target was backlit with a $3\omega_0$ probe beam, which appeared in second order at the OMA as a $3\omega/2$ wavelength fiducial. Measurement of the FWHM of this line shows that the spectral resolution is limited by the crosstalk of the OMA channels to approximately 0.4 Å. The spatial image was aligned at the OMA by illuminating an alignment target with a helium-neon laser and

Fig. 3-96. Layout of imaging diagnostic. Inset shows orientation of the image relative to the entrance slit of the spectrometer.

focusing the image onto a ground-glass screen, which temporarily replaced the OMA detector. The spatial resolution of the optical system corresponds to approximately $10\ \mu\text{m}$ at the target plane. Some smearing of the image occurs because the OMA records a time-integrated picture of emission, which moves with time.

The laser pulses were $\lambda_0 = 1.06\ \mu\text{m}$ and Gaussian in time with a FWHM of 100 ps. The data presented here are for a spot size of $100\ \mu\text{m}$ and an intensity of $1 \times 10^{15}\ \text{W}/\text{cm}^2$, values that are comparable to the parameters in Refs. 163 and 164. The targets were thick molybdenum slabs, which did not burn through during the laser pulse. Many shots were obtained at varying spot sizes and incident energies; the observed threshold of the emission appears to be approximately $1.5 \times 10^{14}\ \text{W}/\text{cm}^2$. We checked the dependence on incident laser polarization by comparing observations in-plane and out-of-plane; no signal was observed in the out-of-plane direction, and the data reported here were observed in the in-plane direction. We verified that the observed emission was not associated with the probe beam and that it was a first-order feature, not second-order.

Figure 3-97 shows typical data and several interesting features. As with previous measurements of $3\omega/2$, we observed features upshifted and downshifted from the center frequency; the spectral feature

Fig. 3-97. Spectral data vs spatial position. The peak at $3\omega/2$ is a wavelength fiducial. Note the region where there is a red-shifted feature but no blue-shifted feature.

that is downshifted in frequency is somewhat brighter than the other component. It is evident from the data in Fig. 3-97 that these components are clearly separated in space, with the upshifted component coming from a region nearer the target surface.

By separately summing over wavelength for both the shifted features seen

Fig. 3-98. Sum over wavelength vs spatial position of data in Fig. 3-97 showing spatial separation of the two shifted features.

Fig. 3-99. Data in Fig. 3-97 averaged over space to give an integrated spectrum. The wavelength shift of the peak of the red-shifted feature is slightly larger than that of the blue-shifted feature.

in Fig. 3-97 and plotting as a function of space, we see from Fig. 3-98 that the peak emission for the two features occurs in different places. This observation could be consistent with theories that account for the emission by plasmons from $n_e/4$ propagating upstream and downstream to regions where they can k match with the pump to produce electromagnetic waves. It is also interesting that the blue-shifted feature peaks well away from the target surface, suggesting that light rescattered from the $9n_e/4$ surface¹⁶⁵ is not the source (for sidescattered light at least). We would expect the density gradients to be steep at $9n_e/4$, which will cause significant refraction of $3\omega/2$ light and could result in either a loss of light if the refracted rays miss the collecting lens of the diagnostic or else an error in the observed spatial position of the emission. The latter spatial error will be on the order of the focusing error of the diagnostic (see, for example, Ref. 170). The focusing error is typically about $10\ \mu\text{m}$.

If, instead, we are losing a larger amount of light from the target surface, we would expect to see a strong angular dependence of the ratio of the strengths

of the two components. This result is not seen for comparable experiments using gold targets.¹⁶³ From this argument, we conclude that while forward-scattered $3\omega/2$ light is probably present, it is by no means the dominant source for the observed, backscattered, blue-shifted component. We also note that this conclusion is consistent with the observation in thin burn-through foils¹⁶⁷ indicating that the backscattered, blue-shifted feature persisted after the $9n_e/4$ surface disappeared.

Another interesting feature of our data is the presence of spectral structure in both peaks. The structure in the red-shifted peak in particular often appears to be periodic in wavelength. The phenomenon of periodic structure has not been observed before in volume-averaged diagnostics of plasmas formed with $\lambda_0 < 1\ \mu\text{m}$, and, as is evident from Fig. 3-99 (which is a sum over space of the data in Fig. 3-98), we would not expect to see such structure in a volume-averaged measurement. Spectral structure has been observed in CO_2 laser plasmas.¹⁶⁹ Our observation suggests that scattering off ion waves occurs during the process between the formation of TPD plasmons and the production of $3\omega/2$ photons.

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Ponderomotive Filamentation Experiments. A laser beam that is incident on an initially uniform plasma with spatial variation (or a hot spot) in the beam intensity produces a ponderomotive force that drives electrons towards regions of lower intensity. The index of refraction in a plasma, N , is given by $N = (1 - n_e/n_c)^{1/2}$, where n_e is the electron density at which the electron oscillation frequency ($\omega_p = 4\pi n_e e^2/m_e$) equals the laser frequency and m_e is the electron mass. The region of lower electron density acts as a convex lens and produces a curved phase front. As a result, the variation in spatial intensity is focused, and the electric-field gradient is enhanced. This process iterates until the electron density in the hot spot is too

low to further refract the beam. This instability is known as ponderomotive filamentation.¹⁷¹ If the perturbation in electron density is also driven by localized heating, then the instability is called thermal filamentation.¹⁷²

The filamentation instability is of particular concern in ICF applications where symmetry of the radiation imploding a pellet is important.¹⁷³ Filamentation is of more general interest because of its role in interacting with parametric instabilities in laser-produced plasmas. Filamentation can produce local laser intensities and density scalelengths that are much different from the average plasma values. The difficulties in understanding filamentation include being able to identify filaments in typical laser plasmas and to provide firm experimental support for theoretical predictions of filament characteristics. Typical laser-beam cross sections have large intensity variations, which can seed the filamentation instability. Methods are being considered that should substantially reduce the instantaneous¹⁷⁴ and time-averaged¹⁷⁵ intensity fluctuations, but we would like to know how large a fluctuation can be tolerated. Hot spots in the beam profile are usually distributed randomly or in diffraction rings, which can change as a function of laser intensity.¹⁷⁶ This property makes diagnosis of the instability difficult.

Earlier experimental investigations of the filamentation instability in laser-produced plasmas used various diagnostics to infer the presence in the plasma of filaments. These diagnostics include x-ray photographs,¹⁷⁷⁻¹⁸⁰ interferometry and shadowgraphy,^{181,182} imaging of backscattered light,¹⁸³ angular distribution of $3\omega/2$ plasma emission,¹⁸⁴ imaging and angular distribution of 2ω emission,^{185,186} dark-field imaging,¹⁸⁷ and Thomson scattering from electron plasma waves generated at $n_c/4$ in the filament wall.¹⁸⁸ Such investigations have been unsatisfactory, in part because most have relied on indirect identification of filaments.

We would prefer to measure directly the density depressions produced by the instability and to correlate them with intensity fluctuations in the laser beam. This type of approach is usually difficult because the spatial cross section of the

filament should be quite small (on the order of the laser wavelength for ponderomotive filamentation). However, density-dependent diagnostics, such as interferometry and Schlieren, are then insensitive because of the small path lengths involved.

Another serious limitation of past work has been the use of uncharacterized noise sources to seed the filaments (i.e., spatial fluctuations in the laser intensity or plasma densities). This limitation results in effectively driving filamentation at different (uncharacterized) wavelengths, resulting in various growth lengths and filament sizes in a single plasma. As noted above, spatial fluctuations can vary as a function of laser intensity.

In our experiment, we used a technique that allows us to conduct a systematic investigation of filamentation. We drive filamentation in a preformed, underdense plasma with a well-defined spatial-intensity perturbation that dominates the intrinsic intensity noise of the laser. Having a well-defined perturbation wavelength makes it easier to identify filamentation structure. By systematically selecting the perturbation wavelength, the driving beam intensity, and, to some extent, the plasma density for each target, we have a sufficient number of tools to explore the dependence of filamentation characteristics on plasma conditions.

Fig. 3-100. Layout in target chamber room of the Phoenix laser facility. The 3ω probe beam can be optically delayed relative to the two 1ω beams. A polarizer allows both the plasma-forming beam and the filament-driver beam to use the same focusing lens; the two beams have orthogonal polarizations.

A principal advantage of our experimental geometry is that the filaments tend to form in sheets so that a probe beam aligned tangential to the sheets will have a large path length through the filament. The combination of the density gradient at the filament wall with the large path lengths means that we should expect significant refraction of the probe beam by the filaments. A diagnostic, such as dark-field imaging, then becomes an important tool for detecting refracted rays and, therefore, identifying filament formation. In addition, it becomes feasible to use interferometry as a second diagnostic to look for the relative phase change between the inside and outside of the filament structure. Our third diagnostic is a target-plane imager that images the transmitted beam at the target plane onto film so that we can look for enhancement of or other change in the modulated beam structure.

We used the Phoenix laser facility, shown in Fig. 3-100. The plasma was formed with a 100-J, 1-ns, 1.06- μm laser pulse incident on a 1- μm -thick CH foil target. These parameters were chosen to produce a plasma density, temperature, and size to provide conditions for which ponderomotive filamentation should be above-threshold. The filament-driving beam was modulated by a pair of partially reflecting plates; the depth of modulation was varied by changing the reflectivity of the plates; the perturbation wavelength was varied by changing the angle between the plates. The driving beam was a 100-ps, 1.06- μm laser pulse of variable energy. A 100-ps, 0.35- μm probe beam was synchronized with the driving beam.

Figure 3-101 shows theoretical predictions¹⁸⁹ for the stability of ponderomotive filamentation as a function of perturbation wavelength. An instability condition occurs when the laser intensity increases exponentially, as shown by the horizontal line in the graph. For comparison, the properties of thermal filamentation are also shown. Note that not only is thermal filamentation predicted to be stable for the chosen plasma conditions, but it is also limited to perturbation wavelengths larger than the electron mean free path. Thus, for very short perturbation wavelengths, only ponderomotive filamentation can grow.

Figure 3-102 shows our early results from the dark-field imager. In Fig. 3-102(a), the modulated beam intensity is $5 \times 10^{13} \text{ W/cm}^2$, the perturbation wavelength is

Fig. 3-101. Comparison of calculated thermal and ponderomotive growth length vs instability k for the listed plasma parameters. Here, $\text{Im } k = 2\pi/\text{instability growth length}$, and $\text{Re } k = 2\pi/\text{perturbation wavelength}$.

Fig. 3-102. Dark-field images (a) with the filament-driving beam above threshold, (b) without a preformed plasma, and (c) with the filament-driving beam below threshold.

40 μm , and the filament structure is quite evident. Figure 3-102(b) shows a null case with a lower driving-beam intensity of $2 \times 10^{13} \text{ W/cm}^2$ and the same perturbation wavelength. Here we see no evidence of filaments. The spatial profile of the filament-driving beam (with an alignment cross hair superposed) is shown in Fig. 3-102(c).

Figure 3-103 shows contours of plasma density corresponding to the plasma of Fig. 3-102(a). The interferometer shows that the filaments begin at a finite plasma density; the dark-field picture suggests that the filaments begin on isodensity and isothermal contours in the plasma. Unfolding of the interferometer shows that the background density is approximately $0.1 n_c$. The results are in qualitative agreement with theory.

Author: P. E. Young

X-Ray Emission from Gold and Beryllium Mixtures. This year, we addressed the effects of material opacity on the interaction of $0.35\text{-}\mu\text{m}$ laser light with high-Z materials and the conversion of laser light to x rays. Our experiments were designed to vary the number density of high-Z radiating atoms by controlling target constituents.

Our work involved the dilution of gold with beryllium in the target. The irradiated targets were pure beryllium, pure gold, and 1/833, 1/100, and 1/20 atomic fractions of gold in beryllium. Disk targets were irradiated at $4 \times 10^{14} \text{ W/cm}^2$ with 1-ns pulses of $0.35\text{-}\mu\text{m}$ light from a single arm of the Nova laser. We observed a strong, nonlinear relation between gold fraction and x-ray production for thermal x rays and gold M-band radiation. This observation indicates the strong effects of opacity in the generation of such radiations.

Targets were manufactured using cosputtering to produce uniform dispersal of gold and beryllium and to prevent islands of either material from forming. When large islands are formed, the radiating environment becomes a linear superposition of pure gold and pure beryllium. If the materials are dispersed,

then the integral along a line of sight will always reflect the average atomic proportions of the target. We measured the effects of the changing-target characteristics on x-rays generated in the laser plasma. We used the Dante system to measure x-ray conversion efficiency and the time-integrated spectral distribution of radiation. We also used Henway crystal spectrographs to measure the absolute yield and spectral distribution of gold M-bands.

As shown in Fig. 3-104, x-ray conversion efficiency falls off with gold fraction. The rate of decrease with gold fraction is

Fig. 3-103. Plasma-density contours corresponding to the plasma of Fig. 3-102(a).

Fig. 3-104. X-ray conversion efficiency vs. gold fraction. Error bars represent shot-to-shot variations.

Fig. 3-105. Low-energy x-ray spectra of gold and beryllium mixture as a function of gold fraction.

Fig. 3-106. Total yield of gold M-band radiation vs gold fraction. Experimental data are contrasted to a linear dependence on gold fraction.

Fig. 3-107. M-band spectral changes in gold and beryllium mixtures.

much slower than linear, indicating that more radiation per gold atom is escaping the target; i.e., the opacity of the target is decreasing. This effect can be observed in the time-integrated spectrum from the targets as a function of gold fraction, as shown in Fig. 3-105. The O-band and 4-4 transitions in the region of 200 to 400 eV approach gold-like emission at 1/20 dilution, and more quickly than the N-band emission, which occurs at 600 to 1000 eV. This effect is also observed in M-bands of gold. As shown in Figs. 3-106 and 3-107, the M-band yield and spectrum change with the gold fraction. As the gold fraction increases, the number of ionization states observed in the spectrum also increases.

Our experiments demonstrate that the concentration of radiating material is critical in determining radiation output from a target and that the output does not scale linearly with the fraction of radiators. The decrease in radiative yields is due, in part, to effects of material

opacity. Further study may lead to a better understanding of high-Z opacity and our ability to select mixtures of materials to yield high x-ray conversion efficiencies.

Author: D. R. Kanla

Pulse-Length Scaling of X-Ray Conversion Efficiency. Future ICF targets will require longer laser x-ray pulses to drive the implosions. We performed an experiment to explore the effects of long laser pulses on the conversion of laser light to x rays. The interrelated questions we addressed were, first, how does x-ray conversion efficiency change with pulse length (time), and, second, does the transition from 1-D to 2-D expansion affect x-ray conversion efficiency. Our results indicate that x-ray conversion efficiency does not decrease for long pulses or for the transition to 2-D expansion, in agreement with the earlier measurements.¹⁹⁰

All of our experiments were performed on one arm of the Nova laser with 0.35- μm light. The laser energy on target was in excess of 1900 J and was focused to approximately $5 \times 10^{14} \text{ W/cm}^2$. The laser pulses were nominally flat-topped and up to 4 ns long. We measured incident laser power as a function of time from streak-camera measurements of incident 1.06- μm laser light, which were cross-calibrated to direct measurements of 0.35- μm laser light in a separate calibration experiment. The finite amount of laser energy available required that the focus diameter D be changed to keep the laser intensity constant as pulse length increased. As a result of the decrease in the size of the laser spot with longer pulse lengths, a transition was made between 1-D and 2-D expansion of the laser plasma; i.e., D was less than plasma sound speed C_s times laser pulse width t_L . This relation can be written as:

$$C_s t_L < D \quad (42)$$

for 1-D expansion. Using the expression for average ionization, sound speed, and temperature for laser-produced gold

Fig. 3-108. X-ray conversion efficiency ($E < 1.5 \text{ keV}$) for 0.35- μm light on gold disk targets vs pulse length. Intensity $\approx 4 \times 10^{14} \text{ W/cm}^2$.

plasmas from Ref. 191 as a function of laser intensity, the criterion can be written as

$$I_{14} - 8.7E^{2/3}t_L^{-2} < 0, \quad (43)$$

where I_{14} is the laser intensity in 10^{14} W/cm^2 , E is the laser energy in kilojoules, and t_L is in nanoseconds. For operating conditions with one arm of the Nova laser, transition to 2-D expansion occurred for pulse widths greater than 2 ns.

We measured the total time-integrated x-ray yield with the Dante soft x-ray spectrometer and the time-resolved soft x-ray emission with a spatially discriminating streak spectrograph (SDSS).¹⁹⁰ Figure 3-108 shows the Dante results for x-ray conversion efficiency $E < 1.5 \text{ keV}$ as a function of laser pulse length. The laser intensity in the data varies from 3 to $7 \times 10^{14} \text{ W/cm}^2$. The longer pulses convert as well as, or better than, 1-ns pulses and significantly better than 100-ps pulses. Although the intensity varies, Fig. 3-109 shows that data for longer pulses are consistently higher than data for 100-ps and 1-ns pulses, indicating that the effect is not a result of intensity dependence of conversion efficiency.

Our results are consistent with measurements of time-resolved conversion efficiency. The plot of conversion efficiency vs pulse length is a rough integral

of the time-dependent conversion efficiency measured by the SDSS. Theoretical expectations are that 2-D effects are detrimental to x-ray conversion efficiency, perhaps indicating that the

Fig. 3-109. X-ray conversion efficiency vs intensity. Data from long-pulse experiments are higher than those for 100-ps or 1-ns pulses, independent of intensity variations.

Fig. 3-110. Average conversion efficiency vs pulse length from Ref. 192 and the present experiment.

Fig. 3-111. (a) Long, single-pulse power at focal spot vs time and (b) picket-fence laser power at focal spot vs time.

present, long-pulse measurements are a lower bound for larger focal spots and higher-laser-energy experiments to be performed in the future.

Figure 3-110 shows the average conversion efficiency from our experiment together with data from Ref. 192. Error bars represent shot-to-shot variations in the data. The trend up to 3-ns pulses is for conversion efficiency to increase. The 4-ns pulses show smaller conversion efficiency than that for 3-ns pulses, but the value is still larger than that for 0.1-, 1-, and 2-ns pulses.

Our study of long-laser-pulse x-ray conversion efficiency shows that the process is at least as efficient for pulses longer than 2-ns as it is for 1-ns pulses, and it is definitely superior to conversion efficiency for 100-ps pulses. The long pulses are in the regime of 2-D expansion; however, we observed no decrease in x-ray yield.

Author: D. R. Kanla

Time-Resolved X-Ray Conversion Efficiencies of Laser-Driven Plasmas. We have completed the analysis of spectrally and temporally resolved soft x-ray data taken during the first phase of Nova experiments on x-ray conversion efficiency. By comparing x-ray yield rates with laser energy-absorption rate, we were able to infer instantaneous x-ray conversion efficiencies. Such information is important in studies of x-ray emission dynamics from such plasmas and is essential to accurately tailor x-ray flux into a capsule in x-ray-driven laser fusion.

(a) Diagnostic setup

(b) Single-pulse experiment

(c) Picket-fence experiment

Our experiments involved the use of both single pulses (~ 2.2 -ns FWHM) and pulses in a picket-fence configuration in which a main ~ 1.0 -ns pulse was preceded by two, small prepulses of ~ 100 ps to initiate and preheat the plasma. Figures 3-111(a) and (b) show representative temporal profiles of pulse power at the focal spot for the single pulse and picket fence, respectively. The ratio of pulse amplitudes for the latter was 1:2:7.8. Incident $0.35\text{-}\mu\text{m}$ laser energies for the main pulses varied between 1.0 and 2.0 kJ. Targets were $2000\text{-}\mu\text{m}$ -diam gold disks that were $\sim 30\text{-}\mu\text{m}$ thick. The angles of incidence in all cases were $\sim 28^\circ$. Nominal laser irradiances were in the range of $\sim 10^{14}$ to 10^{15} W/cm^2 . The main pulses were nominally square with ~ 100 -ps rise and fall times. We obtained laser temporal profiles from the usual optical streak cameras with known, flat-field characteristics.

We collected x-ray data by looking at irradiation spots with a broadband, multichannel, soft x-ray spectrograph^{193,194} coupled to a streak camera with ~ 20 - to 50 -ps resolution. Angular lines of sight relative to the target normal were $\sim 40^\circ$. Figure 3-112 shows the diagnostic setup: the spectrograph's six channels consisted

of x-ray transmission filters paired with grazing-incidence x-ray mirrors to select specific x-ray energies. Filter absorption edges ranged from ~ 190 eV (boron channel) to ~ 1.30 keV (magnesium channel). Cross-calibration relative to an absolutely calibrated x-ray diode system (the Dante,¹⁹⁵ simultaneously fielded during the series) helped to obtain spectral intensities in absolute units.

Figures 3-112(b) and (c) show representative streaked-spectrograph film data from a 2.2 -ns, single-pulse experiment and a picket-fence experiment, respectively. In each figure, photon energies vary horizontally, while time varies vertically. Figures 3-113(a) and (b) show the respective time histories of the most intense signal for each case. X-ray fluences are along the y-axis, time is along the x-axis, and the respective irradiation parameters are listed. Laser energies were ~ 1.0 kJ for the single pulse and ~ 1.4 kJ for the picket fence. Inferred laser energy absorptions from measured scattered light were quite high ($> 90\%$) for both cases despite the difference in nominal irradiation intensities.

Spectral emissions from the prepulse-driven picket-fence experiments with relatively lower irradiance and higher laser

Fig. 3-112. (a) Time-resolved, broadband x-ray spectroscopy of laser-driven plasma radiation. Representative data from (b) single-pulse and (c) picket-fence experiments.

Fig. 3-113. Temporal profiles of brightest signals from (a) single-pulse experiment and (b) picket-fence experiment.

energy were, in general, more intense than those from the long-single-pulse experiments with higher irradiance and lower laser energy. Because emission intensities are implicitly determined by spatial profiles of plasma density and temperature, our data suggest that the two types of laser temporal profiles created plasma conditions that were significantly different from each other. It is possible that, by preheating the plasma and allowing it to temporally undergo some free expansion before the main pulse was applied, the technique caused temperature gradients in the picket-fence experiments to steepen more sharply than was the case with single-pulse experiments, thus enhancing emission coefficients.

Figures 3-114(a) and (b) show representative instantaneous spectra at selected times vs photon energies for both types of experiments. The continuous lines through the data are Planckian fits. The times at which the spectra were measured are shown in Fig. 3-113. The plots show that the spectra were not Planckian—at least for photon energies covered by the diagnostic ($0.1 < h\nu < 1.3$ keV)—a result that was not surprising. In addition, the picket-fence-induced spectra consistently peaked at somewhat higher photon energies than did the single-pulse-generated spectra (~ 600 vs 500 eV), a result that could signify greater N-band contributions. The result could be best confirmed by doing time-resolved higher-resolution spectroscopy.

Fig. 3-114. Representative spectra at selected times for emissions that were (a) single-pulse induced and (b) picket-fence induced.

Fig. 3-115. Single-pulse results for (a) time-resolved x-ray yield and laser energy deposition rate and (b) resulting time-resolved x-ray conversion efficiency.

By integrating over data fits similar to ones shown from 0 to ∞ , we were able to estimate x-ray yields at various times. The yields are plotted vs time in Fig. 3-115(a) for the single pulse. For comparison, we also plotted the temporal profile of laser energy-deposition rate inferred from optical streak camera data and energy-balance measurements. Fiducial timing of both the laser pulses and the x-ray signals helped to temporally unfold the results to less than ± 50 ps.

Instantaneous x-ray yield increased continuously over the course of the laser application, despite modulations present in beam amplitude. By the time laser absorption began to roll off, x-ray yield rate almost doubled just after the initial

rise time and remained at that level for almost 0.5 ns, although laser energy-deposition rate continued to fall. Figure 3-115(b) shows the ratio of the two curves, representing the instantaneous power balance. The dashed line is a fit to help visualize the overall trend. The conversion rate increased somewhat continuously up to the time the laser pulse reached its peak. An intriguing aspect of our results is that computer models of physics interactions have not predicted this trend, although efforts in this respect are continuing, and future experimental studies are already under way.

Figures 3-116(a) and (b) show similar examinations of results for the picket-fence data. Figure 3-116(a) shows both

Fig. 3-116. Picket-fence results for (a) time-resolved x-ray yield and laser energy deposition rate and laser energy deposition rate and (b) conversion efficiencies.

time-resolved x-ray fluxes and the laser energy-deposition rate. Because x-ray emissions last longer than the driving laser pulse, taking time-resolved ratios in the manner discussed previously are meaningless. However, the general trend of the data can still be examined by determining the mean conversion efficiency at selected time intervals, as summarized in Fig. 3-116(b). The dashed line is a measure of the cumulative efficiency up to a given time; horizontal bars show the mean efficiencies between successive time intervals. Although the latter do not represent instantaneous values, they can be interpreted as an indication that the picket-fence-driven, time-resolved x-ray conversion was also increasing.

In conclusion, we have presented results of the first experimental investigation of potential effects of shaped laser pulses on x-ray radiation from the plasmas they drive. Our data show time-resolved spectra up to photon energies of ~ 1.30 keV that can be approximated as Planckianlike. In addition, we showed enhanced x-ray emissions from plasmas preheated by small precursor laser pulses ahead of the main driving pulses. We also showed that instantaneous x-ray conversion efficiencies continuously increased during irradiation time. Our results on conversion efficiency suggest that spatial profiles of plasma density and temperature may not have reached steady-state conditions. A gradual steepening

of temperature gradients, leading to an increase in radiation emission coefficients and optical depths, could lead to increased plasma-radiation rates in the backward direction, as observed in our experiments. Computer physics models and additional experimental activities are under way to follow up on our investigations.

Author: F. Ze

Low-Energy X-Ray Emission from Uranium Plasma. It is of interest to irradiate uranium disks using single beams of Nova to investigate the Z scaling of conversion efficiency and energy balance. Thus, we performed an experimental investigation of the conversion of $0.35\text{-}\mu\text{m}$ laser light from the Nova laser to thermal x rays from uranium targets.

We systematically studied the change in absolute x-ray conversion efficiency with laser light intensity. We performed all experiments with greater than 1000 J of laser energy in 1-ns, flat-topped pulses onto disk targets. The intensity on the target was varied from 7.5×10^{13} to 3.6×10^{15} W/cm² by varying the laser energy and focused diameter of laser light. We measured the x-ray conversion efficiency using the absolutely calibrated Dante soft x-ray spectrometer. Target absorption was monitored on a representative sampling of shots and was found to be near 95% for all of our experiments. We defined x-ray conversion efficiency

as the fraction of absorbed laser light that is converted to x-rays with energy less than 1.5 keV.

Our targets were not pure uranium; rather, they were alloyed with 7% niobium to improve the material characteristics. The targets were 2-mm-diam, 25- μ m-thick disks mounted on thin glass stalks. Figure 3-117 shows our measurements of x-ray conversion efficiency of the U-Nb targets. As with gold targets, the conversion falls off as laser intensity increases. The scaling of conversion efficiency (CE) with intensity can be fit to the function $CE(\%) = 65 I^{-0.3}$, where I is laser intensity in units of 10^{14} W/cm².

All measurements were taken in a two-month period with pre- and post-calibrations of many of the Dante channels. The effort to complete our experiments on a short time scale and careful calibration of the detectors represents a significant part of the low scatter observed in the data. The Dante spectra show some variation with intensity; but, in general, they show a broad, flat radi-
ation intensity below 1.5 keV. Figure 3-118 shows a sample spectrum.

In conclusion, we measured the x-ray conversion efficiency of uranium irradiated with 1-ns pulses of 0.35- μ m laser light for incident intensities from 7.5×10^{13} to 36×10^{14} W/cm². The x-ray conversion efficiency falls off smoothly as laser intensity increases.

Authors: D. R. Kania
and H. N. Kornblum

M-Shell Spectroscopy of Gold and Uranium. During high-Z interaction experiments, x-ray spectra of M-band emission are routinely recorded using crystal spectrographs. In the 1986 Laser Annual, we reported preliminary measurements of the spectra and intensity of M-band emission from gold and uranium disk targets.¹⁹⁶ We added absolute wavelength fiducials to the spectrographs, allowing us to measure x-ray transition energies absolutely. We report here preliminary analysis of transition energies from gold and uranium M-band spectra.

The M-band spectra were measured with the Henway crystal spectrographs

Fig. 3-117. Uranium x-ray conversion efficiency vs incident laser intensity. Error bars represent 10% error in conversion efficiency and laser intensity; conversion efficiency assumes 95% absorption.

Fig. 3-118. Dante spectrum of a U-Nb target irradiated with a 1-ns, flat-topped pulse of 0.35- μ m light at 4.4×10^{14} W/cm².

on the Nova target chamber. Gold spectra in the range of 1.8 to 3.6 keV are dispersed by a potassium acid phthalate (KAP) crystal ($2d = 26.632$ Å) bent in the form of a convex cylinder with a radius of curvature of 25.4 cm. For uranium spectra in the range of 2.8 to 6 keV, a pentaerythritol (PET) crystal ($2d = 8.742$ Å) bent to a radius of curvature of 10 cm is used. Data from both spectra are recorded on SB-5 x-ray film. The relative dispersion of the x-ray spectra on the film has been checked against known helium-like and Lyman emission spectra from Al, Si, and Cl. The dispersion is accurate to better than 2 eV for every keV of dispersion in the regions of interest. To obtain absolute calibrations, fiducial filters having absorption edges near the energies of interest were placed in front of part of the diffracting crystal. For the gold spectrum, the absorption filter is a

C1 plastic whose potassium absorption edge is at 2.8224 keV. For the uranium spectrum, the absorption filter is Sn whose L_{III} absorption edge is at 3.9288 keV. The position of the edges can be determined accurately to better than 1 eV. The overall accuracy for measuring the line energy is estimated to be ± 3 eV.

Figure 3-119 shows an example of the $n = 3 - 4$ transition region from uranium. The spectrum is produced by irradiating a uranium disk with 1300 J of 0.35- μm

light from a single arm of Nova. The average intensity is about 7×10^{14} W/cm². Similar spectra have been shown for gold previously.¹⁹⁶ Transitions for nickel-like (Ni I) configurations are identified in Fig. 3-119, and their measured transition energies are listed in Table 3-18. These are the first measurements reported of nickel-like transitions from uranium. The uranium data agree with calculations by Scofield¹⁹⁷ and by Bauche-Arnoult et al.¹⁹⁸ Nickel-like transitions have also been measured

Fig. 3-119. Uranium $n = 3 - 4$ spectrum.

Table 3-18. Measured nickel-like line transition energies in gold and uranium compared with calculations.

Transition	Gold				Uranium		
	This	Busquet ^a		Scofield ^b	This	Scofield ^b	Bauche-Arnoult ^{c,d}
	exp.	exp.	calc.	calc.	exp.	calc.	calc.
3d _{3/2} —4p _{1/2}	2.058	2.06	2.063	2.062	3.027	3.029	3.028
3d _{5/2} —4p _{3/2}	2.113	2.12	2.117	2.116	3.163	3.166	3.166
3d _{3/2} —4p _{3/2}	2.203	2.21	2.206	2.204	—	3.350	—
3p _{3/2} —4s _{1/2}	2.372	2.38	2.376	2.378	—	3.408	—
3d _{5/2} —4f _{7/2}	2.512	2.51	2.513	2.513	3.703	3.708	3.706
3d _{3/2} —4f _{5/2}	2.601	2.60	2.602	2.602	3.870	3.874	3.876
3p _{3/2} —4d _{5/2}	2.829	—	—	2.828	4.148	4.156	—

^a From Ref. 199.

^b From Ref. 197.

^c From Ref. 198.

^d From Ref. 200.

in gold. These results also agree with calculations by Scofield¹⁹⁷ and with measurements and calculations reported by Busquet et al.¹⁹⁹

In addition to the nickel-like lines, the uranium spectrum also contains a number of satellite lines at lower energy from the nickel-like $3d \rightarrow 4f$ transitions. These lines are transition arrays of $3d \rightarrow 4f$ lines from configurations having more bound electrons in the $n = 4$ level than the excited nickel-like configuration that has only one electron in the $n = 4$ level. Peaks in the spectra shown in Fig. 3-119 can be resolved from isoelectronic series from Cu I to Br I. Each peak consists of many unresolved transitions from fine structure splitting.²⁰⁰ Similar transitions can be observed in the gold spectrum, but only the Cu I and Zn I satellites are resolvable.¹⁹⁹

Figure 3-120 plots the peak energies of the satellites with respect to the nickel-like lines. The relative shift of the satellites is the same for both the $3d_{3/2} \rightarrow 4f_{5/2}$ and $3d_{5/2} \rightarrow 4f_{7/2}$ transitions. For comparisons, predictions by Bauche-Arnoult et al.¹⁹⁸ for the first three ionization states are shown. The predicted shifts are different for the $3d_{3/2} \rightarrow 4f_{5/2}$ and $3d_{5/2} \rightarrow 4f_{7/2}$ transitions and bracket the measured shifts. Bauche-Arnoult et al.¹⁹⁸ also predicted that the widths of the peaks should be 10 to 12 eV due to broadening by the unresolved transitions. The observed line widths are narrower than these calculated widths but agree within errors of the calculated widths before unfolding of the instrumental widths. More analysis and experiments are needed to understand these features.

Author: R. L. Kauffman

▲ Fig. 3-120. Shifts of the $3d$ - $4f$ lines from uranium vs electron configuration.

Fig. 3-121. Schematic of a dot target.

layer. The rich spectroscopic structure of gold allowed us to observe its heating, eventual ablation, and entering the corona region of laser-plasma interaction.

Our dot target shown in Fig. 3-121 consisted of a $300\text{-}\mu\text{m}$ -diam, $500\text{-}\text{\AA}$ -thick gold dot set within a large plastic disc. The gold was buried under approximately $2\text{ }\mu\text{m}$ of plastic on the interaction side, and the disc was effectively infinite in all other directions. The laser spot was larger than the gold-dot diameter by a factor of 2. We used a single arm of the Nova laser to deliver 2-ns pulses of $0.35\text{-}\mu\text{m}$ light focused to $2 \times 10^{14}\text{ W/cm}^2$.

We used several diagnostics to measure x rays emitted from the target as a function of both time and space. The $22\times$ x-ray microscope measured the

Experiments with Imbedded Gold

Microdots. Hydrodynamics and energy transport are important in understanding the interaction of laser light with high- Z plasmas. This year, we conducted experiments using embedded gold dots to experimentally address these processes.

Our target was an embedded dot consisting of a small, thin disk of gold buried in plastic. The plastic was a low- Z material, which provided excellent contrast in emission with respect to the gold

position of M-band emitting regions over time, the gated crystal spectrometer (GACS) measured the M-band spectrum as a function of time in discrete steps, and the transmission-grating streaked spectrograph (TGSS) measured spectral changes in x-ray emission over time. We filtered the 22 × x-ray microscope to measure gold M-bands (2 to 3.5 keV). When the plastic ablates and gold begins to move into the corona, the region emitting M-band radiation should expand away from the target surface when viewed from the side. We also observed the turn-on time of M-band emission in the TGSS. The time evolution of lower-energy (N-band) radiation indicates that

significant energy has been transported into the target and the gold has heated significantly. Such heating should precede M-band emission; i.e., gold is heated by conduction, eventually ablates away, and enters the corona.

Figure 3-122 shows a sample of our GACS data. Here, the M-bands turn on between 400 to 800 ps into the laser pulse. Figure 3-123 is a lineout of TGSS data showing emission intensity vs time for photon energies near 2 keV. Comparing the time evolution of x rays to the laser, we observed a consistent delay of approximately 500 to 600 ps. The 22 × streaked images, which viewed the target from the side, show a distinct change in the slope (expansion velocity) of the M-band-emitting region in the same time interval. Fig. 3-124 shows the distinct character of dot-target emission in the gold M-band compared to an experiment performed in an identical manner but with a pure-gold target.

By examining the temporal evolution of x-ray characteristics at temperatures lower than those of the M-bands, we can measure the time for conduction to heat the buried gold layer and to begin to ablate the material. A lineout of TGSS data shown in Fig. 3-125 yields the temporal evolution of x-ray emission at this energy. We observe no such delay in either pure-gold or pure-plastic targets. This delay is a direct measure of energy-transport and

Gated crystal spectrograph
interframe time = 400 ps;
open shutter time = 200 ps

Fig. 3-122. GACS data of gold M-bands vs time. The M-bands turn on between the end of the second and the third frame (i.e., after 500 ps).

Fig. 3-123. Time evolution of M-band emission measured by the TGSS compared to the driving laser pulse. The measured delay is ~500 ps.

(a) Dot target, 22 × magnification, 2 ns

(b) Pure-gold target, 22 × magnification, 2 ns

Fig. 3-124. Comparison of expansion of M-band-emitting region of (a) a dot target and (b) a pure-gold target.

ablation rate in the dot target. The ablation rate inferred from our measurement is $7 \times 10^5 \text{ g} \cdot \text{cm}^{-2}/\text{s}$, slightly higher than measurements performed with smaller laser spots at slightly higher intensities.²⁰¹

Our dot experiments have yielded useful information concerning material ablation and energy transport in laser-plasma interactions. Further studies are in progress to assess the effects of nonuniformity in the laser-intensity distribution, which may affect the interpretation of our experiments.

Author: D. R. Kania

K α Fluorescence for Characterizing Gold M-Band Emission. Our experiments were primarily intended to test and normalize fluorescence measurements against measurements of the entire spectrum made by a Henway time-integrated spectrograph. The reason for using a fluorescer to measure the energy in a spectral band, rather than simply measuring the spectrum directly, is that a fluorescer integrates not only over x-ray energy but also over angle. When such an average is of interest, one measurement takes the place of many, and greater accuracy is expected. However, the number of

Fig. 3-125. Time evolution of 650-eV x-ray emission from the dot target compared to the driving laser pulse. The observed delay is 200 ps.

fluorescent photons per steradian is usually drastically less than the number of exciting x-ray photons per steradian because of several factors. First, for the low-Z fluorscers we used in our work, the fluorescence efficiency was small. Second, the fluorescer subtended only a small solid angle, as seen from the source, to allow access to the irradiating beams and lines of sight for the diagnostics. Finally, a beryllium filter reduced the x-ray flux on the fluorescer, and reabsorption of the fluorescence further reduced the

Fig. 3-126. Experimental geometry for measuring spectrally and time-integrated gold M-band emission.

efficiency. The net result was that, in terms of photons/sr, our fluorescence signal was only 0.05% as intense as the total gold M-band flux above the fluorrescer K-edge. Our spectrograph looking at the fluorescence did not view directly the laser-irradiated spot and did not see the coronal plasma until it had moved out many hundreds of microns. Even so, many gold lines were strongly visible.

Description of the Experiment. Figure 3-126 shows our experimental geometry. One or two 3512-Å, 1-ns, square, f/4.3 laser beams were incident at an angle of 56° to the normal. Intensity was varied between 2×10^{14} and 2×10^{15} W/cm², and the energy on target was varied between 1.5 and 4 kJ. The time-integrated Henway 2B spectrograph looked at 11.6° to the normal, and the Dante looked at 15° to grazing. The Dante is a soft x-ray spectrometer with discrete channels and filtered x-ray diode detectors. The center of the fluorrescer was at 46.4° to the normal, and the RAPOS time-integrated spectrograph on SIM2 looked at 47° to the fluorrescer normal. Its line of sight was 7° to the back side of the large, laser-irradiated gold plate. The tab at one end prevented the RAPOS spectrograph from seeing the gold plasma until it had moved out beyond the fluorrescer, a total distance of at least 1 mm.

We drilled a small hole where the beams were incident and patched over

the back side with thin tungsten glass. The hole diameter was many times smaller than the laser-beam spot size. We used this window to center the target transversely to the beams using a polar target-alignment viewer (TAV 1). For alignment of the target along the east-west direction, we placed a long wire at the bottom of the plate, directly in line with the hole. The wire was long enough to be seen by both of the two, equatorial target-alignment viewers (TAV 2 and 3). The fluorrescer was 600 μ m wide by 1500 μ m high. The slit of the spectrograph ran along the width; thus, source broadening of the fluorescence line was minimized.

We used an (002) PET crystal bent to a 4-in. radius and Industrex CX film. The target-to-crystal distance was 40 cm, and the crystal-to-film distance was 3.5 cm for the fluorrescer K α x ray. We took several steps to minimize stray x-ray background on the film. First, the spectrograph was lined with plastic. Fluorescence energy was too low to penetrate the 2 mils of beryllium used at the film cassette. The x-ray diffraction crystal was protected from target debris by another 2-mil-thick beryllium filter. This filter was placed on the end of a snout so that recombination radiation from plasma striking the beryllium filter did not have a direct line of sight to the film. A magnet immediately in front deflected most of the plasma from the beryllium window. The

spectrograph interior was completely enclosed, except for a vacuum pump-out hole with several bends to stop plasma from reaching the interior. The outside of the spectrograph was lined with 1/16-in.-thick tantalum.

Modeling. Because we wished to make precise measurements, we wrote a computer code for radiation transport.

Figures 3-127 and 3-128 show the model, including a detailed view of the fluorescer package. Each element of source differential area excites every element of fluorescer differential area. To obtain the total fluorescer signal, we integrate over both the source and fluorescer areas.

To create a fluorescent photon, an incident x-ray photon must first penetrate the filters at the front and then be absorbed by a fluorescer atom and create a K-shell vacancy, which is filled by a radiative transition rather than an Auger process. This fluorescent x-ray must then get through the rest of the fluorescer and the back filters without being absorbed.

Let

$$\mu_F(E) = \rho_i \kappa_i \Delta x_i \quad (44)$$

and

$$\mu_B(E) = \rho_j \kappa_j \Delta x_j \quad (45)$$

be the summed absorption coefficients for the front filters i and the back filters j , respectively. Let θ_E be the angle the incident x-ray photon makes with the normal to the fluorescer and θ_i be the angle the fluorescent x-ray photon makes to the normal to the fluorescer. Then the transmission of the front filters to incident x rays of energy E is $\exp[-\mu_F(E) \sec \theta_E]$, and the transmission of the back filters to fluorescent x rays is $\exp[-\mu_B(E_i) \sec \theta_i]$. Of course, θ_i is determined by the location of the spectrograph, which we assumed to be far enough away so that θ_i is a constant, and θ_E varies with integrations over the source and fluorescer areas. Similarly, $\mu_{fluor}(E)$ is the absorption coefficient for the fluorescer at energy E .

Let the number of escaping fluorescent photons/sr created by a given energy bin of incident x-rays be $dN_F/d\Omega$, and let the

Fig. 3-127. Model geometry and definition of symbols for radiation transport calculation.

Fig. 3-128. Detail of fluorescer package.

number of incident x-ray photons/sr per unit perpendicular area in a given energy bin and in a given direction be

$$\frac{d^2 N_x}{d\Omega d(\mathbf{n} \cdot \mathbf{A}_s)} \quad (46)$$

For a Lambertian angular distribution of x rays from the source, Eq. (46) does not depend upon direction; whereas for an isotropic distribution, it is proportional to $(\mathbf{n} \cdot \mathbf{n}_s)^{-1}$. Here, $\mathbf{n}$ is a unit vector along the radius vector $\mathbf{r}$ joining a source differential area element to a fluorescer differential area element, and $\mathbf{n}_s$ is a unit vector perpendicular to the source differential area element. The vector differential area elements are $d\mathbf{A}_s = \mathbf{n}_s dA_s$, and $d\mathbf{A}_f = \mathbf{n}_f dA_f$. We then have

$$\begin{aligned} \frac{dN_F}{d\Omega} &= f \cdot \mu_{fluor}(E) \sec \theta_E \\ &\times e^{-\mu_F(E) \sec \theta_E} \cdot e^{-\mu_B(E_i) \sec \theta_i} \\ &\times \frac{e^{-\mu_{fluor}(E) \sec \theta_E} - e^{-\mu_{fluor}(E_i) \sec \theta_i}}{\mu_{fluor}(E_i) \sec \theta_i - \mu_{fluor}(E) \sec \theta_E} \\ &\times \int \int \frac{d^2 N_x}{d\Omega d(\mathbf{n} \cdot \mathbf{A}_s)} (\mathbf{n} \cdot d\mathbf{A}_s) \left(\frac{\mathbf{n} \cdot d\mathbf{A}_f}{4\pi r^2} \right) \quad (47) \end{aligned}$$

where the factor on the third line of Eq. (47) is the result of evaluating the integral

$$\int_0^1 e^{-\mu_{\text{fluor}}(E) \sec \theta_E t} \cdot e^{-\mu_{\text{fluor}}(E_I) \sec \theta_I (1-t)} dt. \quad (48)$$

Here f is the fluorescence probability. Given that the x-ray photon is absorbed in the fluorescer material, a fluorescent photon results with probability f . Then

$$f = \omega_K \left(\frac{N_{\text{fluor atoms/molecule}} \sigma_{\text{fluor atom}}}{\sigma_{\text{fluor molecule}}} \right) \times \left(\frac{\sigma_{K \text{ shell of fluor atom}}}{\sigma_{\text{all shells fluor atom}}} \right). \quad (49)$$

Here $t = x/x_i$, where x_i is the fluorescer thickness, and ω_K is the radiative yield or the probability that a K-shell vacancy in a fluorescer atom is filled by a radiative transition rather than by an Auger process.

To calculate the spectrograph film exposure, we need the sensitivity $dS/dF = S/F$. Here S is the film exposure in photons/cm², and F is the fluence in photons/(keV · sr). However, rather than the peak film exposure itself, we need an integral across the width of the line. For a narrow line, both the dispersion dy/dE and the sensitivity dS/dF are approximately constant. Then

$$\int F dE \approx \frac{\int S dy}{\left| \frac{dy}{dE} \right| \frac{dS}{dF}} \quad (50)$$

The integral $\int F dE$ gives the total line intensity in photons/sr and is independent of the shape of the line. Let

$$\frac{d^2 N_x}{d\Omega d(\mathbf{n} \cdot A_s)} = \frac{d^2 N_x}{d\Omega dA_s} \Big|_{\theta=0^\circ} \cdot I(\theta), \quad (51)$$

where the factor evaluated at $\theta = 0^\circ$ is the total number of x-ray photons/sr per unit area from the source in the direction of its normal for some given energy bin. The angular dependence of the source x-ray emission has been put into the

function $I(\theta)$, where θ is the angle to the source normal and $I(0^\circ) = 1$. Both the derivatives as well as the function $I(\theta)$ depend on the x-ray energy. For simplicity, the energy dependence has not been explicitly shown because attention is restricted to one, narrow energy bin of incident x rays at a time. If it were shown, $I(\theta)$ would be replaced by $I(\theta, E)$, and both second derivatives would actually be third derivatives with an extra differentiation relative to the energy of M-band x rays. In the data analysis, we assume the normalized angular distribution $I(\theta, E)$ is independent of energy.

In the experiments we performed, the ratio

$$\frac{dN_F}{d\Omega} \Big/ \frac{d^2 N_x}{d\Omega dA_s} \quad (52)$$

was relatively constant over that part of the gold M-band spectrum that was above the sulfur K-edge of 2.472 keV. Here $dN_F/d\Omega$ is the number of fluorescent photons/sr escaping unabsorbed in direction Ω from the entirety of the fluorescer. Sulfur was the fluorescer atom in the C₆H₄S poly(phenyl) sulfide plastic. Now $dN_F/d\Omega = F dE$. Thus

$$\frac{d^2 N_x}{d\Omega dA_s} = \frac{\int S dy}{\left| \frac{dy}{dE} \right| \frac{dS}{dF} \left(\frac{\frac{dN_F}{d\Omega}}{\frac{d^2 N_x}{d\Omega dA_s}} \Big|_{\theta=0^\circ} \right)}. \quad (53)$$

The total number of photons emitted into the M-band above the fluorescer K-edge is then

$$N_x = \frac{d^2 N_x}{d\Omega dA_s} \Big|_{\theta=0^\circ} A_s \int_0^{\pi/2} I(\theta) 2\pi \sin \theta d\theta. \quad (54)$$

The angular integration has been shown as only over one hemisphere. Of course, because the emitting region moves in front of an irradiated plate of finite size, emission into the backward hemisphere is possible. A 2-D LASNEX calculation shows that the angular distribution of the 2- to 4-keV x rays emitted by a gold disk

Table 3-19. Summary of fluorescence experiments.

Shot number	Intensity (W/cm ²)	Assumed angular distribution	2.472 — 3.5 keV		2.0 — 3.5 keV
			RAPOS results (%)	Henway 2B results (%)	Henway 2B results (%)
17103004	4.9×10^{14}	Lambertian	3.7	4.2 ^a	6.9
AUMSKA-01	3100 J	Isotropic	4.2	8.3	13.4
17110309	2.3×10^{14}	Lambertian	4.8	3.3 ^b	5.3
AUMSKA-02	1465 J	Isotropic	5.5	6.6	10.3
17110313	1.0×10^{15}	Lambertian	3.9	3.7 ^a	5.9
AUMSKA-03	3350 J	Isotropic	4.4	7.3	11.6

^a $a = 0.32$, $b = 2.3$, and $ab = 0.736$. This set of a and b coefficients gives 24% higher fluxes than below the set at small density.

^b $a = 0.28$, $b = 3.25$, and $ab = 0.91$.

Sulfur K α Fig. 3-129. Spectrum showing sulfur K α fluorescence line and lines from the gold coronal plasma.

irradiated with 3512-Å laser light is about midway between a Lambertian and an isotropic angular distribution.

Results. Table 3-19 summarizes our measurements. All percentages for the gold M-band emission are of the laser energy on target. The fluorescence measurement gives a photon number, not an energy. These photon numbers have been multiplied by 2.75 keV to obtain energies. Because much of the solid angle subtended by the fluorescer, as seen from the source, is around 60° to source normal, the energies obtained from the fluorescence measurements do not depend much on whether the angular distribution is isotropic or Lambertian. The Henway 2B, however, looks nearly down the source normal; thus, we obtain a factor of 2 difference, depending upon which distribution is chosen. Because Lasnex simulations suggest that the distribution is about midway between the two, we should take an average of the two numbers.

The Henway 2B spectrum is compared with the RAPOS spectrum for shot number 17110313 in Figs. 3-129 and 3-130. The

RAPOS spectrum consists of relatively few, narrow lines because it sees only a low-density plasma, which has had a long time to cool. For a sufficiently tenuous plasma, electron excitation, ionization, and recombination rates will be long compared to the time for an ion to radiatively decay to its ground state. Nickel-like lines to the $1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10}$ ground state predominate. The gold plasma radiates little around the energy of the sulfur K α line.

The Henway 2B spectrum in Fig. 3-130 shows that most of the emission is actually not in those strong lines visible in the RAPOS spectrum, but rather in overlapping arrays of lines. The SB5 film has been exposed to a high-enough density such that the film sensitivity $dD/d\ln(I)$ has become small. The result is that a small error in the film model can make a large fractional error in inferred intensity. The film model we used is the Henke film model, with physical parameters measured by Henke and Stone and with film constants $a = 0.32$ and $b = 2.3$ for a matched numerical aperture of 0.25.

LASNEX calculations predict that the fraction of laser energy that goes into

Fig. 3-130. Time-integrated spectrum measured by the Henway 2B, which looked directly at the laser-irradiated spot at nearly normal incidence.

Fig. 3-131. Comparison of LASNEX calculations for the M-band energy fractions to those obtained from fluorescence experiments.

gold M-band x rays is much higher at higher intensities (see Fig. 3-131). Our experiments to date do not support this prediction, but they extend over only a narrow intensity range (2.3×10^{14} to 1.0×10^{15} W/cm²). Our future experiments will be at both lower and higher intensities and at both the 56° angle of incidence and at normal incidence. They will also be done with multiple fluorescers so that we can obtain some spectral information as well as additional checks on the measurements.

The fluorescer is heated sufficiently by the incident gold M-band flux so that substantial ionization of the M shell probably occurs. Table 3-20 summarizes the transition energies for the $K\alpha_1$, $K\alpha_2$, $K\beta_1$, and $K\beta_2$ energies. The fluorescence yield does not change much with the ionization state or the L-shell temperature until the sulfur becomes quite highly ionized. The 3p subshell of the M shell is assumed to be statistically populated. In this calculation, electrons are removed first from the 3p subshell, then from the 3s subshell, and finally from the L shell. The various configurations of the L shell are populated according to temperature. For $T = \infty$, the L-shell states are statistically populated. The energy of the $K\alpha_1$ line shifts upwards in energy by about 1.35 eV per M-shell electron removed according to our calculations. Ionization of the first three electrons in the L shell shifts the energy upwards by 16.5 eV per L-shell electron removed. We chose a low-Z plastic with a relatively low atomic percentage of sulfur so that the heat capacity per sulfur atom would be increased without significantly increasing the deposited energy per sulfur atom.

Table 3-20. Sulfur K α , K β transition energies and radiative yields.

Sulfur ion species	Additional ionization energy	K α energy (eV)	K α energy (eV)	K β energy (eV)	K β energy (eV)	K α , β fluorescence yield						
						0 eV	10 eV	20 eV	30 eV	40 eV	50 eV	∞
+ 0	0.00	2308.4	2306.6	2469.65	2469.31	0.0789	0.0796	0.0801	0.0803	0.0804	0.0805	0.0807
+ 1	10.36	2310.0	2307.9	2472.90	2472.30	0.0793	0.0799	0.0805	0.0807	0.0808	0.0809	0.0812
+ 2	23.33	2311.0	2308.4	2477.07	—	0.0796	0.0803	0.0811	0.0811	0.0812	0.0813	0.0816
+ 3	34.83	2312.1	2310.1	2481.69	—	0.0800	0.0807	0.0813	0.0815	0.0816	0.0817	0.0820
+ 4	47.30	2313.4	2312.2	—	—	0.0804	0.0813	0.0818	0.0820	0.0821	0.0821	0.0824
+ 5	72.68	2314.9	2313.7	—	—	0.0817	0.0821	0.0824	0.0825	0.0826	0.0826	0.0828
+ 6	88.05	2316.5	2315.3	—	—	0.0830	0.0830	0.0830	0.0830	0.0830	0.0830	0.0830
+ 7	280.00	2331.9	2331.5	—	—	0.0895	0.0894	0.0895	0.0898	0.0901	0.0904	0.0927
+ 8	338.30	2348.5	2349.0	—	—	0.0960	0.0966	0.0975	0.0988	0.0999	0.1010	0.1060
+ 9	379.10	2366.2	2367.7	—	—	0.1090	0.1060	0.1090	0.1120	0.1150	0.1170	0.1260
+ 10	447.10	—	2389.4	—	—	0.1070	0.1100	0.1210	0.1320	0.1390	0.1430	0.1580
+ 11	504.30	—	2404.7	—	—	0.0951	0.1080	0.1540	0.1790	0.1900	0.1970	0.2160

Conclusions. Our experiments were primarily intended to test and normalize fluorescence measurements against measurements of the entire spectrum as made by the Henway time-integrated spectrograph. In addition, we wished to make measurements of the M-band emission from a gold disk as a function of the laser intensity and angle of incidence. The two spectrographs in our work did not have equivalent lines of sight, but in the experiments at normal incidence, they will. Both the x-ray diffraction crystals and the films used in our experiments were absolutely calibrated; however, the film calibrations are not accurate enough and will be more carefully done using a molybdenum L-line source at 2295 eV. Our measurements are consistent, given the present uncertainties in angular distribution of x rays and in the film calibration.

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X-Ray Laser Experiments.

Progress in Nickel-Like Lasers. Since the demonstration of an extreme ultraviolet (XUV) amplifier at 206.3 and 209.6 Å (Refs. 202 and 203), much effort has been expended on developing an amplifier to produce significant power at wavelengths below the carbon K-absorption edge at 43.76 Å. One motivation is to produce a coherent, monochromatic, high-brightness source suitable for holography of biological specimens in the "water window" between the K-edges of carbon and oxygen.²⁰⁴

Matthews et al.²⁰² achieved gain on 3p $\rightarrow$ 3s transitions in neon-like-selenium ions produced by intense irradiation of a thin selenium foil. The explosion of the foil led to creation of a plasma that served as the amplifier medium. The dominant line expected to be amplified was the $J = 0 \rightarrow 1$ transition at 182.4 Å, predicted to be pumped by electron collisional excitation from the ground state.²⁰⁵⁻²⁰⁹ Matthews et al. did not observe large gain on the $J = 0 \rightarrow 1$ line, but, instead, saw gains on the order of 5 cm⁻¹ on $J = 2 \rightarrow 1$ transitions. Population mechanisms for the $J = 2$ levels are more complicated, including contributions by direct collisional excitation, recombination, and cascading from higher energy levels.

The neon-like-selenium result has been extrapolated to higher-Z ions. For example, Y²⁹⁺ has shown gain at 155.0 and 157.1 Å, while Mo³²⁺ has produced gain of 4 cm⁻¹ at 131.0 and 132.7 Å (Ref. 210). However, the rapid scaling²¹¹ of laser power required with increased target Z makes neon-like schemes unsuitable for extrapolation to the water window. The increase in irradiance between selenium and molybdenum was from 5×10^{13} to 4×10^{14} W·cm⁻². To obtain gain below 43.76 Å would require producing Gd⁵⁴⁺ with an estimated irradiance in excess of 10^{16} W·cm⁻² but would be impossible to apply over a long plasma with currently available laser drivers.

The theory of a nickel-like analogue to the neon-like scheme has been discussed previously²¹²⁻²¹⁴ but has lacked

Fig. 3-132. Simplified level diagram for Eu^{35+} and transitions of interest. Collision rates are for $n_e = 2 \times 10^{20} \text{ cm}^{-3}$ and $T_e = 600 \text{ eV}$. Level energies (eV) and spontaneous-emission rates (s^{-1}) are in parentheses.

experimental validation. The population inversion necessary for amplification is produced between the 4d and 4p levels, which lie above a $3d^{10}$ closed shell.

Figure 3-132 is a simplified level diagram for Eu^{35+} showing the four 4d — 4p transitions most likely to be amplified. The dominant population mechanism for the $3d_{3/2}^9 4d_{3/2} 0$ level is electron collisional excitation from the ground state. The collision rate is shown in Fig. 3-132

for an electron density of $2 \times 10^{20} \text{ cm}^{-3}$ and an electron temperature of 600 eV. This rate is calculated using the multi-configuration, relativistic distorted-wave (MCDW) code.²¹⁴ The $J = 2$ and $J = 1$, 4d levels are fed by collisions from the ground state but are also populated by recombination, cascading from higher- n states, and feeding from the 4f levels. The 4d — 4p population inversion is maintained by the fast 4p — 3d radiative decay, and reduction of this decay rate by trapping will reduce the inversion.

A 137-level, time-dependent model for zinc-, copper-, nickel-, and cobalt-like gadolinium has been incorporated in the XRASER kinetics code,²¹⁵ and the 2-D hydrodynamics output from LASNEX²¹⁶ used as a basis to predict expected gains on the four Eu^{35+} transitions shown in Fig. 3-132. A gadolinium atomic-data file was used to model europium ($Z = 63$) because early, unsuccessful attempts to fabricate thin-foil targets had centered on gadolinium ($Z = 64$). The model included the effect of trapping of the 4p — 3d transitions.

Two types of target foils were used: 50- and 90- $\mu\text{g} \cdot \text{cm}^{-2}$ EuF_2 on 10- $\mu\text{g} \cdot \text{cm}^{-2}$ CH. The targets were irradiated with a total of $7 \times 10^{13} \text{ W} \cdot \text{cm}^{-2}$ in two opposing line foci, 200- μm wide, with a pulse of 1-ns duration. Simulations²¹⁷ of the experiments showed expected gains for both targets on the order of unity on the

Fig. 3-133. Experimental setup at Nova two-beam facility.

four nickel-like lines, with the 65.83- and 71.00-Å, $J = 0 \rightarrow 1$ lines predicted to have gains of 0.8 and 1.0 cm^{-1} , respectively. The gain was calculated to appear near the peak of the heating pulse when the electron temperature and density were expected to be 600 eV and $2 \times 10^{20} \text{ cm}^{-3}$, respectively.

As shown in Fig. 3-133, two beams of the Nova laser were used for the experiment. The principal diagnostics were grazing-incidence grating spectrometers, with microchannel-plate detectors capable of being gated in time. Two spectrometers positioned so as to view the target along the axis of the line focus (the preferred direction for amplification), and a third spectrometer viewed the target from 45° off-axis. Signals from the spectrographs were time-integrated up to 500 ps after the peak of the 0.53-μm Nova pulse, then they were gated off.

Figure 3-134 shows spectra from a $50\text{-}\mu\text{g} \cdot \text{cm}^{-2} \text{ EuF}_2$ target. The upper trace is an off-axis spectrum showing the bright, optically thick emission from $4f \rightarrow 4f'$ transitions in copper- and zinc-like europium, which have been identified previously.²¹⁸⁻²²⁰ The lower trace is an on-axis spectrum with zinc- and copper-like emission, but also shows bright emission at 65.83, 71.00, 100.39, and 104.56 Å, identified as previously unobserved $4d \rightarrow 4p$ transitions in nickel-like europium. Line identifications are based on their proximity to calculated values and their behavior as the length of the target is increased.

Table 3-21 shows predicted wavelengths²²¹ for the lines. Disagreement with measured wavelengths of the two shorter-wavelength lines arises from difficulty in calculating the energy of the $J = 0$ state due to contributions from configuration interaction. As the two

lines share the same upper state, the disagreement with the measured transition energies is similar (0.32 and 0.49 eV). A third, weaker $J = 0 \rightarrow 1$ transition from the same level (observed at 84.83 Å on the longest targets) has a similar energy deviation.

Figure 3-135 shows sections of typical on-axis spectra from targets of different lengths. The $J = 0 \rightarrow 1$ lines show a nonlinear increase in intensity as target length is increased, with the 71.00-Å line increasing more rapidly than the 65.83-Å line. Spectra were scanned by a digitizing densitometer, and the resultant film-density values were converted to intensity using a calibration wedge developed with the data. The intensity of an individual line was then measured by subtracting the background continuum level and integrating over the line profile.

Fig. 3-134. Observed europium $4f \rightarrow 4f'$ off-axis and on-axis spectra.

Table 3-21. Calculated and observed wavelength values (Å) for nickel-like $4d \rightarrow 4p$ transitions. Ytterbium and tungsten $J = 0 \rightarrow 1$ calculations are adjusted for observed europium wavelengths.

Transition	Eu ³⁵⁺		Yb ⁴²⁺		W ⁴⁶⁺
	Theory	Observed	Extrapolated theory	Observed	Extrapolated theory
$3d_{3/2}^2 4d_{3/2} 2j_0 \rightarrow 3d_{3/2}^2 4p_{1/2} 2j_1$	65.72	65.83 ± 0.03	50.24	50.26 ± 0.05	43.15
$3d_{3/2}^2 4d_{3/2} 2j_0 \rightarrow 3d_{3/2}^2 4p_{3/2} 2j_1$	70.80	71.00 ± 0.03	56.10	56.09 ± 0.05	49.34
$3d_{3/2}^2 4d_{3/2} 2j_0 \rightarrow 3d_{3/2}^2 4p_{3/2} 2j_1$	84.62	84.83 ± 0.05	72.04	—	66.39
$3d_{3/2}^2 4d_{5/2} 2j_2 \rightarrow 3d_{3/2}^2 4p_{3/2} 2j_1$	100.32	100.39 ± 0.03	81.03	—	72.27
$3d_{3/2}^2 4d_{5/2} 2j_1 \rightarrow 3d_{3/2}^2 4p_{3/2} 2j_1$	104.56	104.56 ± 0.05	84.44	84.40 ± 0.05	75.29

Fig. 3-135. On-axis spectra in the region near the $J = 0 \rightarrow 1$ lines of Eu^{35+} for three target lengths.

The results of these measurements are shown in Fig. 3-136(a) for the two $J = 0 \rightarrow 1$ lines and for the $4f^2F_{5/2} \rightarrow 4d^2D_{3/2}$ copper-like transition at 93.84 Å. Data shown are for the $50\text{-}\mu\text{g}\cdot\text{cm}^{-2}$ EuF_2 targets. The $J = 0 \rightarrow 1$ data were fit to the formula

$$I \simeq (e^{\alpha l} - 1)^{3/2} (\alpha l e^{\alpha l})^{-1/2} \quad (55)$$

for the output power of a Doppler-broadened, homogeneous, time-independent, distributed source of ASE²²² of gain coefficient α and length l . The 71.00- and 65.83-Å data were fit to gains of 1.11 ± 0.12 and $0.61 \pm 0.14 \text{ cm}^{-1}$, respectively. The relative sizes of the gains are qualitatively consistent with the relative oscillator strengths of the two lines, calculated from the relativistic configuration-interaction code YODA.²²³ Uncertainties in the gains (68% confidence) are calculated assuming an intrinsic error of 15 to 20% in the intensity values. The expected plasma conditions would lead to an absorption coefficient on the order of 300 cm^{-1} for the $4f^2F_{5/2} \rightarrow 4d^2D_{3/2}$ copper-like line; the flat scaling of this line with target length is consistent with it being optically thick.

Figure 3-136(b) shows the length behavior of the $J = 2 \rightarrow 1$ and $J = 1 \rightarrow 1$ lines. Fits to the small αl limit of Eq. (55) give $\alpha = 0.08 \pm 0.14 \text{ cm}^{-1}$ for the $J = 2 \rightarrow 1$ and $\alpha = -0.07 \pm 0.19 \text{ cm}^{-1}$ for the $J = 1 \rightarrow 1$, and they are essentially linear. Simulated gains were much higher (1.6 and 1.0 cm^{-1} , respectively). For the lines to be seen above continuum level, there must be significant emissivity. In the calculation, the excited-state population required to give the necessary emissivity also gave high gain. However, a larger-than-expected population in the 4p lower level, which could be produced by additional trapping (which, in turn, already has a large effect on the $J = 2 \rightarrow 1$ and $J = 1 \rightarrow 1$ simulated gains²¹⁷) would destroy the gain while leaving large emissivity. Such an increase would affect $J = 2 \rightarrow 1$ and $J = 1 \rightarrow 1$ gains more than $J = 0 \rightarrow 1$ gains because of weighting of the lower-level population by the ratio of statistical weights in the expression for population inversion. The gains could also be lower than calculated because of the difficulty in accurately modeling the multitude of $J = 2$ and $J = 1$ population mechanisms, or because of

Fig. 3-136. Intensity vs length for five lines and the continuum at 66 Å from on-axis spectra. Nickel-like line intensities are fit to Eq. (55). The line through 93.84-Å data is not a mathematical fit; the continuum is fit to a straight line.

more refraction by plasma-density gradients than is already included in the simulation.²¹⁷

The optical depth of bremsstrahlung-continuum emission at 66 Å is calculated to be less than 0.2 for the longest target. The behavior of the continuum is shown in Fig. 3-136(b) to be linear, consistent with its being dominated by optically thin emission. The observation of the expected behavior of the continuum and the optically thick copper-like line at 93.84 Å verify that the spectrometers provided a reliable measurement.

Data for the $90\text{-}\mu\text{g}\cdot\text{cm}^{-2}$ EuF_2 targets, analyzed in a similar manner, resulted in gains of 0.60 ± 0.09 , 0.25 ± 0.17 , -0.13 ± 0.14 , and $-0.38 \pm 0.16 \text{ cm}^{-1}$ for the 71.00-, 65.83-, 100.39-, and 104.56-Å lines, respectively. Once again, the $J = 2 \rightarrow 1$ and $J = 1 \rightarrow 1$ gains disagree with calculation, and $J = 0 \rightarrow 1$ gains are now reduced. Thicker targets would be expected to be hotter, denser, more ionized,²¹¹ and also to suffer more from trapping of the $4p \rightarrow 3d$ transition. All of these changes would affect gain coefficients.

The extrapolation of the nickel-like scheme to shorter wavelength was investigated by studying ytterbium. The targets were $100\text{-}\mu\text{g}\cdot\text{cm}^{-2}$ ytterbium on $10\text{-}\mu\text{g}\cdot\text{cm}^{-2}$ CH irradiated at $1.4 \times 10^{14} \text{ W}\cdot\text{cm}^{-2}$ in a 1-ns pulse.²²⁴ Line emissions at 50.26 and 56.09 Å were identified as the $J = 0 \rightarrow 1$ transitions in Yb^{42+} .

The line at 50.26 Å increases nonlinearly with target length, consistent with its being amplified with a gain on the order of 1 cm^{-1} . The length behavior of the 56.09-Å line is closer to linear. Oscillator strengths are calculated to be 0.075 and 0.061 for the 50.26- and 56.09-Å lines, respectively²²³; thus, the shorter-wavelength line would be expected to have slightly more gain. Table 3-21 shows observed and calculated wavelengths of all the nickel-like lines seen to date. The calculated wavelengths for tungsten are also given, demonstrating scaling of the shorter-wavelength, $J = 0 \rightarrow 1$ line to below the carbon K-edge.

In conclusion, we have demonstrated gain in a laser-heated, exploding-foil plasma of nickel-like ions. Our demonstration now opens the way for extrapolation of present-day, relatively long-wavelength XUV lasers^{202,210} to a domain of truly soft x rays. Scaling of the scheme to shorter wavelength is promising, as indicated by our results with ytterbium. We predict that an exploding-foil amplifier of tungsten will produce gain at 43.15 Å, with heating-pulse intensities of approximately $4 \times 10^{14} \text{ W}\cdot\text{cm}^{-2}$.

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Recombination X-Ray-Laser Experiments in Hydrogen-, Helium-, and Lithium-Like Ions. As discussed in the 1986 Laser Annual, one important near-term goal of our x-ray-laser research is the demonstration of soft x-ray amplification at wavelengths near the carbon K-edge at 43.7 Å. In the past year, we pursued nickel-like and recombination schemes as a means of demonstrating short-wavelength x-ray lasers. We described our recent results for nickel-like ions in the previous article. Here, we present data and conclusions from recombination x-ray-laser experiments. Before turning to a summary of recent results from recombination experiments, we briefly discuss our motivation for work in this area based on the relative efficiency of different x-ray-laser schemes.

Figure 3-137 is a plot of the peak electron temperature T_e required vs x-ray-laser wavelength for various short-wavelength laser schemes. Several of the

points are labeled with the elements appropriate for a given wavelength: at 206.4 and 209.8 Å the neon-like laser element used is selenium, at 50.3 Å it is nickel-like ytterbium, and so forth. (For clarity, the curve for hydrogen-like 3 → 2 recombination systems is not shown; it lies between the nickel- and lithium-like 4f → 3d curves.) The peak electron temperature required is plotted in that it represents a rough measure of the relative efficiency of a system; systems heated to a lower peak temperature require, in principle, less power than those heated to a higher temperature. In this figure, we assumed steady-state ionization balance and an electron density of 10^{21} cm^{-3} . For each of the four schemes shown, the temperature has been chosen to produce plasmas composed of (in roughly equal amounts) the lasing species and the next-higher ionization stage.

A more detailed analysis of the efficiency of various x-ray laser (XRL) schemes requires clarification of the relationships among peak T_e and parameters, such as target thickness and incident laser intensity. Figure 3-137 serves as a general guide, however, to show that recombination schemes (especially in lithium-like ions) hold the promise of greater efficiency than both neon- and nickel-like collisional schemes, and it is for this reason that we are investigating these systems. (The detailed scaling of neon- and nickel-like-ion x-ray lasers is discussed in Refs. 225 and 226, respectively.)

During our research on recombination schemes at LLNL in the last year, we further investigated the aluminum systems described in the 1986 Laser Annual. In addition, we performed preliminary investigations on recombination schemes in higher-Z lithium-like ions, such as Cr^{21+} and Ca^{17+} . We first summarize data relevant to hydrogen-like aluminum systems, and we then turn to a discussion of higher-Z lithium-like schemes.

Aluminum Results. Our experiments on aluminum recombination schemes in 1987 focused on obtaining further data regarding the plasma ionization balance and possible presence of gain on various transitions. As discussed in the 1986 Laser Annual, the candidate transitions for gain are the 5f → 3d transitions in

Fig. 3-137. Peak electron temperature required vs x-ray-laser wavelength for various short-wavelength laser schemes.

Fig. 3-138. Experimental setup for the Nova two-beam chamber showing an x-ray laser target and associated diagnostics.

Al XI (lithium-like) and Al XII (helium-like) at 105.7 and 88.9 Å, respectively, and the $4f \rightarrow 3d$ transition in Al XII at 130.1 Å. (Gain on the 105.7-Å, lithium-like line has been observed elsewhere.²²⁷)

Our targets in these experiments consisted of 100- μm -wide aluminum strips ranging in length and thickness from 0.5 to 2 cm and 1000 to 4000 Å, respectively. Further description of the targets is presented in last year's Laser Annual. We observed the plasma by several McPIGS soft x-ray spectrographs looking both on and off the x-ray-laser axis and by time-resolved and time-integrated x-ray spectrometers. The experimental setup for the experiments described in this article is shown in Fig. 3-138.

The additional diagnostic developed in 1987 and shown in Fig. 3-138 was a McPIGS spectrograph capable of spatial resolution in the horizontal plane (the plane of the incident laser). The spectrometer uses a cylindrical mirror to image the source in the horizontal direction onto the detection plane of the instrument. The resulting 2-D film image contains spectral information in one dimension and spatial information in the other. Spatial resolution of the instrument in the target plane is 20 μm , and the microchannel-plate detector was gated off 2 ns after the laser pulse.

Figure 3-139 shows sample data from the McPIGS and plots the measured

spatial extent of various hydrogen-, helium-, and lithium-like aluminum lines. On this shot, a 1000-Å-thick aluminum-strip target was irradiated with a 100-ps FWHM pulse at an intensity of $1.3 \times 10^{14} \text{ W/cm}^2$. The spatial extent of hydrogen-like emission is 100 μm . This result is consistent with hydrogen-like emission occurring over a period of 200 to 300 ps (during and just after the driving laser pulse). Thus, the bulk of hydrogen-like emission occurs when plasma is hottest, consistent with the idea that this emission is, in large part, due to collisional excitation.

With regard to lower stages of ionization, the 88.9-Å helium-like and the 105.7-Å lines have spatial extents of 270 μm and 1 mm, respectively. The larger spatial extent of the helium- and lithium-like emission implies that these transitions emit well after the end of the laser pulse; thus, a significant fraction of the total emission in these lines occurs as a result of recombination. Taken as a whole, these data support the idea that hydrogen-like emission arises primarily due to excitation, while helium- and lithium-like radiation is due, in large part, to recombination. One caveat to our data is that the relative timing of the two incident laser beams on this shot was displaced by 200 ps. However, the basic interpretation of the data remains unchanged, especially for the helium- and lithium-like lines, because their spatial

Fig. 3-139. Sample data from McPIGS spectrograph showing measured spatial extent of hydrogen-, helium-, and lithium-like aluminum lines.

extents are considerably greater than the product of expansion velocity and total laser-pulse duration.

These data and x-ray spectra presented in last year's Laser Annual support the notion that plasma is ionizing to primarily the hydrogen-like stage, as opposed to the fully stripped state necessary to produce population inversion in hydrogen-like ions. This notion suggested the possible presence of gain in helium- and lithium-like aluminum. To check for such a possibility, we carried out a length study at an on-target intensity of 2×10^{14} W/cm². An initial measurement using 0.5-, 1- and 2-cm-long targets showed evidence for nonlinear growth with increasing target length for the 88.9-, 130.1-, and 105.7-Å transitions. We are currently engaged in efforts to determine whether or not there is gain on these lines.

Higher-Z Lithium-Like Results. We also carried out a preliminary series of experiments to investigate the spectroscopy of lithium-like ions of calcium and chromium. We chose these elements because they each possess candidate lasing lines below 44 Å: the 4f → 3d transition in lithium-like chromium at 38.8 Å and the 5f → 3d transition in lithium-like calcium at 39.5 Å.

The target geometry was the same as that for our aluminum experiments; the CaF₂ strip thickness in this case was

3000 Å. The 39.5-Å, 5f → 3d and the 57.7-Å, 4f → 3d transitions in lithium-like calcium were observed, and the 4d → 3p and 4p → 3s transitions at 56.7 and 54.4 Å, respectively, were also identified. (Calcium wavelengths quoted here are theoretical values.)

We also used the CaF₂ targets in an attempt to produce measurable gain on the 3 → 2 transition in hydrogen-like fluorine at 80.8 Å. In the first series of experiments, we observed no nonlinear growth on this or any of the above calcium transitions. We also carried out several shots in chromium, which resulted in identification of the 4f → 3d line at 38.8 Å, but, once again, without evidence for amplification. From the observed spectra, it appears that the laser intensity we used may have been too low to produce the helium-like state necessary for inversion on these 4f → 3d and 5f → 3d transitions.

Our future work in these systems will focus on ionization-balance measurements to determine the conditions appropriate for production of helium-like ions necessary to pump population inversions in the lithium-like ionization stage. The physics experiments will be done on smaller facilities at higher shot rates and will result in a set of laser parameters necessary to provide appropriate ionization-balance conditions (i.e., sufficient amounts of helium-like ions) for gain in lithium-like ions. In addition, we

are working towards the goal of conducting recombination experiments on Nova using 20-ps laser pulses, which should result in higher electron density (and thus higher gain) during the plasma recombination phase.

Conclusions. Our recombination x-ray-laser experimental efforts during 1987 focused on the aluminum systems first discussed in the 1986 Laser Annual and on preliminary exploration of higher- Z lithium-like systems. With respect to aluminum schemes, soft x-ray measurements of aluminum-strip plasmas corroborated our conclusion that plasma ionization peaks out at the hydrogen-like stage. Spatially resolved measurements of soft x-ray emission were especially useful in this regard. Efforts to measure gain in helium- and lithium-like aluminum resulted in evidence for nonlinear growth of the 88.9- and 130.1-Å lines in helium-like aluminum and the 105.7-Å line in lithium-like aluminum.

Our preliminary experiments with lithium-like chromium and lithium-like calcium resulted in identification of candidate lasing transitions, but we observed no amplification, and the targets were not optimized. Our future work in these lithium-like systems will involve ionization-balance measurements to determine laser and target conditions that are optimum for producing gain. In particular, we will examine plasmas created by short (20-ps) driving laser pulses. With such results in hand, we will clearly define a parameter space for Nova experiments. The systems we have identified have the potential for high efficiency, and the information gained from measurements on aluminum as well as forthcoming ionization-balance data should help point the way to higher-gain recombination schemes.

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Multipass Amplification of Soft X Rays in a Laser Cavity. We have been working since 1985 on the development of

resonant cavities for use with our recently developed soft x-ray sources of amplified spontaneous emission (ASE).²²⁸⁻²³² Cavity development is a crucial step toward the ultimate transformation of these x-ray gain media into relatively efficient soft x-ray lasers with optimized divergence and coherence properties.

The fundamental challenges in laser cavity development have been optics survival, radiation transport, and gain lifetime.²³⁰⁻²³² Typical soft x-ray gain media are short-duration, high-temperature, high-density plasmas produced by intense optical laser irradiation of foil, fiber, or solid targets. These gain media provide a hostile x-ray environment for cavity optics. They may also be subject to local inhomogeneities that could compromise coherent transport of amplified radiation, and the short gain lifetime (typically a few hundred picoseconds) leaves little opportunity for many amplification passes.

Significant progress has been made in addressing these challenges. There are reports in the technical literature on the development of relatively efficient and robust multilayer cavity optics.²³²⁻²³⁴ In addition, double-pass cavity experiments have been performed that demonstrate the survival of cavity optics and the transport of amplified radiation.^{232,235,236} However, until our work in 1987, no significant double-pass amplification had been achieved (i.e., the double-pass output was not much greater than the single-pass ASE), nor had multipass amplification of soft x rays been accomplished.

We have made a significant advance in the development of soft x-ray laser cavities. In a series of XRL cavity experiments conducted on the Nova laser facility, we achieved a double-pass output that was approximately an order of magnitude more intense than the single-pass ASE, and produced triple-pass amplified output from a multipass XRL cavity as well. Double- and triple-pass amplification were achieved on the neon-like selenium lines at 20.63 and 20.96 nm, and cavity output measurements were time-resolved, allowing temporal separation and distinct identification of the single-, double-, and triple-pass beams. Multipass

amplification in these experiments was limited to only three passes by the finite duration of the gain.

The experiments were conducted at the two-beam target facility of the Nova laser system and had a typical pulse duration of 500 ps (FWHM). Each XRL target used in these experiments consisted of a 2-cm-long, 80-nm-thick strip of selenium supported on a 100-nm Lexan foil. Despite the 2.0-cm length of the XRL target, however, gain length was only 1.8 cm because of a 0.2-cm-diam hole at the center of the target—to match a similar “hole” in the line focus of the optical laser. The 2 kJ (total) each target received was delivered in a 2.3-cm-long, 0.03-cm-diam line focus whose length ensured uniform illumination of the entire 2-cm-long target. Such laser-illuminated selenium targets exhibit significant amplification of $2p^53p \rightarrow 2p^53s$ transitions in neon-like selenium. The dominant emissions are the $J = 2 \rightarrow 1$ lines at 20.63 nm and 20.96 nm.^{228,229,237,238}

The XRL cavity consisted of a spherical multilayer mirror and a flat multilayer beam splitter placed on either side of and orthogonal to the axis of the gain medium, whose ends were typically a few centimeters away from the optical components. (The beamsplitter was removed for double-pass cavity experiments.) The spherical mirror was a molybdenum-silicon multilayer deposited on a highly polished BK-7 substrate. Its multilayer

coating was comprised of 30 Mo-Si layer pairs with a nominal period of 11.1 nm and an Mo-Si ratio of 2:3. The beam splitter was an Mo-Si multilayer supported by a 40-nm-thick silicon-nitride substrate. Its multilayer coating was comprised of 6 Mo-Si layer pairs with a nominal period of 11.4 nm and an Mo-Si ratio of 1:1. When the performance of the x-ray optical components was tested at near normal incidence (0.5 degrees off normal) at 20.63 nm and 20.96 nm, spherical mirror reflectivity was 20% and beam splitter reflectivity and transmission were 15 and 5%, respectively.

We measured XRL cavity output with a time-resolving x-ray spectrometer.^{239,240} This spectrometer had a temporal resolution of 20 ps, a spectral resolution of 0.1 nm, and a radiation acceptance angle of 1.4×10^{-4} sr.

Figure 3-140 is a representative data record from a double-pass cavity experiment; it shows the sum of signal intensities at 20.63 and 20.96 nm plotted vs time. (The signals at 20.63 and 20.96 nm were virtually identical.) A total of 2.05 kJ was delivered to the target in this experiment, and the cavity consisted of a flat mirror placed 2.75 cm from the end of the gain medium.

The single-pass ASE and double-pass amplified signal are easily identified in Fig. 3-140. Their peak-to-peak separation of approximately 0.275 ns is within 10% of the expected value for the 7.5-cm path difference for single-pass and double-pass beams. The double-pass signal is seven times more intense than the single-pass ASE, indicating a second-pass amplification factor of approximately 800 when reflection and divergence losses are taken into account.

Figure 3-141 is a representative data record from a multipass cavity experiment; it shows the time-dependent signal intensity at 20.63 nm (the 20.96-nm signal is virtually identical), along with an illustration of the multipass cavity geometry. A total of 2.3 kJ was delivered to the target in this experiment, and the cavity consisted of a spherical mirror ($R = 12$ cm) placed 3.5 cm from one end of the gain medium and of a flat beam splitter placed 3.0 cm from the opposing end.

Fig. 3-140. Time-resolved double-pass cavity output at 20.63 and 20.96 nm (summed together) for experiment No. 17071004.

The single-, double-, and triple-pass amplified signals are easily identified in Fig. 3-141. The temporal separation of the three signals is again within 10% of that expected from their optical path differences. The double-pass signal is 22 times more intense than and the triple-pass signal approximately equal to the single-pass ASE. (The low-intensity triple-pass signal may be attributed to the rapid fall-off of the time-dependent gain.)

The data in Figs. 3-140 and 3-141 demonstrate that cavity optics can significantly enhance the output intensity from soft x-ray gain media. The data are consistent with cavity optics survival and efficient radiation transport during the multipass amplification process. Indeed, the observed amplification pattern (i.e., strong double-pass amplification, weaker triple-pass amplification) appears attributable to the short duration of the time-dependent gain, while the gain lifetime appears to be approximately equal to the duration of the driving optical laser pulse. In interpreting our double- and triple-pass data, however, we cannot totally rule out the possibility of cavity losses due to slight mirror misalignment, late-time mirror damage, or transport inefficiency resulting from plasma gradients. While we have no evidence of the importance of any of these mechanisms in our experiments, short gain lifetime is clearly a limiting factor in the multipass amplification process.

If gain lifetime can be increased without compromising the quality of radiation transport or the survival of cavity optics, the prospects for soft x-ray lasers with optimized efficiency, divergence and coherence properties will be promising. These goals are being actively pursued.

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Demonstration of X-Ray Holography with an X-Ray Laser. X-ray holography offers the potential for obtaining high-resolution, three-dimensional images of *in vitro* biological microstructures.

Fig. 3-141. Time-resolved multipass cavity output at 20.63 nm for experiment No. 17071705. Total cavity length 8.5 cm. Reduced amplification for three or more passes was due to the short gain lifetime.

Significant progress toward this goal has been achieved with holography systems using synchrotron x-ray sources,²⁴¹ and spatial resolutions as small as 40 nm have been recently demonstrated.²⁴² These experiments required x-ray exposures of one hour or longer, which makes high spatial resolution difficult to achieve in live biological specimens because of blurring of the image. The blurring, caused by specimen motion, prohibits the imaging of dynamic processes within the specimen.

A possible solution to the problem of blurring is to exploit the extremely high brightness and long coherence lengths produced by x-ray lasers and to create the hologram with exposure times of less than 1 ns. This article presents results from an experiment in which an x-ray laser was used to produce x-ray holograms.

Our holography used Gabor²⁴³ in-line geometry modified by including a high-reflectivity, multilayer x-ray mirror²⁴⁴ as a narrow-bandpass filter. The x-ray mirror had a flatness and roughness of less than $\lambda/100$ (where λ is the wavelength) at the x-ray laser wavelengths.

The soft x-ray laser in our experiments was the neon-like selenium laser (ionized 24 times) produced at the Nova laser two-beam facility.²⁴⁵ This continuum comprised the dominant noise source in the hologram experiments. The longitudinal coherence length of the laser emission is estimated to be greater than 100 μm based on the laser line width.

Fig. 3-142. Schematic arrangement of Gabor in-line holography geometry modified by inclusion of a narrow-bandpass, multilayer x-ray mirror.

assuming a Doppler-broadened line profile with an ion temperature of 400 eV (Ref. 246). The lower bound on the transverse coherence length is $3.5\text{ }\mu\text{m}$ at the end of the x-ray laser amplifier. This bound is based on the measured source size of $70\text{ }\mu\text{m}$ for a 4-cm-long laser amplifier. The effect of coherence brightening²⁴⁷ due to the high gain-length product of the laser ($gL \approx 16$) was ignored so that these coherence lengths are lower bounds. The transverse profile of the x-ray laser beam has a double-peaked structure with most of the energy in one of the peaks. This structure is due to refraction effects in the laser amplifier.²⁴⁸ The inner portion of the higher intensity peak was used to produce the holograms.

The geometry used for the holography was a modified version of the Gabor in-line geometry, as shown in Fig. 3-142. In this geometry the x-ray beam propagates in vacuum to the multilayer x-ray mirror where it is reflected at 60° to the object and the detector. The narrow bandpass (10% at 20.8 nm) of the mirror reduces the broadband continuum energy to less than 10% of the x-ray laser energy recorded at the detector. Without this reduction, the continuum emission would significantly reduce the fringe visibility in the hologram. In this geometry, forward-scattered x rays from the object become the object beam for producing the hologram. X rays that miss the object become the reference beam. Both beams interfere at the detector producing the hologram.

The x-ray mirror consisted of a low-scatter, highly polished, fused-silica substrate (General Optics, Moorpark, Calif.) and a multilayer coating of silicon and molybdenum. The measured roughness of the finished mirror was 0.07 nm rms,

and the flatness over $100\text{ }\mu\text{m}$ was $\lambda/100$ at 20 nm (Ref. 249). A typical scalength for the holograms produced was $100\text{ }\mu\text{m}$. A flatness of at least $\lambda/10$ at 20 nm (x-ray laser quality) was required to avoid destroying the coherence of the x-ray laser beam. The multilayer coating consisted of 20 alternating 9.8-nm layers of silicon and 3.0-nm layers of molybdenum. The coating produced a measured x-ray reflectivity of 25% for 20.8-nm light at a 60° grazing angle. The coatings were designed using the LLNL computer code MLPLUS²⁵⁰ and were coated at the Lawrence Berkeley Laboratory Center for X-ray Optics, where the x-ray reflectivity was also measured.

The detector was Kodak 101 x-ray film, which has a sensitivity at this wavelength range of approximately $1\text{ photon}/\mu\text{m}^2$ and a grain size of $1\text{ }\mu\text{m}$.^{251,252} The spatial resolution is about 5 to $10\text{ }\mu\text{m}$. The film was protected from stray visible and ultraviolet light by a $2.3\text{-}\mu\text{m}$ -thick, free-standing, 1-cm-diam aluminum filter manufactured by Luxel Corporation (Friday Harbor, Washington). This filter had a measured surface roughness of 0.6 nm rms to ensure that filter nonuniformities did not destroy the interference produced by the coherent x rays on the film.

The distance from the x-ray laser to the object was 3.65 m, and the distance from the object to the film was 5.08 cm. At this distance, spatial coherence of the x-ray laser, which scales with distance according to the Van Cittert-Zernike theorem,²⁵³ was approximately $300\text{ }\mu\text{m}$. This spatial coherence allowed us to produce holograms of up to 1 mm in extent. The spatial resolution of the holography system, estimated from the resolution limits for Gabor holography,²⁵⁴ was approximately $5\text{ }\mu\text{m}$, with the dominant limitation being the film spatial resolution. The 1-cm-diam aperture of the filter permitted us to produce several holograms in one laser shot. By using a portion of the x-ray beam, where the intensity as a function of position in the beam was rapidly varying, we could obtain a variety of exposures on a single laser shot.

We used two types of objects in our experiments. The first type was an $8\text{-}\mu\text{m}$ -diam carbon fiber; up to five fibers were employed on each laser shot. The carbon

Fig. 3-143. X-ray hologram of an 8- μ m-diam carbon wire.

fibers were used to generate holograms with predictable interference fringe patterns. The dimensions of the holography system were chosen to optimize the visibility of these fringe patterns and to ensure that the interference-fringe spatial frequency did not exceed the spatial resolution of the x-ray film. The second type of object was a 3-D structure consisting of three, 10- μ m gold bars in an orthonormal configuration at the tip of a 40- μ m glass stalk. This figure allowed us to produce more complicated 3-D holograms. Both types of objects were opaque at the x-ray laser wavelengths.

Figure 3-143(a) shows the fringe pattern obtained from a carbon fiber. Four to five fringes can be observed. This figure was obtained by digitizing the data with a 30-to-1 aspect ratio rectangular slit and converting the film density to linear exposure (photons per unit area). The density-exposure curve for the conversion was obtained from Ref. 250.

Figure 3-143(b) shows the calculated fringe pattern for an opaque strip. The calculated fringe pattern was obtained with the analytic results derived in a Fraunhofer diffraction calculation including the effects of a partially coherent source.²⁵⁵ A 70- μ m-diam, uniformly circular source and plane-wave illumination were assumed. The correction for the curvature of the wave front is negligible. The normalized theory from Ref. 254 was scaled to agree with the experimentally measured peak amplitudes.

Agreement between theory and experiment for the wire object is quite good.

The fringe peak positions and relative amplitude variations as a function of position agree very well. Only at large transverse distances on the order of 200 μ m do theory and experiment appear to disagree. Here, the spatial frequency of the predicted fringes is too large for the film spatial response; consequently the fringes are not resolved.

Holograms from the wire and 3-D stick figure were successfully reconstructed using a helium-neon laser. Both images were visible above a large background noise in holograms.

Our experiments show that current x-ray lasers are sufficiently well characterized and have enough brightness to be used successfully in the creation of x-ray holograms. In addition, x-ray mirrors now have the necessary flatness, roughness, and reflectivities to allow their use in phase-sensitive x-ray experiments. The experiments also show that by using x-ray lasers, holograms can be created with exposure times of a few hundred picoseconds.

The trend in x-ray laser development is toward ever-decreasing wavelengths and improved coherence. Lasing has been observed as low as 6.6 nm in nickel-like europium and 5.03 nm in nickel-like yttrium (see the article, "Progress in Nickel-Like Lasers," earlier in this section). This scheme should be scalable to sub-4.4-nm wavelengths in nickel-like tungsten (see the article, "Analysis of Nickel-Like X-Ray Lasers with Wavelengths Approaching the Water Window," in the previous section). There

have been proposals for significantly improving the spatial coherence for these types of x-ray lasers.²⁵⁶ X-ray laser cavities with partially transmitting x-ray beam splitters as output couplers have also been demonstrated (see the preceding article). This development offers the possibility of both improved efficiency and coherence. With laser wavelengths in the water window (4.4 to 2.3 nm) where there is high contrast between protein and water, x-ray holograms of living cells can be produced.

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Photoionization of Argon with the Selenium Soft X-Ray Laser. With the recent development of soft x-ray lasers, many applications requiring bright, short-wavelength radiation are now possible. In a joint experiment with AT&T Bell Laboratories, we used the LLNL selenium soft x-ray laser to photoionize argon gas. Our immediate objective was to determine the absolute output of the x-ray laser by measuring resulting emission of the Ar II transition at 476.5 nm. We ultimately used the experiment to investigate the possibility of obtaining a soft x-ray laser by photoionizing sodium gas.

Figure 3-144 shows our experimental apparatus. Our x-ray source was the LLNL selenium soft x-ray laser, which emits approximately 120 μJ of 206- and 209-Å radiation from a 4-cm-long selenium foil.²⁵⁷ Two, state-of-the-art, 75-mm-diam multilayered mirrors first collimated and then focused the beam through a 400-Å-thick baron-nitride window into a cell filled with 20-Torr of argon gas. We optimized the two-mirror focusing configuration with a computer ray-tracing code, which predicted that a 50- μm -diam source would grow only to 56 μm at the focal plane. A 100-Å aluminum filter was placed between the final mirror and gas cell to attenuate stray 0.53- μm light scattered from the x-ray-laser target. Resulting emission from the argon transition exited a quartz window at the rear of the cell. A lens collected this light and focused it onto a grating spectrometer and a photomultiplier equipped with an oscilloscope.

We conducted preliminary pointing and focusing tests to ensure that sufficient x-ray-laser radiation would enter the 1-mm-diam gas-cell window located 4.6 m from the x-ray-laser source. In our preliminary tests, we placed a film pack (film, aluminum filters, and cross hairs) at the location of the gas-cell window, as shown in Fig. 3-145. We achieved alignment of the film-pack cross hairs, multilayer-mirror axes, and x-ray laser by means of a telescope and external mirror manipulators.

Fig. 3-144. Two multilayered mirrors point and focus the x-ray laser into a gas cell containing argon gas. After photoionization, resulting emission at 476.5 Å is recorded with a photomultiplier.

Results from firing both 3- and 4-cm x-ray-laser targets into the film pack showed that a 235- μm -diam spot size (at 5 mm from best focus) was attainable at 5 mm from theoretical best focus. The focal spot missed the film-pack cross hairs by 300 μm , which translates to a 75- μrad -pointing accuracy. Although we recorded no data at the theoretical best focus, results at the defocused positions were consistent with achieving spot sizes of less than 100 μm . By post-calibrating the mirror and filters, we found that the focusing system could provide 1 to 2 μJ to the gas-cell window, which gave us high confidence that we could make the argon photoionization scheme work.

For our photoionization experiment, we replaced the film pack with the gas cell. We then fired a 4-cm-long x-ray-laser target, which should have put 0.5 to 1 μJ into the cell.²⁷ Output of the photomultiplier indicated that 0.5 μJ of x-ray-laser energy entered the cell. This value represents good agreement with expectation, considering the large uncertainty ($\sim 100\%$) in the film calibration used in our preliminary tests described above. Nonetheless, in obtaining 0.36 μW of 476.5-nm light, we concluded that the gain-length product in argon at 476.5 nm was ~ 2.2 .

In an attempt to provide more output energy from the x-ray laser, we tested a 5-cm-long x-ray-laser amplifier. This target should have placed over 4 μJ of nominal energy into the cell; however, the gas cell recorded only 0.15 μJ . The discrepancy indicates that radiation probably missed the cell window, a result that is not surprising given that we did not first test the pointing and focusing characteristics for 5-cm targets.

Had a higher-transmitting, 1000-Å aluminum filter (~ 80 instead of 35%) been used with better-reflecting mirrors (~ 35 instead of 20%), there would have been enough energy in the cell to produce gain-saturated ASE, which would dominate argon emission at 476.5 nm. Such conditions would have produced a laser at 476.5 nm, and the device could then be used as an x-ray-laser detector. In any case, we successfully used the soft x-ray laser in a useful application. We are

Fig. 3-145. Soft x-ray-laser emission is directed into the x-ray mirror system, collimated, and pointed onto a film pack equipped with 100- μm -diam wire cross hairs. The x-ray-laser axis is made co-linear with two x-ray mirrors and alignment cross hairs.

encouraged to attempt an experiment using sodium with the same experimental configuration, but with longer targets and higher-reflectivity mirrors.

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Implosion and Hydrodynamics Experiments.

Preliminary Rayleigh-Taylor Experiments. The objective of Nova Rayleigh-Taylor experiments is to radiatively accelerate a mass-modulated package and to measure the growth rate of the Rayleigh-Taylor instability seeded at the wavelength of the initial modulations.^{25, 26a} For directly driven targets, the effect of ablation has been shown heuristically and numerically to stabilize Rayleigh-Taylor growth by ablative flow.^{25, 26a} Drive wavelength is reduced by using shorter-wavelength lasers; predictions of the growth rate vary from less than classical for high-opacity materials^{26a, 26b} to essentially zero for low-opacity materials.

The theoretical Rayleigh-Taylor growth rate of a perturbation is $\gamma = \min [A(ka)^{1/2} - kV_A, A(ka)^{1/2}]$, where V_A is the ablation velocity, and A is the Atwood number $(\rho_{\text{high}} - \rho_{\text{low}})/(\rho_{\text{high}} + \rho_{\text{low}})$. With short-wavelength drive, the ablation velocity is higher,^{26b} and density gradients are shallower, leading to a lower effective Atwood number.

This year, we made preliminary measurements on two modulated target

Fig. 3-146. Scheme for backlighting through a target and simultaneously obtaining a reference-unobstructed view of backlighting target for absolute opacity measurements via two sectors of Wölter x-ray optics.

Fig. 3-147. Dysprosium backlighter spectrum after attenuation through yttrium filters and fluorosilicone, convolved with reflectivity of the Wölter x-ray microscope.

materials that were radiatively accelerated by Nova. The high-opacity material, fluorosilicone, exhibits some growth of initial modulation, in approximate agreement with the value predicted numerically. The low-opacity material, plastic, gives no observable growth within the range of current instrument sensitivity.

For the high-opacity target material to be radiatively accelerated, we chose fluorosilicone, $C_4H_7F_3SiO$. The fluorine and silicone in this compound fill in windows in the x-ray opacity at their K-photoabsorption edges, making it an effective high-opacity material. Fluorosilicone also can be cast and has low density ($\rho = 1.3$). The material, supplied by Exitech Ltd., Blaydon, Oxon, England, was cast into the form of packages with an average thickness of 28 to 35 μm , peak-to-valley modulations of 1 to 5 μm , and wavelengths of 50 or 100 μm . For all of our experiments, the average thickness, peak-to-valley modulation, and

wavelength were 28, 3.5, and 50 μm , respectively. We shot some targets with modulated polystyrene packages of the same wavelength, but with 10- μm peak-to-valley modulations and 60- μm thickness.

We used eight of the ten beams of Nova to radiatively drive the packages. We used beam 3 as a backlighter in either the short-pulse mode (0.25 ns) or the long-pulse mode (2.0 ns) at a wavelength of 0.53 μm . Beam 3 was focused to an ~ 0.5 -mm spot on the 1-mm-square, dysprosium-foil backlighter. We used a 25- μm hole in the center of this foil to define a position on the backlighter for prealignment of targets. The packages were radiatively accelerated parallel to the line of sight, with a line of sight through the package to a dysprosium foil behind the package. The target was prealigned using the technology developed from the mix experiments (see the article, "Characterization of a High-Magnification X-Ray Microscope on Nova," later in this section).

We observed transmission of x rays from the dysprosium backlighter through the accelerated packages with the Wölter x-ray microscope. We obtained the shadow of the driven-fluorosilicone target and a separate view of the backlighter in the flash mode by forming two images of the backlighter on the Wölter x-ray microscope using the scheme shown in Fig. 3-146. This scheme works because the backlighter is slightly out of

focus and can be seen by one of the two open sectors of the Wölter over the top of the target.

To isolate a well-defined band of x rays for quantitative absorption measurements, the dysprosium backlighter was filtered by an yttrium filter. The dysprosium backlighter gives a spectrum dominated by the $3d \rightarrow 4p$ transitions of states of ionization close to nickel-like ions. The combination of this spectrum—the L_{III} edge of yttrium and a fluorosilicone package—is essentially a band with a width of 250 eV centered at 1.8 keV.

To prove that we could obtain quantitative results with this system, a shot was fired with various thicknesses of cold fluorosilicone and plastic packages on the Wölter film pack, thereby forming a test-step wedge. Figure 3-147 shows the theoretical attenuation. The yttrium L_{III} edge and the silicon K-edge at 2.07 and 1.84 keV, respectively, are clearly visible. (Dysprosium was chosen because the $3d \rightarrow 4f$ clusters from nickel, copper, zinc, and gallium states of ionization are below the yttrium L_{III} edge.)

Figure 3-148 shows measured transmission. The transmission ranges from 100% (no absorbers) to 0.01. Theoretical transmissions calculated for the spectrum of Fig. 3-147 and using cold opacity values

are shown for comparison. The good agreement between Figs. 3-147 and 3-148 corroborates our measurements of the spectrum of x rays used for the measurement as well as the x-ray transmissivity of the package. Quantitative radiographic analysis is notoriously difficult; thus, we believed it was important to verify the system's capability by such simple experiments.

We obtained most experimental results with a 0.25-ns backlighter and x-ray film as the detector on the Wölter x-ray microscope, or with a 2-ns backlighter onto an x-ray streak camera as the detector. Figure 3-149 shows a flash radiograph of

Fig. 3-148. Measured transmission of fluorosilicone and plastic step wedge placed on the x-ray film pack of the Wölter x-ray microscope.

Fig. 3-149. Radiograph of accelerated package with a 0.25-ns backlighter flash delayed 1.5 ns from start of drive pulse.

an accelerated package at 1.5 ns, the time delay that gave us the greatest observed modulation. The double view of the backlighter allows us to predict the position of the backlight spot with respect to the image, as shown in Fig. 3-149. The relative absence of x rays outside

the spots shows the small effect of self emission on this shot. The three accelerated packages were a modulated fluoro-silicone package (center), a modulated plastic package (left), and a flat fluoro-silicone package (right). The two gaps where the packages are imperfectly

Fig. 3-150. Streaked transmission of modulated fluorosilicone. The package covers the right-hand side of the image. Modulations in x-ray intensity emerge from noise at the end of the backlighter pulse. (a) Lineout at 1.4 ns; (b) backlit image; and (c) lineout at -1.4 ns.

butted together transmit a higher level of x rays. Initial modulations on the central accelerated package have grown and are clearly visible in Fig. 3-149. By using the film calibration curve (from Industrex C) and the intensity of unattenuated backlight from the other image, the transmitted signal was converted to modulation in opacity. The brightness of the image that was not transmitted through the fluorosilicone package allows us to measure the absolute opacity at a 1.5-ns time delay. We also made measurements at 0.2- and 0.8-ns delays, which gave smaller modulations in opacity.

In addition, we obtained a few streaked-backlit shots. Figure 3-150 shows the results from a shot for a backlighting pulse that was flat-topped in time, starting 0.5 ns before the drive pulse. The package half covered the observation hole, with a fiducial wire out of the field of view. Modulations can be clearly seen at the end of the backlighting pulse, albeit at a low level. At this stage, the effects of the x-ray streak camera on the contrast transfer function (CTF) of the system are imperfectly known.

In measuring modulations in opacity, we can correct for at least two possible systematic experimental errors: self emission and poor instrument contrast. Figure 3-151 shows instrument contrast measured for an infinite-contrast mask on film. The measured CTF depends on the size of the sector of the x-ray optic used and the direction of measurement. Moreover, because of modulations in brightness of the backlighter, the CTF varies over the area of the backlighter beam. A simple model suggests that scatter occurs primarily in the plane of incidence of x rays for the section of the annulus used. Contrast in a direction normal to the plane of incidence, i.e., the horizontal direction, is therefore high (25) in Fig. 3-151, but low (~ 3) in the plane of incidence as shown by the vertical lineout of Fig. 3-151. When a large sector is used, the directional scatter from each part of the sector adds to give a scatter that becomes more isotropic as the angle of the sector increases. Furthermore, there appears to be a variation in contrast around the azimuth of the optic.

For flash-backlit measurements, we used the instrument with a 5° horizontal

Fig. 3-151. Flash radiograph of x-ray-opaque resolution grid. Upper and lower images are transmitted through upper 30 and lower 150 sectors of the Wölter. Contrast varies with direction and is best for a small sector normal to the plane of incidence. A contrast of 4 is an MTF of $(4 - 1)/(4 + 1)$.

section and vertical target modulations, giving a CTF of ~ 0.50 for a $50\text{-}\mu\text{m}$ period. This method reduced all perceived contrasts $\Delta\tau_{\text{perc}}$ by approximately $\Delta\tau_{\text{perc}} = 2\tanh^{-1}[\text{CTF} \times \tanh(\Delta\tau)]$. However, in detail, the reduction depends on the shape of the point-response function. We require further analysis of this effect before generating measurements of quantitative opacity.

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Measurement of Turbulent Mixing at Unstable Interfaces in Driven Foils.

When an interface between materials of differing density is accelerated, small perturbations can grow. For continuous acceleration, the phenomenon is known as the Rayleigh-Taylor instability²⁶³; for shock acceleration, it is known as the Richtmeyer-Meshkov instability.²⁶⁴ Although analysis via linear perturbation theory is straightforward for incompressible flow,²⁶³ the nonlinear phase of compressible fluids at high pressure is complex.

A limited number of experiments conducted at low pressure in the nonlinear phase^{265,266} give an empirical value for the thickness of the region of intermixing between two unstable materials subject to constant acceleration, namely:

$$h = 0.14 A(0.5 at^2), \quad (56)$$

where A is the Atwood number, and $0.5 at^2$ is the distance moved by the interface. For a shock-driven interface, where v is the piston velocity, if $d^2h/dt^2 = 0.14$ from Eq. (56), and as $h(0) = dh(0)/dt = 0$, then

$$h = 0.14 Avt. \quad (57)$$

Although such an analysis is a simple treatment that ignores damping of turbulence and is based on experimental conditions far from those in high-pressure plasmas, it approximates more complex calculations.²⁶⁷

In laser-driven fusion, mixing occurs both during inward acceleration of the shell and during deceleration of the shell by a lower-density, higher-pressure fuel at stagnation. However, the pressures of

matter are significantly higher than those in the experiments on which the simple model is based. For this and other reasons, we are engaged in a series of experiments on Nova to measure the level of mixing in laser-accelerated, hydrodynamically unstable targets.

In this article, we present the first indications of mixing in impulsively accelerated, layered, plane targets. The novel technique of point-projection spectroscopy has enabled us to identify the position of the materials and the amount of mix. Control experiments in which no mix should occur because the target is designed with a small Atwood number are indeed seen to exhibit little mix.

Previous experiments on hydrodynamic instabilities have measured seeded, single-mode growth²⁶⁸⁻²⁷⁰ by spectrally integrated x-ray-transmission measurements in which the accelerated target is either face-on^{268,269} or side-on²⁷⁰ to an accelerated target. In the more discriminating technique of point-projection spectroscopy, spectral information is retained in the radiographs so that characteristic edges and lines of different materials can be uniquely identified.

Figure 3-152 shows the technique we used in our experiments on Nova. A planar disk with an interface between a low- and a high-density material was accelerated at high pressure. After some delay, a flash of x rays was produced by focusing one of the Nova backlighting beams with a pulse length of 100 to 250 ps onto a $20\text{-}\mu\text{m}$ -diam fiber coated with mixtures of platinum, gold, and bismuth. The spectrum of the resultant x-ray flash was quasicontinuous in the region from 2.5 to 3 keV and had spatial and temporal extents of $\sim 15\text{ }\mu\text{m}$ and 100 to 200 ps, respectively. Because the flash came from a source small enough that it was essentially a point, we used it to project an image of the package in-flight onto x-ray film after reflection off a KAP Bragg diffraction crystal. However, because the crystal reflected only when Bragg's law was satisfied, the image had spectral as well as spatial resolution in one direction.

We selected the low- and high-density package materials to have photoabsorption edges in the wavelength range over which an image is formed. A serendipitous

Fig. 3-152. Schematic of the technique of point-projection spectroscopy.

feature of this technique is that absorption lines form close to the cold edges as material is heated and ionized due to vacancies forming in the otherwise-full upper states. In this way, the extent of the material could be identified by the degree to which spectral features of the different materials of the two layers overlaid one another.

The accelerated packages consisted of disks of molybdenum accelerated by a low-density ablator of either chlorinated plastic or sulfonated plastic and tamped by a $2\text{-}\mu\text{m}$ layer of plastic. The high Atwood number in this package came from the density mismatch between molybdenum (with a density of 10) and the plastic (1.3). We designed some experiments as nulls with a low Atwood number achieved by having an ablator of sulfonated plastic and a "payload" of chlorinated plastic. The initial Atwood number of these targets is low; we calculate that after shock compression the Atwood number still does not exceed 0.2.

In the initial stages of this experiment, radiographs of the in-flight packages showed a bowed package with a ratio of displacement of the middle of the package to the edge of the package of 1.3. Because the measurement of mix is a line-of-sight measurement, the bowing would make an unmixed package appear to be mixed. We eliminated the problem by mounting the ablator of these packages

in a guard ring of plastic so that the edges of packages were not at the edges of the drive.

Figure 3-153 shows an example of data obtained in this manner where both the package and a spatial fiducial can clearly be seen. A package consisting of a Parylene-C ablator (initial density of 1.3 g/cm^3 , 35 wt% chlorine) driving a $2\text{-}\mu\text{m}$ -thick molybdenum foil (initial density $\sim 10.2\text{ g/cm}^3$) with an initial aspect ratio of 5 was accelerated by eight of the Nova beams. The flash radiograph shown in Fig. 3-153 was taken at a time when the package was coasting, its center of mass having traveled 2 package diameters from its starting position with some decompression evident. The crystal spectrometer had a flat KAP crystal, which has the effect of making the magnification equal in the two directions, but no bowing of the package is visible. The tip of the spatial fiducial was $10\text{ }\mu\text{m}$ from the axis of the cylindrical package and gives a reference for the radial edges of the package and its axial position. The package has expanded radially by a factor of 1.5. The fiducial also demonstrates the spatial resolution of backlighting. Moreover, the motion smearing due to the package velocity is small because the duration of the flash is short compared to its time delay relative to the drive.

We obtained most of the detailed information about the package from the

Fig. 3-153. Spectral radiograph from a package consisting of a chlorinated ablator driving a molybdenum package.

absorption spectra in Fig. 3-153. The back-lighting spectra shows the spectra of gold and bismuth. The silver edge arises from the filter placed over the film pack. The K edge of chlorine lies spectrally in the middle of the package and can be seen shifting to higher energy in the chlorine closer to the hot ablator. The molybdenum from the package is characterized by lines appearing at the position of the L_{III} and L_{II} edges, although a detailed lineout shows these photoabsorption edges. The degree to which these chlorine and molybdenum features appear to overlap indicates the level of mix. The spectral features of the bismuth-and-gold-cosputtered backlighter are mainly from the $3d \rightarrow 4f$ transitions of the states of ionization close to the nickel-like state of ionization and are evident in Fig. 3-153.

To display the spectral absorption by the package, we divided part of the unobscured spectrum taken from across the

bottom of the image in Fig. 3-153 into all other points of the image at the same wavelength. To achieve this result, we used the data-analysis code IMAGIC, which has a provision for dividing along the spectral lines that are, of course, slightly curved. Figure 3-154 shows the resulting divided image. This figure shows absorption as a function of wavelength and position in the horizontal direction and as a function of position only in the vertical direction.

Absorption features at the K and L edges are more clearly evident. These features are $1 \rightarrow 3$ and $2 \rightarrow 4$ transitions in chlorine²⁷¹ and molybdenum, which become visible when chlorine and molybdenum are heated and ionized into L and M shells, respectively, creating vacancies in the otherwise-full upper states. Because the line opacity is greater than the continuum opacity, the features are sensitive qualitative indications of the extent of these materials along the line of flight. Once the line strengths and widths are known, the features can become quantitative measures of material density. In Fig. 3-154 the $1s \rightarrow 3p$ transitions of chlorine at 2.82 keV can be seen to penetrate approximately half-way through the foil, as identified in the figure. The line feature at the position of the L_{III} edge is due to $2 \rightarrow 4$ transitions of slightly ionized molybdenum and clearly overlaps the chlorine K line in the acceleration direction.

To fully exploit the backlighting capabilities of Nova, we employed two laser beams to generate two, separate back-lighting sources on the same shot. In two such sequential radiographs, the lines of sight of the two views were separated by 44° . In this case, we used a sulfinated plastic (polyphenol sulfide) as an ablator to accelerate a high-density molybdenum foil. This ablator allows a better measurement of the position of the ablator because molybdenum has a lower opacity at the sulfur K edge than at the chlorine K edge. Once again, overlap of the sulfur K edge and the molybdenum L_{III} features suggested apparent mixing. This target is predicted to have a higher degree of mixing than the previous chlorinated-ablator target since sulfur in the ablator is a more effective x-ray-preheated shield. The molybdenum thus remains cooler

and at a higher density. Indeed, a larger overlap region was clearly seen.

The apparent mixing of ablator and the high-density foil could be a line-of-sight effect. If the package were dished (i.e., central-drive pressure greater than edge-drive pressure along the line of sight) there would be an overlay of the two materials in the line of sight and, consequently, apparent mixing. Figure 3-154 shows that there is little such dishing in the plane normal to the direction of observation. However, edge effects due, for example, to blowby of the ablator are harder to disprove; therefore a null target was shot.

Our null target consisted of sulfinated plastic as an ablator driving chlorinated plastic as a payload. In simulations, this combination results in a low Attwood number because of the similar densities of the two plastics and, therefore, little mixing. Edge effects and dishing would, however, be similar to those for the high-mix packages. Figure 3-155 shows the result from a null package: little mixing is evident, and the sulfur and chlorine lines are in distinctly different regions of space in the direction of motion.

The simplest expression for a theoretical value of mix thickness taken from Eq. (57) is 0.14 times the distance moved

▲ Fig. 3-154. Spectrally divided image corresponding to Fig. 3-153. Most spectral features of the backlighter have been divided out.

Fig. 3-155. Spectrally divided image of a null target where no mixing should occur. Sulfinated plastic ablator is accelerating chlorinated plastic, resulting in an Attwood number near zero. Sulfur and chlorine lines and edges do not overlap; thus, little mixing has occurred.

by the package. Our measured values are in good agreement with this value, although several canceling effects have been ignored. Such effects include the decompression of the package as it travels and further acceleration of the package once it has been shock-accelerated. A detailed comparison of measured and calculated mix thickness depends on knowledge of the drive profile and the equations of state of the materials. This analysis will be reported in a future publication.

In conclusion, we have demonstrated the first use of point-projection radiography to demonstrate mixing in foils radiatively accelerated by a laser plasma. Possible systematic errors in the experiment are discussed and eliminated by the use of a null target for which mixing does not occur.

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Interaction of Matter with High-Intensity Lasers. Our investigation of the interaction of high-intensity laser radiation with matter has been divided into two broad categories. The first area is concerned with the response of bulk material to intense, short-pulse, visible and near-ultraviolet radiation. These experiments are designed to extend our knowledge of laser-plasma interactions by examining plasma formation and heating on time scales near 1 ps. At these time scales, complications associated with the hydrodynamic motion of plasma are eliminated. Such experiments are described in the article, "X-Ray Generation from Preformed Plasmas with Picosecond Lasers," later in this section.

The following two articles are devoted to our second area of interest: the interaction of intense laser light with matter under collisionless conditions. Our experiments in this area are designed to study the response of individual atoms to the strong laser field. By learning how energy is coupled into the atom, we can begin to understand the initial stages of

plasma formation in solid targets, and we may eventually learn how to store energy in multiply excited states of the atom. Such highly excited atoms may serve as an efficient source of coherent, extreme ultraviolet or x-ray radiation.

Author: M. D. Perry

Nonresonant Multiphoton Ionization of Rare Gases. To date, we have constrained our investigation to the rare gases. For rare-gas species, the energy of a single laser photon is insufficient to excite or ionize the atom. Hence, excitation or ionization can occur only by multiphoton processes. The probability of such a process is a function of both the laser frequency and intensity. However, the majority of experiments have been limited to studying only the intensity dependence of multiphoton ionization at the fixed frequencies of high-power excimer²⁷² or Nd:glass²⁷³ laser systems.

We have investigated both the intensity and frequency dependence of multiphoton ionization by using the tunability of our picosecond-dye-laser system.²⁷⁴ In this article, we describe our work on nonresonant multiphoton ionization of rare gases at intensities such that the strength of the applied field approaches a significant fraction of the Coulomb field experienced by the outer electrons. In this regime, it may be inadequate to treat the applied field merely as a perturbation on the atomic system.

Our experiments consisted of measuring the absolute yield of multiply charged ions of argon, krypton, and xenon as a function of laser intensity. The emphasis on determining the actual numbers of ions produced, as opposed to only relative yields, is dictated by our desire to perform quantitative comparison with theory.

Figures 3-156 and 3-157 show some of the experimental results²⁷⁵ at a laser wavelength of 586 nm. The general shape of the curves in Fig. 3-156 is similar to that observed in previous experiments.²⁷⁶ When the ion yield is relatively low, the parent atom/ion population is not depleted anywhere in the focal volume, and ion yield is a strong nonlinear function of peak laser intensity. In perturbation

Fig. 3-156. Detected absolute ion yield of (a) argon and (b) xenon vs laser intensity.

theory, ion yield is proportional to I_0^N where N_0 is the minimum number of photons required to ionize the parent species. When the ionization probability approaches unity, the parent species becomes depleted throughout the focal volume. Further contribution to ion yield then comes from the expanding focal volume, which grows asymptotically as $I_0^{3/2}$ (Refs. 277 and 278).

Figure 3-156 shows the threshold intensity, defined as the intensity at which the ionization probability is 10^{-4} as a function of the ionization potential. The

smooth, monotonic increase in threshold intensity for production of the higher charge states is a general result, independent of the atomic or ionic system investigated. This relation suggests that, far from resonance, the ionization rate is a fairly weak function of the details of atomic structure. Instead, the ionization rate appears to be determined primarily by a gross feature of the atom, its binding energy. This is the same qualitative conclusion reached by Keldysh²⁷⁹ by an application of Fermi's golden rule in which a "dressed" or Volkov²⁸⁰ state was

Fig. 3-157. Threshold laser intensity, defined as the intensity at which ionization probability is 10^{-4} , vs ionization potential of a given charge state.

used to describe the outgoing photoelectron. This theory was later extended by Faisal²⁸¹ and Reiss.²⁸² At the intensities considered here, the differences between the Keldysh and perturbation approaches are significant.

We have found that the "dressed" electron theory is in quantitative agreement with our experimental results for production of only the first charge state. The agreement between theory and experiment is poor for the second charge state and becomes progressively worse for higher charge states. We attribute this result to neglect of the effect of the Coulomb field of the residual ion on the outgoing electron.

We extended the Keldysh-Reiss-Faisal theory by using the Wentzel-Kramers-Brillouin (WKB) wave function of an electron in a constant potential, subject to an intense, oscillating electromagnetic field for the outgoing electron.²⁸³ The magnitude of the potential was set equal to the ionization potential of the parent atom/ion. The results of our calculations with no adjustable parameters appear as the solid curves in Figs. 3-156 and 3-157.

Our model has recently been applied to the experiments of others²⁸⁴ and was once again found to be in good agreement with no adjustable parameters. To date, our model has generated the best agreement between theory and experiment for non-resonant multiphoton ionization.

Authors: M. D. Perry (LLNL) and O. L. Landen (Brigham Young University)

Resonant Multiphoton Ionization.

The multiphoton ionization probability of an atom is a function of both laser frequency and intensity. The frequency dependence is a result of the influence of multiphoton resonances to intermediate atomic states. In the previous article, we described a series of experiments in which the laser frequency was tuned to avoid any intermediate dipole-allowed resonances. Here, we describe a series of experiments designed specifically to examine resonant enhancement of the multiphoton ionization probability.²⁸⁵

For wavelengths > 250 nm, three or more photons per atom are needed to ionize the valence electrons of rare-gas atoms. The high order of these multiphoton processes requires laser irradiances I above 10^{10} W/cm² to ionize a substantial fraction of the atoms within a typical laser pulse length < 10 ns. For such laser intensities, the electronic levels of the atom undergo measurable energy shifts²⁸⁶ due to the AC Stark effect. These shifts have been well modeled by perturbation theory²⁸⁷⁻²⁸⁹ for laser intensities up to 10^{12} W/cm².

However, no data existed on the behavior of the levels when the laser field E_L reached a few percent of the value of the Coulomb field E_C on the outer electron, where the ratio $E_L/E_C = [I/(7 \times 10^{16} \text{ W/cm}^2)]^{1/2}$. We now present such data in the form of detailed three-photon resonance profiles for krypton obtained via four-photon ionization in the intensity range from 3×10^{12} to 8×10^{13} W/cm². The resonances exhibit up to 0.3-eV shifts and are still well described by perturbation theory.

Our experiment consisted of measuring the number of singly charged krypton ions produced under collisionless conditions as a function of laser frequency and intensity. The output of a picosecond-dye-laser system operating at 2.5 Hz was frequency doubled in a 1-cm potassium dihydrogen phosphate (KDP) crystal to give a 0.5-mJ, 1.3-ps, transform-limited 0.1-nm-bandwidth beam tunable between 285 and 310 nm. The linearly polarized ultraviolet beam was separated from the remaining fundamental by a prism and focused by an $f/15$ optic in krypton gas at

a pressure of 10^{-6} to 10^{-5} Torr. The ions produced on each shot were extracted by a 5000-V/cm applied field, sorted according to charge by a time-of-flight spectrometer, and detected by direct impact on a microchannel plate.

Figure 3-158 shows krypton yields with clear resonant structure for increasing peak-laser intensity. Each datum point corresponds to a 20- to 60-shot average. The vertical lines denote resonance positions in the limit of zero intensity. The observed resonances, all shifted to shorter wavelengths, are taken to be the $4p \rightarrow 4d$, $4p \rightarrow 4d'$, and $4p \rightarrow 5d$, $\Delta l = 1$ and 3 allowed three-photon transitions identified in the caption of Fig. 3-158, with j, l coupling notation. As expected, the resonances shift and broaden as the intensity is increased until the ionization process saturates, as is the case for the prominent $4p \rightarrow 4d[3/2]$ resonance at 2×10^{13} W/cm 2 .

Figure 3-159 shows the $4p \rightarrow 5d[1/2]$ krypton resonance in greater detail for increasing peak-laser intensity. The asymmetric profile is typical of laser pulses with realistic spatial and temporal intensity distributions.

The resonance profile can be modeled by an effective two-level atom, with the assumption of a rate-limiting three-photon-excitation step followed by rapid one-photon ionization from the excited state. The ionization rate $W(\omega, I)$ as a function of laser detuning from a three-photon resonance, assuming a resonance shift proportional to intensity with a width dominated by lifetime broadening of the excited state, is given²⁸⁷⁻²⁸⁹ by

$$W(\omega, I) = \frac{\Omega^2}{2} \frac{\delta I}{(3\omega - \omega_0 - \alpha I)^2 + \delta^2 I^2}, \quad (58)$$

where Ω is the three-photon Rabi rate (proportional to $I^{3/2}$), αI is the AC Stark shift of the resonance, δI is the photoionization rate of the excited state, ω_0 is the unshifted resonance frequency, and ω is the laser frequency. In the limit of large detuning, the nonresonant four-photon ionization rate, proportional to I^4 ,

Fig. 3-158. Yield for Kr^+ vs laser wavelength for various peak laser intensities I_0 . Vertical lines mark unshifted resonance positions:
A = $4p \rightarrow 5d[7/2]_{3/2}$;
B = $4p \rightarrow 4d'[3/2]_{3/2}$;
C = $4p \rightarrow 5d[1/2]_{1/2}$;
D = $4p \rightarrow 4d'[5/2]_{3/2}$;
E = $4p \rightarrow 6s[3/2]_{1/2}$;
F = $4p \rightarrow 4d[3/2]_{1/2}$;
G = $4p \rightarrow 4d[5/2]_{3/2}$;
and
H = $4p \rightarrow 4d[7/2]_{3/2}$.

is recovered from Eq. (58). The parameters Ω , α , and δ are taken to be independent of frequency over a given resonance profile, a good approximation even for Figs. 3-158(c) and 3-159(e). In addition, the laser bandwidth and the Doppler and radiative lifetime broadening of the excited state could be neglected, even for Fig. 3-158(a).

Below saturation, the total number of ions produced at a given laser wavelength is obtained by the integration of the rate $W(\omega, I)$ over the spatiotemporal intensity distribution at the laser focus. In our experiment, this distribution was found to be well modeled by

$$I = \frac{I_0 \sec h^2 1.76 t / \tau_1}{1 + A \lambda^2 z^2 / \pi^2 r_0^4} \times \exp \left[- \frac{2r^2}{r_0^2 (1 + A \lambda^2 z^2 / \pi^2 r_0^4)} \right], \quad (59)$$

Fig. 3-159. Krypton 4p → 5d[1/2], three-photon resonance profiles for various peak intensities I_0 .

where $\tau = 1.3$ ps, the temporal FWHM of the laser pulse; $A = 20$, an experimentally determined constant accounting for a divergence rather than diffraction limited focus; z is the distance along the laser axis from the focal plane; $r_0 = 12.5 \mu\text{m}$, the $1/e^2$ beam radius at $z = 0$; and I_0 is the peak laser intensity at $r = z = t = 0$.

The peak laser intensity was known with 20% accuracy from an absolute

measurement of the saturation intensity at a nonresonant wavelength of 293 nm. Our determination was made by measuring the maximum ponderomotive energy U_p associated with the photoelectron upon ejection^{290,291}, where U_p is related to laser intensity by

$$U_p(\text{eV}) = 9.3 \times 10^{-14} I \lambda^2, \quad (60)$$

where I is in W/cm^2 , and the laser wavelength λ is in microns.

The curves in Fig. 3-159 represent best fits to the data, normalized to the peak height, but with α and δ kept constant for all fits. The values deduced for α and δ are 3.9 and 1.4 meV/(TW·cm⁻²), respectively, yielding a photoionization cross section from the 5d [1/2]₁ state, σ_{5d} , of 3×10^{-18} cm² at $\lambda = 289$ nm. The fitting error in the ratio δ/α reflects possible departures from a Gaussian spatial profile and hyperbolic secant-squared temporal profile. This error is small ($\pm 25\%$) because the calculated resonance shape is fairly insensitive to the exact focal-intensity distribution chosen and is fairly sensitive to the ratio α/δ .

The 4p-4d[3/2]₁ three-photon resonance was also well modeled by Eq. (58), yielding $\alpha = 8.0$ meV/(TW cm⁻²), $\delta = 4.0$ meV/(TW cm⁻²), and, hence, $\sigma_{4d} = 8 \times 10^{-18}$ cm² at 300 nm. We note that the ratio of the measured photoionization cross sections from the 4d and 5d states, which was 2.7, is in agreement with the calculated value of 3 obtained from a simple quantum-defect approximation.²⁹²

In summary, our experiment extended to 3×10^{13} W/cm² the intensity range for which resonance enhancement of ion yields has been observed. The resonance profiles recorded for over one decade's changes in laser intensity were well modeled by a simple, single-rate analysis, even when the laser electric field reached 3% of the atomic field strength.

Authors: O. L. Landen (Brigham Young University) and M. D. Perry (LLNL)

X-ray Generation from Preformed Plasmas with Picosecond Lasers. The interaction of a high-intensity, picosecond laser beam with a preformed laser plasma has been shown to yield an ultra-short (< 10-ps), ultras-small-diam (< 3- μ m) source of 1-keV x rays.^{293,294} In addition, the thickness of the 1-keV emitting region was measured as less than 0.3 μ m, ideal for use as a point backlighter source. The short duration of the x-ray burst could be explained by rapid cooling of the hot emitting plasma slab by thermal conduction. The primary absorption mechanism for producing the hot, transient plasma was determined to be resonance absorption, which efficiently couples laser energy into the electrons,

Fig. 3-160. Time-integrated x-ray yield from gold target in a 400- to 1000-eV band vs angle between laser beam and target normal. (a) High-intensity, short pulse preceded by a low-intensity, long pulse. (b) Yield without the low-intensity, long pulse.

resulting in a rapid increase in electron temperature and ionization state of the plasma.

The fraction of energy absorbed via resonance absorption and reemitted through x rays is a function of the electron-density scalelength at the critical density and the angle between the laser axis and the target normal²⁹⁵ (see Fig. 3-160). Thus, we can, in principle, measure plasma-density gradients on a picosecond time scale for any preformed plasma at any time.

Figure 3-161 shows the experimental setup for such a measurement. The experiment, which is scheduled to begin in early 1988, consists of producing a laser plasma with a "pump" picosecond laser pulse and measuring the x-ray emission resulting from resonance absorption of a delayed, second "probe" picosecond pulse in the preformed plasma. By varying the time delay between the two pulses, we can measure the evolution of the plasma density gradient and, hence, the time history of the plasma acceleration away from the target surface.

The laser is an amplified, synchronously pumped, picosecond dye laser operating at 2.5 Hz. The laser-beam output is 2 mJ in 1 ps at $\lambda = 587$ nm. A nanosecond prepulse, present in earlier experiments,²⁹⁴ is kept below 0.1 μ J in

Fig. 3-161. Configuration of the pump-probe experiment.

energy to avoid damaging the target. The beam is split into two beams to provide pump and probe beams that are optically synchronized to each other. The pump beam is sent through a variable-delay line and focused by an $f/20$ lens to a 50- to 100- μm -diam spot on the target. The probe beam is focused by an $f/7.5$ lens to a 25- μm -diam spot within the larger focal spot defined by the pump beam. The pump beam polarization is optically rotated to provide S polarization on the target, while the probe beam remains P polarized (in the plane of incidence), the optimum polarization for resonance absorption to occur. The ratio of probe-to-pump intensities on target is greater than 3 to ensure that the majority of x-ray emission is due to the probe pulse at all angles of incidence.

Targets consist of 3-in.-diam, polished-silicon wafers overcoated with 5000 Å of gold. The target is mounted on remotely driven x,y translation stages so that the laser can be fired on a fresh target surface on each shot. In addition, the target assembly is mounted on a remotely driven, vertical-axis rotation stage such that the intersection of the two laser beams at the target surface is also on the

axis of rotation of this stage. Thus, the target can be rotated over 70° while keeping both beams focused on the same spot on the target. Alignment is accurate to within 25 μm .

Under ideal circumstances, the pump beam should maintain a constant angle of incidence as the target rotates, but this objective proves to be too complicated experimentally. Instead, both beams are fixed at an angle of 35° to one another (see Fig. 3-161). In this manner, the maximum angle of incidence on target for the pump beam is 35°, representing only a 20% change in focal-spot area from normal incidence irradiation. In contrast, the probe beam angle of incidence varies between 0 and 70°.

Plasma x-ray emission is recorded by a filtered channeltron detector placed 15 cm from the plasma. The 60° angle between the detector line of sight and target normal is fixed by mounting the detector on a support bracket attached to the rotation stage.

A run will consist of 312 shots fired as the target rotates by 70°. Between runs, the delay between pump and probe beam will be varied by a few picoseconds. The x-ray signal will be sent to a gated boxcar integrator and computer for further averaging and analysis.

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Diagnostics Development

Streak and Framing Camera Technology.

Flat-Field Characterization of Optical Streak Cameras. Last year we reported techniques for calibrating image gain nonuniformities (flat-field) and geometric distortion in optical streak cameras.²⁹⁶ These characteristics of streak cameras can distort the amplitude response of measured signals by as much as a factor of 2 (see Ref. 297); however, with careful characterization of these effects, measured streak signals can be corrected by analysis software when the calibration data for each streak camera are known.

Our efforts during 1987 were directed toward two goals. The first goal was to characterize several new charge-coupled device (CCD) optical streak cameras to be installed on Nova. Five new CCD streak camera systems were installed after being fully characterized for flat-field response and geometric distortion. The second goal was to better understand the characterization measurements. Our progress toward these goals is summarized below.

To characterize the effects of gain nonuniformities and geometric distortion in any imaging system, we measure the response to a uniform or known input signal. In a streak camera system, a spatially and temporally uniform input signal is used to generate an output signal called the flat-field response. Since the amplitude of the input signal is flat, the flat-field response gives the point-for-point amplitude response due to the effects of system gain nonuniformities and geometric distortion. Nonuniformity in the flat-field response can be used to

correct arbitrary input signal distortions and therefore to improve the accuracy of the measurement. The transformation between input and output signals I_i and I_o is given by the equations

$$I_i(x, t) = [I_o(x', y') - I_{\text{bkg}}(x', y')] \times \left[G(x', y') \frac{dx' dy'}{dx dt} \right], \quad (61)$$

$$\left(\frac{dx}{dt} \right) = \begin{pmatrix} \frac{\partial x}{\partial x'} & \frac{\partial x}{\partial y'} \\ \frac{\partial t}{\partial x'} & \frac{\partial t}{\partial y'} \end{pmatrix} \begin{pmatrix} dx' \\ dy' \end{pmatrix}, \quad (62)$$

where (x, t) are the input coordinates, (x', y') are the output coordinates, $I_{\text{bkg}}(x', y')$ is the background image that results from dark current in the CCD, and $G(x', y')$ is the gain function. The last term is a correction to the amplitude from the Jacobian transformation from (x, t) coordinates to (x', y') . Equation (62) describes the mapping between input coordinates (x, t) and output coordinates

Fig. 3-162. Flat-field response measurements for a streak camera operating with sweep durations of 3.5 and 10 ns. The lineouts show the raw data before correcting for the Jacobian transformation for each sweep rate.

(x',y') using the Jacobian matrix. If the input signal is flat, i.e., if $I_i(x,t)$ is constant, then the measured output signal $I_o(x',y')$ is basically the reciprocal of the flat-field response (the product of G and the Jacobian). To use these characterization data to correct input signals, one simply multiplies a measured output image by the normalized flat-field response image. The task is easily performed with a mini-computer and takes only a few seconds of CPU time for two 0.5-Mbyte images.

To characterize the geometric distortion produced by the streak camera, a spatial and temporal grid of equally spaced pulses is generated as the input signal and the distorted image produced by the streak camera is recorded. Since the grid at the input has a constant spacing in both the spatial and temporal directions, the nonlinear spacing in the output image may be measured, and the transformation from input to output may be calculated from Eq. (62).

Using the geometric distortion image, the Jacobian transformation may be

computed as described above. The gain function $G(x',y')$ from Eq. (61) is independent of geometric distortion and should be the same at different sweep rates because the nonuniformities attributed to it are spatially dependent only.

Geometric distortion images were taken for sweep durations of 3.5 and 10 ns; the flat-field response was measured for each. Each image was computer-corrected for its corresponding sweep rate nonlinearity, and lineouts were compared for each. Figure 3-162 shows an example of lineouts taken in the time direction of two flat-field response images before making a correction for the Jacobian. These signals represent the product of the gain function $G(x',y')$ and the Jacobian for both cases and differ because of sweep rate nonlinearity differences. Figure 3-163 shows the same signals after being corrected for the Jacobian and represents the local gain function for the camera. The two profiles in Fig. 3-163 are in good agreement, differing mostly in the higher frequency noise. This result demonstrates

Fig. 3-163. Flat-field response signals of Fig. 3-162 after correcting for their corresponding Jacobians.

that the flat-field response needs only to be measured at one sweep rate, along with the corresponding geometric distortion image. Then it is necessary only to obtain the geometric distortion image at other sweep rates and compute the flat-field response for each based on the transformation corrections.

In summary, we have made significant progress in achieving our goals this year. Several cameras were characterized and installed on the Nova laser facility. Our understanding of the characterization measurements will allow us to significantly improve the calibration of optical streak cameras and the techniques for correcting measured signals.

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Software for the Charge-Coupled-Device Camera. We have replaced the LLNL charge-coupled-device camera that was long used for beam diagnostics with a new, lens-coupled, thermo-electrically cooled CCD camera. The CCD camera is described in more detail in the 1985 Laser Annual.²⁹⁸ At the same time, we replaced the stand-alone software originally used to support operation of the new camera with a new software package. This software supports multi-camera operation, integral with that of Nova, as a component part of laser diagnostics.

The configuration of the system and operation of the camera are similar to that described in the 1985 Laser Annual. The software, on the other hand, is significantly different because of the many integration issues and our need for flexibility to accommodate a variety of potential applications.

The system is configured for a maximum of 18 cameras, and each camera is individually supported by an LSI-11/23 front-end processor (FEP). The FEP is connected via Novanet to a designated VAX 11/780 computer in the Nova laser control system. Novanet is the Nova 10-megabyte local-area network. All system activity is initiated and controlled through the VAX, with the FEPs used as

slaves to perform time-critical activity relevant to camera operation. User interaction with the system is via both video-graphics terminals and the Ramtek-based, laser-diagnostics control room console. On-line data storage is supported by a pair of dedicated RA81 hard disks attached to the VAX.

The FEP software consists of a single, memory-resident image that is down-line loaded from the VAX. The VAX-resident CCD software is a multitask structure centered around a command executive, with separate processes used to support user-interface functions, system synchronization, post-acquisition data processing, and hard-copy generation. Nova shared memory is used for process-to-process communication and the interlocking of activities within the VAX. We selected the multitask structure to prioritize activities and to resolve conflicts that could interfere with time-critical activities. All software is written in Praxis, the implementation language developed for Nova.

The software supports two modes of operation, known as shot and local. The shot mode is for operation of the system as a single entity, with operation supported through the control room console. The local mode is for stand-alone operation of individual cameras, with support through video terminals. Both modes of operation support image and background acquisition, background subtraction, color-enhanced image display, generation of 3-D plots of an image, and lineout plots of processed data. The two modes differ in that the shot mode emphasizes simplicity of operation, synchronized with Nova shot sequences; the local mode emphasizes flexibility, with camera operation determined through the use of setup commands.

Image display and plot generation are supported by both the VAX and FEPs. The VAX supports console Ramtek display, a video terminal, or hard copy as output media. The FEPs use the color video monitor indirectly attached to the frame buffer, which stores CCD images. Although they lack the data-processing and plot-annotation capability of the VAX, the FEPs have the advantage of speed and can generate and display plots

in real time. They also support the display of lineout reference markers that we use as an aid for laser-beam setup.

From the operator's console, user interaction with the system is via Ramtek-supported, touch-sensitive menus and an associated keyboard. We use the menus for command input and the keyboard for cursor control and text entry. The cursor is used along with the color-enhanced display of an image to interactively select lineout positions and widths. From a video terminal, user interaction with the system is strictly from the associated keyboard; we call up menus, select cameras, and issue commands through 2- to 3-letter mnemonics along with required arguments. An input parser permits commands to be concatenated, if desired. If a parameter is either missing or erroneous, then a user is prompted for its entry or reentry.

Each CCD image consists of a 384-column by 576-row array of pixels, or 442 368 bytes of data, and each image is stored as an individual file. Records that are appended to the file provide essential information relevant to identity, camera setup and operation, and pre-established lineout parameters. Each file is uniquely named to identify the shot, camera, and type of data.

We maintain a record of all system activity by means of an on-line log. This log is accessible in the form of a Ramtek or terminal-supported display or as a file that can be edited or printed. The principal purpose of each log is to confirm system operation.

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X-Ray Streak Camera for Hydrodynamic Instability Experiments. Experiments on hydrodynamic instability are concerned with determining the growth of (usually preformed) modulations in an accelerated material. To perform such experiments on Nova, the modulations must have characteristic wavelengths such that the available accelerations produce measurable growth during the time of a laser pulse. The change in amplitude

of modulations is measured as a change in the apparent contrast of a backlit, modulated foil. Such conditions require an instrument whose spatial resolution introduces little degradation of the contrast or, at least, one for which the degradation is known. A discussion of the x-ray microscope used for our experiments is provided in the article, "Characterization of a High-Magnification X-Ray Microscope on Nova," later in this section. An investigation into the spatial resolution of x-ray streak cameras suggested to us that a simple change could greatly improve this part of the system.

A streak camera system may be considered to consist of three parts: photocathode, electron optics, and image intensifier and data readout. Because the photocathode introduces essentially no degradation in spatial resolution of the system, we concentrated on electron optics, image intensifier, and film.²⁹⁹ We found that the image intensifier we used (ITT model 4113) introduces significant degradation in the spatial resolution of the system as measured by the contrast transfer function (CTF) for low-magnification (about 1.2) streak cameras. (The spatial-frequency input to the intensifier is determined by magnification of the electron optics of the streak camera.) Figure 3-164 shows the CTF of a typical intensifier.

One way to improve the overall resolution of our streak camera system is to concentrate on improving the electron focusing of the camera. Such improvement has been done successfully in various commercial and experimental models.³⁰⁰ Such cameras are then limited almost entirely by the resolution of the image intensifier. Image intensifiers with greater spatial resolution are currently available, but with significantly reduced signal gain (about luminous gain 100 compared with luminous gain $>10^4$ for microchannel-plate intensifiers, such as the 4113). Another way to improve the overall spatial resolution is to increase the magnification of the image, either at the input to the camera or in the electron optics. We decided to concentrate on the latter.

The Photochron II streak tube is commercially available from Kentech, Ltd. as

a streak camera with modular power supplies. We calculated the electron optics of the Phototron II streak tube using the SLAC electron-optics program by W. B. Herrmannsfeldt, as modified by P. A. Jaanimagi. Such a calculation shows that the magnification of the optics may be increased by increasing the voltage of the extraction grid in the tube. The normal magnification for this tube is 2.0. The increase in magnification that may be attained without changing the shapes of electrodes is limited; the aperture placed at the crossover of the electron beam appears to limit the magnification to 2.9. We constructed a power supply to replace the commercially available module. Our supply allows us to vary the extraction-grid voltage between +1.6 kV relative to cathode (RTC), to +3.0 kV RTC. We found that a voltage of about +2.8 kV RTC gave us a magnification of 2.9.

We have now characterized the camera in both streaked and static modes and found it to have very good spatial resolution. Figure 3-165 shows a comparison of the modified and original CTF measured in the streaked mode.

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Transmission-Grating Streak Camera.

In 1982 we combined an x-ray transmission grating with a soft x-ray streak camera to provide continuous x-ray spectra from 0.1 to 2 keV as a function of time.³⁰¹ The instrument gives good time resolution ($\Delta t \geq 20$ ps), moderate spectral resolution ($\Delta\lambda \geq 1$ Å), and is unique in terms of this combination of characteristics in its energy regime.

The transmission-grating streak spectrometer (TGSS) has been successfully fielded previously on the Argus and Novette laser systems in various configurations.³⁰¹⁻³⁰⁴ We have now adapted the TGSS for the Nova laser system and have obtained data on all of the target types shot.

With regard to x-ray continuum measurements, we have used the TGSS in practice primarily to provide time profiles in specific spectral regions rather than spectral shapes as a function of

Fig. 3-164. Contrast transfer function for a typical ITT 4113 image intensifier.

Fig. 3-165. Contrast transfer functions for original and higher-magnification Phototron II cameras.

time. Such use is due, in large part, to the continuing disagreement between the TGSS results and those obtained from the Dante-filtered x-ray diode spectrometer. While Dante is quite inferior to the TGSS in terms of time and energy resolution, it retains the advantage of having fairly easily calibrated components and a simple enough configuration to lead us to believe that these calibrations can be combined to provide a total, absolute sensitivity. Until recently, on the other hand, we relied on a calculated grating response and calibrations of generic streak camera cathodes to analyze TGSS data. In addition, there may be areas of nonlinear behavior in the streak camera tube and microchannel plate intensifier that can affect amplitude information. We have now begun to investigate some of these effects to determine whether disagreements between the TGSS and Dante results can be reduced.

Calibration of a nominally 0.3- μ m-period, 0.6- μ m-thick gold transmission grating by J. Kerner at the Lawrence

Fig. 3-166. Calculated vs measured grating response. Grating period = $0.3\text{ }\mu\text{m}$; thickness = $0.6\text{ }\mu\text{m}$; lines/space = 1.2.

Fig. 3-167. TGSS spectral unfolds using aluminum and gold cathodes.

Fig. 3-168. Amplitude corrections for streak camera nonlinearities 1.8 cm from the end of the streak.

Fig. 3-169. Comparison of TGSS and Dante spectral unfolds.

Berkeley Laboratory Center for X-ray Optics has indicated some deviation from the response calculated with these values, as shown in Fig. 3-166. The deviation may be due to differences in the nominal versus actual dimensions of period and thickness or in variations from ideal cross sections of the grating rectangular bar. Additional measurements will be made on a variety of gratings to determine if the source of discrepancies can be found. In any case, we are now using the calibrated response for spectral unfolds.

We have performed partial calibrations of the gold and aluminum cathodes used on the Nova TGSS with the LLNL ion-accelerator subkilovolt x-ray facility. The calibration is partial in the following sense. In the TGSS operating configuration, x-ray photons are first collimated by the cathode slit—in this case, $300\text{-}\mu\text{m}$ wide—before being transmitted through a plastic substrate and the active metal layer. If the calibration were attempted in this way, the area of the metal irradiated would be so small because of the cathode slit that the resulting signal would not be measurable by the standard technique. Instead, we irradiate a large area of the metal directly from the uncollimated back side. This approach requires that we then correct the resulting sensitivity by the calculated transmissions through the plastic and metal layers.

Figure 3-167 shows unfolded TGSS x-ray spectra from two nominally identical Nova shots using a gold and an aluminum cathode and the appropriate partial-cathode calibration. Dante measurements confirm that the x-ray spectra were indeed nearly identical. The gross differences seen in the unfolded spectra in Fig. 3-167 indicate that our partial-calibration technique is not adequate to provide a valid cathode response. In the near future, we intend to attempt full-cathode calibrations in the actual TGSS geometry.

We have developed correction factors for nonlinear effects arising from the streak camera tube and image intensifier by uniformly illuminating the streak camera slit with x rays on the Phoenix laser

system. We measured amplitude profiles along the photon-energy dimension at five different time locations. We then derived correction factors to be used as multipliers of experimental data. Figure 3-168 shows a typical curve.

Such additions to our understanding of the properties of the TGSS have improved somewhat the agreement between the TGSS and Dante results. Figure 3-169 shows an example of this improvement, where the curves have been normalized at 500 eV. We hope that the significant differences yet remaining will be further reduced if our full-cathode calibrations are successful. If such is not the case, then it would become necessary to attempt a calibration of the TGSS as an integral unit, including grating, cathodes, and streak camera, to remove the remaining discrepancies.

Author: H. N. Kornblum

Four-Frame, Gated Optical Imager with 120-ps Resolution. In studies of subnanosecond phenomena, particularly those involving short-pulse lasers, 2-D, time-resolved pictures are extremely useful. In this article we describe a novel approach for framing cameras, which provides four frames with 120-ps-wide gate times. We then discuss a specific application of the camera to illustrate its versatility.

Figure 3-170 shows the gated optical imager (GOI) along with a high-voltage unit that provides dc bias voltages to the camera and a planar-triode pulse generator that provides the gating pulse. The GOI (from Grant Applied Physics, Inc.) is designed for four-frame operation during a single event. Each channel uses a wafer image-intensifier tube, which has an S20 cathode with a quartz window. (We chose the quartz window in our particular camera to take pictures in ultraviolet light. A single-frame GOI was previously used successfully with a fiber-optic face plate.) In fast-pulse operation, the cathode is capacitively coupled to a conducting electrode, which is pulsed relative to the microchannel plate (MCP) of the tube. To turn the tube off and maintain a

narrow gate pulse, the cathode is reverse biased relative to the MCP by approximately 100 V.

The high-voltage driver is a planar-triode pulse generator with an output pulse of 2.5 kV and a rise time of less than 1.5 ns. This pulse is then shortened to approximately 100 ps by feeding it to a speed-up module containing a diode that is reverse biased to operate near its avalanche-breakdown point. Because the avalanche-breakdown process is known to occur rapidly (<100 -ps typical time scale), we can use well-characterized

Fig. 3-170. (a) Rear view of the framing camera showing connections with the speed-up module (far right), d.c. bias voltage supply (lower center), and planar-triode pulse generator. (b) View showing the arrangement of four gated intensifier tubes.

diodes to produce a very fast rising pulse. This pulse is then split and fed to opposite sides of the tube to optimize impedance matching and to obtain symmetrical propagation of the pulse across the photocathode. The large voltage pulse is required because the capacitive coupling to the tube leaves only one-tenth of the applied gate voltage on the cathode-to-MCP gap.

Multiple-frame operation is achieved by using multiple-gated intensifier tubes. The operation requires that the optical system provide a separate image for each GOI channel, but such separation is easily accomplished. The output pulse from the planar-triode pulse generator is fed to a single speed-up module, which is split between the four tubes. The relative timing among channels is determined by the relative lengths of the cables between each tube and the speed-up module.

The width of the gate pulse has been verified optically by a Hamamatsu laser diode pulser (pulse width = 80 ps, wavelength = 840 nm) and a photodiode to measure the light emitted at the P20 phosphor. Using a sampling-type setup, we obtain a plot of integrated light output vs relative delay between the gate and the laser. The gate pulse is approximately Gaussian with a FWHM of close to 120 ps. By uniformly illuminating the tube with the diode pulser, we have examined the spatial dependence of the gate time by moving the photodiode receiver across the phosphor; the variation is less than 20 ps.

Because only the cathode is gated in this system, the MCP provides the same gain as in dc operation. The MCP voltage can be varied incrementally in six steps (each step = 50 V) from 920 V (radiant gain = 10^3) to 670 V (radiant gain = 30).

Further characteristics of the tube are as follows:

- The tube has a P20 phosphor and is used in conjunction with a standard streak-camera film holder.
- The aperture of each tube is 18 mm.
- The shutter ratio is $>10^6$.

The latter value is indicated by the tube specifications and has been verified by comparing gated and ungated exposure

times to obtain similar intensity levels with the MCP biased. The lower limit corresponds to defects in the MCP.

We also investigated the spatial resolution of the gated tube. We obtained a contrast transfer function using a linear zone-plate grid. The CTF is greater than 30% at 10 line pairs/mm (lp/mm), which corresponds to the smallest spacing in the target grid. Comparison between the CTF obtained in the fast-gate mode and that obtained by gating the MCP (300- μ s gate time) shows no significant difference. The resolution is limited by the intrinsic spatial resolution of the intensifier tube and is not degraded by external electrodes.

At present, we use the four-frame GOI as a routine diagnostic in conjunction with the Nova laser. With the GOI, we can study the spatial and temporal evolution of thermal emission around the target or look at light scattered by parametric processes driven in laser-produced plasmas.

A Cassegrainian telescope (magnification = 8.47; input f/number = 10; telescope located 2 m from target) images the target onto the GOI; thus, the diffraction-limited object-plane resolution at a wavelength of 450 nm is 9 μ m/lp. In the image plane, the resolution is therefore larger than 76.2 μ m/lp or 13 lp/mm. This is comparable to the image resolution of the tube. Four images of the target of nearly equal intensity are provided by a system of beam splitters and mirrors. Small differences in the relative channel intensities can be compensated by adjusting the MCP gain of each tube individually.

Figure 3-171 shows an example of our experimental use of the GOI. We focused two beams of Nova onto one side of a 63.7- μ m-thick, planar aluminum slab. The focused energy produces a shock wave in the target, which shows up as a bright flash when reaching the opposite side. The figure shows four frames taken during a single event, with each frame separated by 200 ps. The first frame shows emission due to preheating. Shock breakout begins by the second frame and continues in the remaining frames. Although

Fig. 3-171. GOI data showing breakout of a shock wave produced in an aluminum-slab target.

the beams were intended to be overlapping, two distinct spots are observed instead of a single spot. We later determined that alignment errors in the target positioner were the cause of the two spots. Such information would be difficult to obtain with a streak camera alone without fortuitous alignment.

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Characterization of Gated Microchannel-Plate Detectors. We have built and fielded gated, stripline microchannel-plate detectors since 1984 (Ref. 305). We have always coated the

input (or stripline) side with 1 μm of gold and the output (or ground plane) side with 1.5 μm of gold. It now appears that the coating thickness on the input side is excessive, significantly reducing the gain obtainable from the detector. The strong gain reduction is correlated with a slight closing up of the 12- μm openings of the microchannels in the input side of the MCP.

We first observed the gain reduction during our analysis of data from the gated x-ray imager (GX!) used on the Nova laser; we observed that the gain from an area inadvertently shadowed during coating was greater than that from an area coated to the usual thickness (Fig. 3-172). The shadowed area

was produced when we modified a coating mask to make a 12.5- Ω stripline that was open-ended or "split strip." It was not clear at that time how much thinner the gold was, but the higher gain was obvious.

The input sides of our MCPs are coated with impedance-matched striplines for propagation of short (<150-ps), ~1-kV pulses. Such pulses allow us to gate the MCP on and off for a short time, thus temporally resolving or framing an image. One- or two-dimensional imaging is possible, depending on strip width. We use strip widths of 15 mm

(6 Ω), 7 mm (12.5 Ω), or 3.1 mm (25 Ω).

We have always coated these strips with 1 μm (10 000 $\text{\AA}$) of gold to prevent them from being lossy or degrading the fast pulse, as would occur if we used the 1000- $\text{\AA}$ coatings that are standard on MCPs for dc use. The input (front) side of the MCP is coated at an angle of 75° to the normal^{306,307} so that the gold does not penetrate the microchannels deeply (penetration is about 12 μm with this coating angle). The output side is coated at 45° to the normal, which allows the gold to penetrate 25 μm up the channel wall. The deeper coating, called end spoiling, improves spatial resolution by reducing the angular spread of the secondary electrons leaving the plate.³⁰⁸

To quantify the connection between gain and coating thickness, we coated a 40-mm Galileo MCP (channel $L/D = 40$) with four 25- Ω (3.1-mm) striplines. Four successive coatings, each nominally 2500 $\text{\AA}$ thick, were laid down with the e-beam coater. (Profilometer tracings of witness plates coated during the runs indicated actual coating thicknesses of ~2250, 2500, 2800, and 3000 $\text{\AA}$, in order of coating.)

After each coating run, another strip was covered so that the nominal coating

Fig. 3-172. Stripline image with diffuse x-ray source. Arrow shows the region of greater gain than adjacent areas, which was less heavily coated with gold because of an inadvertent masking effect.

Fig. 3-173. Relative intensity profiles for the four test strips.

(a) 2500 Å

(b) 5000 Å

(c) 7500 Å

(d) 10 000 Å

Fig. 3-174. SEM pictures of microchannel holes in the four coated test strips.

thicknesses on the strips were 2500, 5000, 7500, and 10 000 Å. The corresponding dc resistances for each strip (measured across the 40-mm MCP) were 5.0, 2.1, 1.3, and 1.0 Ω , respectively.

The test plate was then installed in our 6-in., hard-sealed, 7-stripline intensifier. Time-domain-reflectometer (TDR) pictures showed essentially the same impedance (25 Ω) for each strip, independent of gold coating thickness. (The 7500-Å strip had a bad mismatch at its connection to the MCP, which caused considerable noise in the TDR trace. This problem has been corrected.)

We used a Garton spark source as an x-ray source for relative gain measurements on the four strips. The source output was filtered through a 0.5-mil (12- μ m) Mylar film coated with 2000 Å of aluminum to block ultraviolet light.

Visual inspection of the output fiber optics indicated a substantial difference in gain among the strips. Several shots were required to determine the correct exposure for the Kodak 2484 film. We

added a P-11, No. 127 wedge to the exposed film, which was then processed in a Hope 400 processor.

The film was digitized, and our VAX computer program, WEDGE, was used to convert film density to relative intensity (Fig. 3-173). The gain of the 2500-Å strip is a full order of magnitude greater than that of the 10 000-Å strip.

The MCP was then removed from the intensifier and mounted in a scanning electron microscope (SEM). The SEM photographs of the input surface (Fig. 3-174) show considerable differences between the strips at different coating thicknesses. The 12- μ m holes in the 2500-Å strip are circular; those in the 10 000-Å strip are egg-shaped. The change in shape and size of the microchannel entrances, while not drastic, apparently changes the electric field configuration enough to make a great difference in the gain of the channel.

Coating to a depth of 2500 Å or less would appear to be desirable because of the increased gain obtainable. Until we

know exactly how much gold is needed to ensure against signal loss, however, we are using 5000 Å of gold to coat all our pulsed-MCP diagnostics on Nova, which include the GXI, gated active crystal (GAX), 22X (MAX), and GAX II. The gain differences are obvious, and there is no degradation of the fast pulse.

Authors: G. D. Power, P. M. Bell, and J. D. Killenny

High-Speed, Gated X-Ray Imagers. In this article, we describe single-frame and multiframe, gated x-ray images with a time resolution as fast as 150 ps. Such systems are based on the gating of microchannel plates in a stripline configuration. Gating voltage comes from the avalanche breakdown of a reverse-biased p-n junction, producing high-power voltage pulses as short as 70 ps. We also describe results from single- and four-frame x-ray cameras used on Nova.

Gated x-ray cameras and spectrometers with gate times as short as 100 ps have aroused much recent interest. For ICF applications, this time resolution is necessary to freeze implosions with velocities of 10^7 cm/s and resolutions of $10\text{ }\mu\text{m}$. Two different techniques are available for gating: shuttering electro-optic tubes and voltage gating of a proximity-focused device. Several schemes have been proposed for subnanosecond shuttering of electro-optical tubes.^{309,310} Such approaches are attractive in that several frames can, in principle, be achieved,³¹⁰ there is little problem from x rays that

are transmitted through the cathode, and a uniform response can be obtained over a large photocathode area. However, the schemes are complex and, to date, have found little practical application.

Gating a proximity-focused device is much simpler. A voltage pulse is applied across either the photocathode-phosphor gap³¹¹ or across a microchannel plate.^{312,313} However, several disadvantages arise from this approach:

- A fast, high-voltage drive is required because a large detector area implies high capacitance.
- The device is inherently single-frame.
- Straight-through hard x rays can cause problems.
- The response across a large, sensitive area is nonuniform because of finite propagation velocity of the gating voltage wave.

Until recently, fast, high-voltage pulses for electrical gating could be generated only by photoconductive switches with the concomitant complexity of a short-pulse laser.³¹³ However, the well-known phenomenon of avalanche breakdown³¹⁴ has now been developed so that high-power electrical pulses as short as 70 ps can be produced by purely electronic drivers. We apply the voltage pulses to a strip transmission line with a microchannel plate as the dielectric. The temporal and spatial resolution are described later in this article. By using a voltage-doubling scheme, sufficient voltage from four parallel, reverse-biased diodes can be produced to gate four microchannel plates arranged in the four corners of a rectangle. The field uniformity and gain-width variation of this configuration are also described later in this article. For use on Nova, our microchannel plates are gated at different times and detect four-gated images from an array of four pinholes.

Four-Channel, Gated X-Ray Detector.

When a 1-kV, 1-A p-n junction is reverse-biased to a high voltage from a low-impedance source, it can undergo avalanche breakdown. In previously reported work,³¹⁴ the fall time of the diode impedance was 0.5 ns. By taking care to minimize inductances, we have been able to cut the voltage fall time to 70 ps. Moreover, by using a pulse-forming network, a

Fig. 3-175. Pulse shapes produced by various passive pulse-forming networks from a p-n junction.

Fig. 3-176. Four-gated detectors. The voltage pulse propagates along each microstrip line and doubles at the open-circuit end.

pulse instead of a step can be formed.³¹² These systems are pulse-charged by a 3-kV pulse with a 1-ns rise time, based on a microwave triode. The repetition rate, which is limited by the pulse charger, can be up to 1 kHz with negligible jitter in the avalanche-breakdown time of the pulse former.

A single diode can readily produce a 1.5-kV pulse into 50 Ω, thereby switching 160 kW. As shown in Fig. 3-175, we can produce a variety of pulse shapes by varying the pulse-forming network. We can obtain rise times of 70 ps, and the shortest pulse (not shown) can have a FWHM of 70 ps. To obtain x-ray gating, we use a 0.5-mm-thick MCP with $1/d = 40$ as the dielectric of a planar microstrip transmission line, with the amplified electron cloud at the MCP output accelerated onto a phosphor at +3 keV. The phosphor is on a fiber-optic faceplate with optical film in contact with the output of the faceplate.

We performed standardization on 12-Ω microstrip transmission lines coated on

▼ Fig. 3-177. MCP gain and gain width vs distance x away from open-circuit end.

the MCP because they can be driven without mismatch by two 25- Ω coaxial lines in parallel, and the 7-mm width on the MCP provides an adequate detector area. A single diode can switch enough power to provide a pulse for one such line. To obtain four images, we arranged four 12- Ω lines in the rectangular pattern

Fig. 3-178. Calculated and measured gain vs distance from open-circuit end of transmission line.

Fig. 3-179. Measured contrast transfer function.

shown in Fig. 3-176. To obtain sufficient switching power, four diodes are charged in parallel. By tuning the in-line inductance to each diode, the diodes can be made to avalanche simultaneously, all four pulses combining before being split again, as shown in Fig. 3-176. This technique ensures that the relative timing of all four microchannel plates, which is set by the difference in cable lengths, remains fixed. We measured the relative timing of gating pulses onto the MCP by a sampling oscilloscope to an accuracy of 50 ps. The blocking capacitor shown in Fig. 3-176 allows a dc bias to be applied to the plate.

X-ray images, which are aligned onto the open-circuit end of the strip lines coated onto the MCP, allow us to use the voltage doubling at an open circuit. This scheme has the advantage of effective pulse shortening for detector areas away from the open circuit, although gain decreases away from the open circuit in an accurately calculable manner. For detector positions displaced at distance x from the open circuit, incoming and reflected voltage pulses add with a time difference $\tau_d = 2x\epsilon_r^{1/2}/c$, where ϵ_r is relative permittivity of the transmission line, as shown in Fig. 3-177. Because MCP gain is a strong function of voltage ($\sim V^{11}$), once $\tau_d \approx \tau_{FWHM}$, the factor of 2 in the superposition of the two voltages causes a large decrease in gain. For the ideal case of a square voltage pulse, gain width is reduced for $0 < x < \tau_{FWHM}$, as shown in

Fig. 3-180. Schematic of four-frame, gated x-ray pinhole camera.

Fig. 3-177. This reduction is true if electron transit time in the MCP is small, i.e., $\tau_d < 100$ ps (Refs. 313 and 314). For our instrument on Nova, we used a 300-ps FWHM voltage pulse, which is well represented by a Gaussian curve, giving the calculated gain profile shown in Fig. 3-178. For this voltage profile, the gain narrowing illustrated for an ideal square voltage pulse in Fig. 3-177 does not occur.

We measured gain uniformity by exposing all four MCPs to a 150-ps x-ray flash from laser-produced gold plasma on Nova. With 250-ps interframe spacings, only frame 3 was gated on during the pulse, and gain uniformity with length was as shown in Fig. 3-178. Thus, we obtained good agreement with the simple calculation discussed above.

The effect on an image that is not large in the direction parallel to the stripline is small. The gain effectively falls to zero in ~ 15 mm for our 300-ps gating pulse. Because the width of the channel plate is 7 mm, there is $\sim 40\%$ edge-to-edge gain variation in a square image. The compressed core is usually much smaller for implosion work. If such gain variation were considered to be too detrimental, it could be removed by flaring out the width of the line at its open-circuit end.

Spatial resolution of a proximity-focused MCP device is usually set by electron spreading between the output of the channel plate and the phosphor. For our system, we measured the resolution with an opaque resolution grid with variable spacing in contact with the MCP, and we converted film density to exposure. Figure 3-179 shows the computed contrast transfer function. Even at 10 line pairs/mm, the contrast is 40%. Measurements with a finer-resolution grid show that features with $30\text{-}\mu\text{m}$ scalelengths can still be discerned visually.

We measured the absolute sensitivity of our MCP system by a calibrated electron-beam x-ray source. At an MCP bias of 900 V, an x-ray exposure of 0.3×10^8 photons/cm² was sufficient to produce an optical density of 1 at RAR2484 film in contact with phosphor and with photon noise of $\pm 20\%$.

Four-Frame, Gated X-Ray Imager on Nova. Figure 3-180 shows how four

composite x-ray pinholes image the x-ray-emitting plasma onto the four gated areas of the channel plate for use on Nova. Due to the large radius of the Nova target chamber, our system had to be run in on a rail system through a vacuum airlock. The pulse-forming modules are close (~ 20 cm) to the gated areas to minimize pulse-shape distortion due to propagation through the cable. The 1-ns rise-time pulse driver remains outside the vacuum chamber.

We observed no measurable jitter in the avalanche pulse-forming scheme; yet there are long-term timing changes due to variations in the trigger pulse of the driver pulse. As shown in Fig. 3-181, we monitor shot-to-shot variation in the timing of gating with respect to the laser pulse by displaying the fiducial laser pulse and the voltage pulse on a fast oscilloscope. Once timing has been established with a short flash of x rays, subsequent variations in the gating timing can be measured with an accuracy of 100 ps.

Results from Nova Experiments. The four-channel, gated x-ray imager is now in routine use on Nova. We have used the GXI for single-beam and multi-beam shots in high-neutron and electromagnetic-pulse environments. The most interesting picture comes from direct-drive implosions,

Fig. 3-181. Pulse-timing monitoring system.

where about 20 kJ of 0.35- μm laser light is incident on D-T-filled glass shells with 1-mm $\times$ 2.4- μm walls. The Nova system is not designed to optimize such implosions, and the rather poor implosions are used primarily as calibration neutron detectors. Figure 3-182 shows one such implosion for which the thermonuclear yield was 1×10^{13} neutrons. The five beams on the right-hand side of the image were timed to arrive 200 ps early; this side of the ball can clearly be seen as imploding faster on the frame at 0.5 ns.

The core of direct-drive implosions is nonuniform, as can be seen on the 1-ns frame; however, Nova is mostly used for indirectly driven implosions. Figure 3-183

Fig. 3-182. Direct-drive implosion with one array of beams arriving early.

Fig. 3-183. Images of the fuel regions of indirectly driven implosions: results of (a) asymmetric drive and (b) symmetric drive.

shows two shots in which the drive was tuned for asymmetry (a) and for symmetry (b).

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Gated X-Ray Spectrometer. During the past year, the field of x-ray spectroscopy on laser-produced plasmas has undergone what could be described as a revolution brought on by the somewhat routine availability of x-ray spectrographs that are both highly dispersive and time-resolved. We described the first generation of such a spectrograph, the gated crystal spectrometer (GCS) in Section 3 of the 1986 Laser Annual. This instrument has now been fielded scores of times on shots ranging in complexity from simple disks to imploding capsules located inside a hohlraum. The GCS has proven itself to be highly reliable (with a failure rate of less than 5%) and extremely versatile.

Figure 3-184 shows the GCS as it now exists. X rays from a source enter the array of entrance slits at the front of the spectrograph (left side of the figure). The x rays subsequently pass through a set of filters and baffles and are diffracted from the diffraction crystals. The diffracted x rays are incident upon microchannel

Fig. 3-184. Schematic layout of the gated crystal spectrometer.

Fig. 3-185. Sample spectra from a directly driven glass sphere.

plate striplines, which are gated on sequentially for 200 ps. The time interval between striplines is adjustable and typically is about 200 to 500 ps. The front slits serve the dual roles of background rejection and imaging. As shown in the figure, the two crystals are not necessarily the same, thus providing flexibility in that two distinct spectral regions may be

observed simultaneously, or, alternatively, additional filters may be added on one side to extend the dynamic range of a single spectral region. Because each stripline is only about 3.1 mm wide, and the magnification of this instrument is of the order of 10, only a single spatially resolved spectrum may be obtained at a time. When the spectrograph is used in

Fig. 3-186. Data from two similar shots consisting of a tightly focused beam 100 ps in duration incident on a gold slab.

(a) Nickel-like gold, M-shell fluorescence

(b) Thermal radiation

Fig. 3-187. Schematic layout of GCS II with a bent crystal.

(All dimensions in cm)

this mode, we plan to add a film cassette containing x-ray film in place of one of the baffles to thereby obtain a time-integrated spectrum along with time-resolved spectra. A time-integrated spectrum should be especially useful in obtaining absolute values for the brightness of measured spectra.

Our alignment scheme consists of a light bulb properly placed behind the slit assembly. The light source is used to backlight a central slit built into the slit assembly, which is viewed with a telescope located at the opposite wall of the chamber. We discovered empirically that the alignment could be accomplished to the high accuracy needed (about $50 \mu\text{m}$) using this very simple system. The entire alignment procedure takes only about 1 minute after the spectrograph is in place.

Figure 3-185 shows sample spectra from a directly driven glass sphere. Although our GCS was not a primary diagnostic on this series of experiments, the data show the type of information that can be obtained from the instrument and are remarkable in that they are almost noise-free even though this particular shot produced over 10^{12} neutrons. We have obtained such good noise rejection even for targets located inside a hohlraum. The resolution of these spectra is determined primarily by the source size and should therefore improve for high-convergence targets.

Figure 3-186 shows raw data taken from two similar shots, both of which consisted of a tightly focused beam 100 ps in duration incident on a gold slab. For both shots, we used the GCS in the one-dimensional imaging mode; however, we changed the crystal angle to give the spectral coverage shown. In

Fig. 3-186(a) the radiation is primarily from nickel-like gold, m-shell fluorescence; in Fig. 3-186(b) the radiation is primarily thermal in character. It is remarkable that, although the thermal radiation lasts only while the laser is on, the m-band fluorescence lasts for well over one nanosecond after the laser is turned off. It is also interesting to note that the spatial extent of x-ray emission is very narrow compared to the region emitting during the laser pulse.

The success of the GCS was so encouraging that another instrument has recently been built with somewhat different operating parameters. This instrument, which we call GCS II, was designed to compliment the original GCS in two ways. First, its magnification has been decreased. Therefore, GCS II may be used to image rather large targets fully on a single stripline. Second, the object distance has been decreased. This means that a given crystal can cover a larger spectral range.

Figure 3-187 shows the GCS II together with its bent crystal that was designed to give a spectral coverage of from 1.96 to

4.29 keV. The detector is identical to that of GCS so that no new technology needed to be developed. This spectrograph images horizontally, whereas GCS images vertically; in addition, we designed GCS II to operate in SIM III while not interfering with the line of sight in SIM IV (SIMs are 6-in. manipulators, standard 6-in. ports on Nova).

Author: R. S. Thoe

Pulse Generators and Pulse Shapers.

The gated MCP imagers described in the previous articles require fast, high-voltage electrical gating pulses. A typical gating pulse is 1000 V in amplitude into a gating strip having 6.25- Ω characteristic impedance and is from 100 to 400 ps in duration. We are using a gate generation system made up of two parts: a driver followed by a shaper.

The driver is a planar-triode-based pulse generator that produces a negative 3000-V pulse into 50 Ω with a rise time of less than 2 ns. We use two types of drivers: a commercial unit that has a single output and an LLNL-designed unit that has two simultaneous outputs. We use

(a) Shaping network

Fig. 3-188. (a) Shaping network for driving into a 6.25- Ω load. (b) A typical output pulse.

the latter unit when multiple imagers must be driven with low relative timing jitter.

The shaper can be described as a self-triggered, solid-state spark gap. It is based on fast avalanching of a diode junction when it is rapidly reverse-biased beyond breakdown.³¹⁵ To drive the 6.25- Ω characteristic impedance, we connect four diode sections in parallel, as shown in Fig. 3-188. The input pulse charges the 25- Ω coaxial charge lines CL1 to CL4 through the decoupling and wave-shaping inductors L1 to L4. When the voltage across diodes D1 to D4 exceeds that of avalanche breakdown, the diode resistance drops to a few ohms within 100 ps. The charge lines discharge through their respective diodes, resulting in a nominally square output pulse into a 25- Ω transmission line. The four lines are combined into a single 6.25- Ω transmission line made up of four, parallel 25- Ω cables. The diodes are 1N5408 rectifiers and are matched for similar breakdown voltages. Inductors L1 to L4 are short pieces of wire; the length of each is adjusted to cause the four diodes to switch simultaneously. Inductances L5 to L8 are small enough to short out the charging signal that is capacitively coupled through the diodes before breakdown, but large enough to pass the higher frequencies of the shaped pulse. They also sharpen the trailing edge of the output pulse. The result is a pulse with rise and fall times of 100 to 150 ps, jitter of less than 40 ps, and excellent long-term trigger stability (see the photo in Fig. 3-188).

These drivers and shapers are an integral part of three gated imaging systems on Nova. We plan to install several more systems in 1988.

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High-Resolution X-Ray Framing Camera. The achievement of high gain in a laser fusion implosion depends critically on the condition of the DT fuel near the time of maximum compression. Two parameters of particular interest are the final radius of convergence and the

symmetry of the imploded core, which can be determined directly and unambiguously through time-resolved imaging of the intense self-emission of x rays from the fuel pellet core. Time-resolved imaging is essential for suppressing potentially overwhelming x-ray emission from the hot corona and other sources throughout the implosion process. However, for the moderate size fuel pellets studied on Nova (initial diameter ~ 1 mm), the diameter of the imploded core is expected to be tens of microns and the duration of its confinement only ~ 100 ps, which will impose severe requirements on the spatial and temporal resolution of the imaging system. Accordingly, we have developed a high-resolution x-ray framing camera (HRXFC) that can record an x-ray image of ~ 1 cm² in area with a spatial resolution of less than 50 μ m at the image plane and a temporal resolution of less than 100 ps. Coupling the HRXFC to the 22 $\times$ Wöller x-ray microscope will provide resolution at the target of ~ 2 μ m with a field-of-view of $\sim 500 \times 500$ μ m². The HRXFC will be triggered optically with a laser pulse that is synchronous with the Nova driver beams.

The design of the HRXFC and the characteristics of its major components have been described in detail previously.³¹⁶⁻³¹⁹ A thorough discussion of the techniques used to fabricate the HRXFC can be found elsewhere.³²⁰ To review the principle of operation briefly, we show a schematic representation of the HRXFC in Fig. 3-189. Here an x-ray image is incident on a photocathode built into a suspended-strip transmission line, which, in the OFF state, has a reverse dc bias of ~ 700 V that prevents photoelectrons from leaving the surface of the photocathode. So that framed images can be recorded, a high-voltage gate pulse is generated with a gallium-arsenide photoconductive switch that is mounted directly in the transmission line and propagated across the photocathode. This photoconductive switch isolates a charge-line section that is pulse-biased to ~ 10 kV. The switch is activated with a short light pulse of moderate energy

Fig. 3-189. Schematic representation of the HRXFC.

Fig. 3-190. Experimental configuration for dynamic characterization of the HRXFC.

(~ 0.25 mJ) coupled in through a fiber-optic light guide, launching a ~ 4 -kV gate pulse of ~ 70 -ps duration. The pulse then propagates along the vacuum transmission line with negligible dispersion and is eventually dissipated in a thick-film resistor.

As the gate pulse traverses the photocathode, a photoelectron replica of the x-ray image is accelerated across the transmission line gap. The photoelectrons pass through a microchannel plate incorporated into the groundplane and are detected directly by a charge-coupled device (CCD). The microchannel plate is not electrically biased, but serves only as a direction-sensitive filter, effectively blocking the x rays transmitted by the semi-transparent photocathode. Excellent image quality is achieved in the proximity focus geometry by accelerating the

photoelectrons to many kilovolts while keeping the total transit distance short (~ 2 mm).

In the past year, we conducted laboratory tests to evaluate the performance of the first prototype HRXFC, using the short-pulse dye laser (pulse length $\tau = 1$ ps, wavelength $\lambda = 600$ nm) to both trigger the HRXFC and to generate an intense pulse of soft x rays. The dynamic test configuration is shown in Fig. 3-190. By focusing ~ 1 mJ of laser light onto a gold target, we were able to produce x-ray pulses of less than 10-ps duration at an energy of ~ 1 keV (Ref. 321) using a $1\text{-}\mu\text{m}$ -thick carbon foil to filter out ultraviolet radiation and protect the camera from target debris. The x rays backlit a linear zoneplate resolution pattern positioned directly in front of the photocathode, in these tests a $3\text{-}\mu\text{m}$ -thick

Fig. 3-191. (a) Linear zoneplate resolution pattern and (b) image lineout.

Fig. 3-192. Temporal response of the HRXFC at different dc bias voltages.

aluminum foil coated with 120 nm of cesium iodide. After passing through a variable delay line, a portion (1 mJ) of the laser pulse was used to trigger the HRXFC. Since the relative timing between the optical trigger and the x-ray pulse was free of jitter, we were able to improve the image contrast significantly by integrating over many laser shots.

Figure 3-191(a) shows a typical gated image of the resolution pattern, recorded with a pulsed bias voltage of ~ 10 kV and a dc bias of 500 V. The line width of the linear zoneplate pattern [a lineout of which appears in Fig. 3-191(b)] varied continuously from 500 to 50 μm , where the image contrast was found to be $\sim 10\%$. It should be recognized that the measured resolution represents the convolution of the actual photoelectron spread with the response of the CCD, which is limited by its 30- μm pixel size.

We determined the frame duration of the HRXFC by measuring the integrated

intensity of the recorded image as the relative delay between the x-ray pulse and optical trigger pulse was varied. Figure 3-192 shows the image intensity plotted as a function of the delay for dc bias voltages of 250, 500, and 700 V. Applying bias voltages of more than 700 V resulted in electrical breakdown. Since the duration of the x-ray pulse was negligible, the curves in Fig. 3-192 can be interpreted directly. It is evident that the shutter profile varied significantly with the dc bias voltage. In particular, the profile exhibited a long tail at 250 V, which was suppressed as the bias voltage increased. Consequently, the frame duration decreased with increasing dc bias, having values of 200, 80, and 50 ps (FWHM) at bias voltages of 250, 500 and 700 V, respectively.

The long tail of the shutter profile at low bias voltage indicated there were photoelectrons in the transmission line gap hundreds of picoseconds after the arrival of the x-ray pulse. In addition, we observed that images recorded at the long delay times exhibited substantially degraded spatial resolution. We believe that the long tail corresponds to photoelectrons emitted at the surface and within the channels of the microchannel plate. These photoelectrons spread transversely while drifting toward the photocathode in the dc bias field and are eventually accelerated back through the microchannel plate by the high-voltage gate pulse or are absorbed at the photocathode. When bias voltage is increased, the drift time of the photoelectrons in

the transmission line gap decreases, thereby resulting in a reduced signal at long delay times. A quantitative analysis of the variation in shutter profile with bias voltage is in progress. It is evident, however, that the shortest frame duration and the best spatial resolution are achieved by operating the HRXFC at maximum dc bias voltage.

We have also measured the shutter efficiency of the HRXFC, defined as the ratio of the image intensity to the residual intensity recorded when the camera is not triggered. The measurements showed the residual intensity obtained by integrating over 200 laser shots with the optical trigger disabled was less than the background noise of the CCD readout. When this result was compared to the image intensity measured with the optical trigger enabled, the ratio implied a shutter efficiency in excess of 1000.

The characterization of the HRXFC is complete, and it has been shown to satisfy important performance specifications: a frame duration of 50 ps, spatial resolution of $50\text{ }\mu\text{m}$, and a shutter efficiency greater than 1000. Now a second prototype HRXFC is being designed and fabricated specifically for implementation on the Nova target chamber. The instrument package will be compatible with the output stage of the $22\times$ Wölter x-ray microscope. The first laser experiments using the HRXFC will be conducted on the Short Pulse Facility within the next year. We expect to implement it on the Nova laser in FY 89.

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X-Ray Optics and Technology.

Characterization of a High-Magnification X-Ray Microscope on Nova.

We have implemented a Wölter-type x-ray optic in a high-magnification ($22\times$) microscope on Nova. We have measured the point-spread function, contrast transfer function at selected spatial frequencies, x-ray throughput, and alignment accuracy, all as configured for Nova experiments.

To measure alignment accuracy, and precision, we constructed a series of

resolution grids backlit by illuminating a dysprosium backlighter disk with one beam of the Nova laser. Our scanning-electron microscope grids have reference letters every $300\text{ }\mu\text{m}$. Figure 3-193 shows a typical image from such a grid. After measuring four such images with careful alignment, $\pm 10\text{ }\mu\text{m}$ in both vertical and horizontal directions, we found a $30\text{-}\mu\text{m}$ offset in the vertical and no offset in the horizontal direction.

During measurement of these targets, we observed that the edge response was

Fig. 3-193. Image of a backlit resolution grid with image aligned optically to cross hairs.

Vertical offset = $30 \pm 10\text{ }\mu\text{m}$
Horizontal offset = $0 \pm 10\text{ }\mu\text{m}$

Fig. 3-194. Result of measurement of tilted resolution grid.

not as sharp as expected ($8\text{-}\mu\text{m}$, 10-to-90% rise compared with $3\text{-}\mu\text{m}$, as measured off line). We therefore included a measurement of the z-axis offset, i.e., focusing offset, in this series of characterization measurements. To measure the z-axis offset, we constructed a resolution target that was rotated around its vertical axis. When using the entire Wölter optic instead of a small sector, the reduced depth of field causes the resulting image to vary rapidly across the 1-mm target. The resulting vertical contrast should be at a maximum at the correct focal point, as shown in Fig. 3-194. (The improvement in contrast near the edges occurs because the image near the edges is formed mainly by a small vertical sector.) The measured offset between the optical-image best focus and the x-ray-image best focus is $500\text{ }\mu\text{m}$.

We measured the point-spread function for a 5-mm sector of the optic by backlighting a pinhole array with dysprosium, as described above. Figure 3-195 shows the resulting image. The visible spots are formed by 40-, 20-, 10-, and $5\text{-}\mu\text{m}$ pinholes. The FWHM of the $5\text{-}\mu\text{m}$ images averaged about $6.7\text{ }\mu\text{m}$ in the horizontal direction and about $10.6\text{ }\mu\text{m}$ in the vertical direction.

We measured the contrast transfer function for the same 5-mm sector of the

optic by backlighting a resolution grid with a $50\text{-}\mu\text{m}$ -wavelength grid ($25\text{-}\mu\text{m}$ open spaces and $25\text{-}\mu\text{m}$ bars). Figure 3-196 shows the resulting image. The horizontal contrast averaged across the image is about 0.45; the vertical contrast is about 0.30.

X-ray throughput can be estimated from an image on film. Comparison of such an image to the static-spectrometer (Henway) spectrum of the backlighter, corrected for filtration, solid angle, protective screen, and sector size, suggests that at 1.5 to 2.0 keV, about 30% of the anticipated signal is found in the image. This value depends strongly on accurate knowledge of the density vs log exposure curve for spectrometer film and microscope film. Such curves are known to about 40% and may introduce a large error in measurement.

Author: S. G. Glendinning

Demonstration of Guided-Wave Phenomena at Extreme Ultraviolet and Soft X-Ray Wavelengths. We have performed the first explicit demonstration of classical guided-wave propagation at extreme ultraviolet (XUV) and soft x-ray wavelengths. The practical value of guided-wave transmission has long been appreciated, and considerable effort has gone

Fig. 3-195. Point-spread function from backlit pinhole array.

Fig. 3-196. Contrast transfer function at $v = 10$ line pairs/mm input.

into the development and optimization of classical waveguide structures for much of the electromagnetic spectrum. In general, the classical waveguide is a long, narrow structure within which geometrical or ray optics do not provide a viable approximation to electromagnetic wave propagation. As intended here, a classical waveguide structure is one for which $a/\lambda > 1$ and $L/\lambda \gg 1$, where λ is the wavelength being propagated, a is a characteristic transverse dimension of the waveguide (e.g., diameter), and L is its length. (In the notation of Fig. 3-197, $a \leftarrow s$ and $L \leftarrow t$.)

Until now, classical waveguide structures for XUV and soft x radiation have not received much attention for at least two reasons. First, only recently has the technology been available for fabricating structures with dimensions on the scale of the soft x-ray wavelength.³²² Second, the general sense of the x-ray community is that propagation losses would be prohibitive at wavelengths longer than a few tenths of a nanometer.^{323,324} As a result, the majority of guided x-ray research has been on x-ray "light pipes" (having $a \gg \lambda$) (Ref. 325) that rely on multiple

internal reflections of "hard" ($\lambda \approx 0.1$ nm) x rays from the interior surfaces of macroscopic capillary tubes ($a \approx 10^4$ to 10^5 nm) (Refs. 326-329).

The waveguide propagation experiments reported here were conducted in a range of parameter space previously unexplored for XUV and soft x rays: $a/\lambda \approx 3$ to 20, for $\lambda \approx 5$ to 30 nm, with $L/\lambda \leq 140$. In these experiments synchrotron

Fig. 3-197. Experimental setup for the measurement of the diffracted synchrotron beam passing through the thick grating (waveguide) structures.

radiation was propagated down the long, narrow channels of a free-standing, sub-micron-period transmission grating. The output diffraction pattern was observed as a function of tilt angle between the grating normal and the incident beam direction. In general, grating tilt produced strongly asymmetric diffraction patterns, allowing directional control over the flow of diffracted x-ray energy. Distinct diffraction patterns for the transmitted radiation were observed even at very large tilt angles, for which the beam had no

Fig. 3-198. Measured diffraction patterns at $\lambda = 20.8$ nm emerging from grating G-1 ($l = 100$ nm, $s = 200$ nm, $p = 300$ nm, $t = 470$ nm) at a variety of tilt angles (θ_T). The angular separation between diffracted orders is $\Delta\theta = \lambda/p$.

geometric line of sight (LOS) through the grating channels. Although measured propagation efficiencies were modest ($<42\%$), novel waveguide designs are herein proposed to significantly improve propagation efficiency.

Experimental Detail. The x-ray waveguide experiments were conducted at the radiometric laboratory of the PTB Institut Berlin at the Berlin Electron Storage Ring (BESSY).^{330,331} Measurements were conducted at four wavelengths: 5, 20.8, 21, and 30 nm. In each case, the synchrotron radiation incident on the waveguide structure was narrowband ($\lambda/\Delta\lambda \approx 500$), linearly polarized (along the x direction in Fig. 3-197), and collimated into a narrow beam ($\approx 200\text{-}\mu\text{m}$ diam with 0.25-mrad divergence).

The waveguide structures were free-standing (no substrate or support membrane) transmission gratings with a 300-nm grating period.^{332,333} Results from two gratings (G-1 and G-5) are reported here. G-1 had a 100-nm line width, a 200-nm space width, and a 470-nm thickness (see Fig. 3-197). G-5 had a 200-nm line width, 100-nm space width, and a 700-nm thickness. The grating lines were gold and the spaces vacuum. The lines (and spaces) were aligned along the vertical (the y direction in Fig. 3-197) and were supported by a $6\text{-}\mu\text{m}$ -period, orthogonal array of gold cross bars (aligned along x , but not shown).

Figure 3-197 shows the experimental setup. The synchrotron beam was incident along the $+z$ direction, with its E-field polarized parallel to the x axis. At zero tilt angle, the grating's lines were aligned along y , its periodic variation along x , and its thickness along z . The grating tilt axis M passed through the grating and was parallel to the y axis. A channeltron detector operating in the photon counting mode was used to measure the pattern of diffracted radiation emerging from the grating structure. The detector moved along a circular path in the dispersion plane (x - z plane in Fig. 3-197). (Note that the grating and detector are not coupled in a θ , 2θ motion. The grating is fixed at its tilt angle while the detector scans the dispersion plane.) The circular detection path had a 20-cm radius and was centered on the grating tilt axis.

Data Presentation and Analysis. In the following paragraphs, waveguide propagation phenomena are illustrated by diffraction pattern asymmetries resulting from changes in the transmission grating tilt angle and/or the incident x-ray wavelength. The asymmetries arise from variations in the phase and amplitude distribution across the output channels of the transmission grating. These variations are produced by changes in the propagating mode structure as grating tilt angle and/or incident x-ray wavelength are changed.

Figure 3-198 shows a representative data record illustrating the dependence of diffraction pattern asymmetry on grating tilt angle. The data are for $\lambda = 20.8$ nm radiation incident on transmission grating G-1. Plotted are values of relative diffraction efficiency, into 0, ± 1 , ± 2 orders for grating tilt angles varying from -6 to $+6^\circ$. It is important to note that in these measurements the absolute value of diffraction efficiency is determined primarily by external geometrical factors not germane to the issues of waveguide propagation. We are concerned instead with relative changes in diffraction efficiency as relevant parameters are varied.

In the data of Figs. 3-198 through 3-200 the zeroth order of the diffraction patterns is coincident with the incident beam direction. The measured angular separations between adjacent diffracted orders is, as expected, $\Delta\theta = \lambda/p$, where λ is the x-ray wavelength and p is the grating period. The measured values of diffraction efficiency presented in the data are not a direct indicator of waveguide mode propagation efficiency. The diffraction efficiency values are determined primarily by the throughput of the grating support structure. For example, in the $\theta_T = 0^\circ$ data of Fig. 3-198 when the grating support structure is taken into account, we find that 42% of the x-ray energy incident on the open grating channels appears in the 0, ± 1 , ± 2 orders of the diffraction pattern.

The measured diffraction patterns of Fig. 3-198 show a distinct asymmetry, with energy preferentially directed into those orders toward which the grating is tilted. As tilt angle is increased, the asymmetry of the diffraction pattern increases (with some loss of overall

diffraction efficiency), providing some directional control over the flow of diffracted x-ray energy. Another application of these phenomena relates to the use of thick transmission gratings as dispersive elements in x-ray spectroscopy,^{334,335} wherein the dependence of diffraction efficiency on tilt angle and x-ray wavelength may play an important role in instrument calibration.

Fig. 3-199. Measured diffraction patterns emerging from grating G-5 ($l = 200$ nm, $s = 100$ nm, $p = 300$ nm, $t = 700$ nm), tilted 6° toward the positive diffraction orders for a variety of incident wavelengths. $\Delta\theta = \lambda/p$.

Fig. 3-200. Measured diffraction patterns at $\lambda = 30$ nm emerging from grating G-5 tilted at angles θ_T greater than the cutoff for a geometrical LOS through the grating channels by the incident synchrotron beam. $\Delta\theta = \lambda/p$.

Figure 3-199 shows a data set illustrating the wavelength dependence of diffraction pattern asymmetry and waveguide propagation efficiency. The data are for radiation at $\lambda = 5, 21$, and 30 nm incident on grating G-5, tilted at 6° toward the positive diffraction orders. Two things happen as wavelength is increased. First, the asymmetry of the diffraction pattern becomes more pronounced, indicative of the changing phase distribution within the waveguide channels as the wavelength dependent mode structure is changed. Second, the overall diffraction efficiency of the grating structure is decreased because of increased wall losses (greater gold absorption) at the longer wavelengths.

Figure 3-200 shows transmission of soft x radiation through the grating structure at extreme tilt angles for which the beam had no geometric LOS through the grating channels. Synchrotron radiation at $\lambda = 30$ nm is incident on grating G-5 at tilt angles of 12 and 18° . The cutoff of the geometric LOS through the channels of G-5 occurs at a tilt angle of approximately 8° . As noticed earlier, diffraction pattern asymmetry continues to increase and diffraction efficiency continues to decrease with increasing tilt angle, but there is no precipitous change in waveguide propagation characteristics beyond the geometric cutoff angle. One may compare, for example, the $\lambda = 30$ nm data (at 6° -tilt angle) of Fig. 3-199 with the data of Fig. 3-200.

A quantitative analysis of the diffraction patterns produced by the thick transmission grating structures under discussion is in preparation for publication. In this classical analysis, a proper accounting is made of the interaction of the soft x radiation with the three-dimensional, periodic structure. The radiation propagates down the waveguide channels and emerges with an amplitude and phase distribution (within each channel) determined by the geometry and optical characteristics of the channel walls. Changes in wavelength and/or tilt angle significantly alter the propagating mode structure within the waveguide channels leading to complex phase and amplitude distributions emerging from

the periodic waveguide structure. The variations in these amplitude and phase distributions produce the wavelength and tilt angle dependent modulations of the diffraction patterns observed in Figs. 3-198 through 3-200.

In contrast to an early sense of pessimism within the field,^{323,324} we have explicitly demonstrated the propagation of soft x radiation through classical waveguide structures. This work is of immediate and direct relevance to researchers using thick, periodic structures for spectroscopic^{334,335} and/or x-ray imaging (e.g., "jelly roll" zone plates)³³⁶ applications. In order for this technology to play an effective role in the control and transport of soft x radiation, waveguide efficiency must be significantly improved. This can be achieved through the use of resonant multilayer structures as optimal wall materials in submicron-period vacuum waveguides. A detailed discussion of optimized, soft x-ray waveguide design is in preparation for publication in *Applied Optics*.

Author: N. M. Ceglio

High-Precision Alignment of X-Ray-Transmission Diffraction Gratings.

Gold transmission diffraction gratings have been coupled with other x-ray diagnostic instruments (such as microscopes, telescopes, and temporally resolved detectors)³³⁷⁻³³⁹ to make sophisticated instruments with expanded capabilities. Coupling gratings to such diagnostic instruments occasionally requires a high degree of accuracy in rotational alignment of the grating normal to a specific axis of the instrument.

In the past, it has been relatively easy to rotationally align a grating with the axis of a second instrument³⁴⁰ because grating periods have been large (>1000 nm), and optical imaging or diffraction techniques could be used. Submilliradian rotational alignment can be achieved by optical diffraction. However, high-line-density transmission diffraction gratings with periods ranging from 200 to 300 nm are now being used for diagnostic measurements on Nova.

The principal difficulty with alignment of these diffraction gratings is the absolute determination of the direction of grating lines. The grating period is too small for optical diffraction and imaging techniques, and polarization techniques³⁴¹ are cumbersome and limited to milliradian accuracies. In addition, the alignment of a grating often cannot be easily verified on the Nova target chamber. We have developed a simple technique that can determine the grating direction to within a few microradians and that allows the alignment of the grating to be easily verified in situ using conventional optical techniques.

Procedures and Results. Thick, gold diffraction gratings were fabricated on silicon wafers by a combination of several techniques, including x-ray lithography and holographic lithography. These gratings have spatial periods of 200 to 300 nm, gold thicknesses ranging from 100 to 500 nm, and are described in greater detail elsewhere.^{342,343} The gold gratings were overcoated with a thin layer (<250 nm) of PMMA, which is a high-resolution, electron-beam-sensitive resist. Thicker layers of PMMA were not as successful because the ability of a scanning electron microscope (SEM) to image the gold grating through PMMA is essential to this technique. For layers of PMMA thicker than 500 nm and for incident electron-beam energies less than 20 keV, lateral spreading in the beam due to forward scattering³⁴⁴ degraded the resolution so that the gold grating could not be imaged.

We modified the stage of a Bausch and Lomb Nanolab LE2100 SEM so that the 2-axis stage motion was motorized and controlled by a joystick. By appropriately configuring the SEM stage and the stepping-motor controller, we were able to obtain a minimum motion of approximately 32 nm per step. This minimum step size represents a small fraction of a grating period and allowed us to move the stage accurately relative to a single grating line. (The gold grating on the silicon wafer was mounted on the SEM stage.) The stepper motor controller was programmed to rotate at a constant, but programmable, frequency (steps per second) that determined the lateral stage

translation speed, and to travel in the direction dictated by the joystick. The operator imaged any grating line in the SEM and moved the stage along the direction of the grating line, independent of the relative orientation of the stage to the grating. Our gold grating lines were typically ~100-nm wide, and we followed a single line for 1 cm with an accuracy of ± 0.5 linewidth. This value translates to an SEM line written parallel to the original grating with an accuracy of $\pm 5 \mu\text{rad}$.

After exposure, the SEM-written line was developed for 1 min in a mixture of 50% methyl-isobutyl-ketone and isopropyl alcohol. The SEM-written line had a line width much greater than the area directly exposed to the electron beam, which was typically 2000 to 5000 nm. The larger line width arises because PMMA was exposed to the incident electron beam in the SEM and to electrons backscattered from the gold grating and silicon substrate (see Fig. 3-201). The resultant line width was ~0.01 mm; however, the exact width of the line was dependent on incident energy of the electron beam, beam current, backscattered electron range, and speed of the stage translation. (We used a 15-keV electron beam, and stage translation speeds ranged from 0.05 to 0.25 mm/min.)

Fig. 3-201. (Top) A thin PMMA layer overcoats a sub-micron-period gold grating and is exposed to an electron beam. (Bottom) Energy deposited in PMMA has a spatial profile determined by forward- and back-scattered electrons. An area wider than the SEM electron-beam width receives appreciable dosage.

Fig. 3-202. (Top) After exposure, the developed area in PMMA is that exposed to the electron beam. (Bottom) After overexposure, the area exposed to the electron beam is insoluble in the developer. Areas surrounding the SEM-exposed area receive less exposure through backscattered electrons, resulting in two exposed and developed lines separated by several microns.

Fig. 3-203. Two gold-plated parallel lines (alignment markers) produced by SEM-overexposure. Overexposed PMMA in the center was not removed, making the submicron-period grating in that region difficult to image.

Because some of these variables can change during exposure, the exact width of the SEM-written line varied; nevertheless, the center of the line was always aligned to the grating.

The written line was made a permanent part of the grating by either gold microplating or ion-beam etching to transfer the pattern from PMMA into the grating. The next step was to remove PMMA by soaking in chlorobenzene and exposing to a UV ozone cleaning process. Because it is a permanent addition to the grating, the SEM-written line can be used at any time to optically align or verify alignment of the grating.

Under some conditions, the SEM-writing process resulted in two parallel lines in the PMMA. These two lines were each approximately 5000-nm wide, separated

by another 5000 nm because heavily exposed PMMA behaves as a negative resist. Under slow translation speeds or high beam currents, the area exposed to the SEM beam was heavily exposed; however, areas immediately surrounding the region were exposed only to backscattered electrons (see Fig. 3-202). The heavily exposed area in the center was insoluble in the developer, but areas exposed to only the backscattered electrons were less exposed and developed normally. The result is two, parallel lines separated by the heavily exposed PMMA. As an alignment mark, the parallel lines are functionally equivalent to a single SEM-written line. Further processing for parallel lines was identical to that for a single line. Figure 3-203 shows two, parallel lines produced by an overexposure and gold microplated.

Final processing of the gold grating with the optical-observable alignment line depended on the application of the gold grating. For spectroscopy of x-ray energies greater than 1 keV, or for x-ray lithography, the grating can be supported by a thin membrane, such as 500 to 1000 nm of polyimide, which offers minimal x-ray absorption at these energies. Under such conditions, the silicon wafer can be etched away, leaving a gold grating on the support membrane. For x-ray energies less than 1 keV, energy-dependent x-ray absorption by the support membrane severely complicates analysis of a

spectrum. Therefore, we made freestanding transmission diffraction gratings supported by a gold grid using conventional photolithographic processes and gold microplating.^{342,343}

A typical grid consists of a 0.01-mm-period grating (0.003-mm-wide gold bars) orthogonal to a 0.25-mm-period grating (0.025-mm-wide gold bars). Using a mask aligner, we aligned the grid pattern to the SEM-written line. After exposure, development, and gold microplating, the resulting structure is a periodic array of optically observable gold lines that are parallel and perpendicular to the submicron-period grating (see Fig.3-204), any of which can be used for instrument alignment. Final processing of our freestanding gratings required etching of the silicon wafers, resulting in the desired freestanding grating over a 7- × 10-mm area. We have now used these gratings successfully on several experiments at LLNL.

Alignment of the freestanding grating to the support grid was tested against a polarization measurement. Our polarization tests measured the transmission of polarized light through the grating as a function of grating rotation. The accuracy of our polarization tests was ± 5 mrad. Within this limitation, we could distinguish no discrepancy between the orientation of the grating lines, as determined by either polarization tests or the SEM-writing procedure.

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Fabrication of Soft X-Ray-Laser Cavity Components.

Introduction. Recent experiments have demonstrated the production of ASE at soft x-ray wavelengths.³⁴⁵⁻³⁴⁷ The reliable production of significant amplification at soft x-ray wavelengths provides strong motivation for normal-incidence mirrors and output couplers for use in an x-ray-laser cavity. Such a cavity could provide significant enhancement of x-ray-laser emission and possible single, transverse-mode operation, which is useful for many future applications.

Fig.3-204. Gold-plated SEM-written line used as an alignment marker to rotationally align a support grid to the 300-nm-period grating.

Fig. 3-205. Fabrication process for the x-ray beam splitter.

Fortunately, recent advances in the field of microfabrication technology have spurred a revolution in the field of x-ray optics. Transmission diffraction gratings and Fresnel zone plates have been made³⁴⁸⁻³⁵¹ with minimum feature sizes of less than 100 nm, and multilayer mirrors for high normal-incidence x-ray reflections ($R > 25\%$) for wavelengths as short as ~ 5 nm have also been reported.³⁵²⁻³⁵⁶

We have developed two new x-ray optical components: an x-ray beam splitter and a highly dispersive multilayer mirror (HDMM). Both structures can be used as efficient output couplers for an x-ray-laser cavity. The x-ray beam splitter reflects and transmits some fraction of the incident x radiation, while the HDMM reflects and diffracts normal-incident radiation. In both cases, the reflected beam provides the necessary feedback into

the laser cavity, while transmitted and diffracted beams provide the output coupling.

Fabrication. Figure 3-205 outlines the fabrication of our x-ray beam splitter via a multistep process using many of the recent advances in the field of microfabrication. A silicon wafer is first prepared by a thorough cleaning process, and an etch stop (a 30-nm layer of silicon nitride or boron nitride) is deposited by low-pressure chemical vapor deposition. It is important that this layer be x-ray transparent because x rays transmitted through the beam splitter will pass through this thin layer. Therefore, the layer, which covers both the front and back sides of the silicon wafer, is both thin and composed of low-Z materials.

Next, the multilayer mirror is deposited onto the front side of the silicon wafer by one of several different techniques. Our multilayer mirrors are deposited by a dual-head, magnetron sputter-deposition system with a revolving wafer holder. The number of layers, chosen to give a reasonable reflection and transmission coefficient, depends on the x-ray wavelength and mirror materials. We typically use approximately 10 layer pairs of molybdenum and silicon multilayers for use at x-ray wavelengths from 13 to 30 nm. The multilayer mirrors were overcoated by ~ 1 nm of carbon to protect them from oxidation. The back side of the silicon wafer was patterned using conventional lithographic techniques so that the thin etch-stop layer was removed from a small area (up to 5×15 mm) to expose the underlying

silicon. This silicon was then fully etched from the back to the thin etch-stop layer on the front surface by using a highly selective anisotropic etching bath (500 g KOH in 1 L of water at 70°C from Ref. 356). This bath etches the (100) and (110) planes in the silicon crystal at approximately 500 nm/min, but etches less than 5 nm of silicon nitride in the 8 to 10 hours necessary to etch through the silicon wafer. Unfortunately, the KOH etch bath attacks the multilayer and completely destroys it within a few minutes. A special teflon fixture with a double O-ring seal was machined to protect the multilayer surface from the etch bath.

Our fabrication of amplitude-modulated HDMMs also involved advanced fabrication techniques. Amplitude-modulated HDMMs require the lateral modulation of the reflection coefficient of an x-ray multilayer mirror. This requirement can be met by several techniques (see Fig. 3-206), such as patterning a thick gold grating on the surface of the multilayer, etching a grating pattern into the multilayer, or depositing a multilayer onto a previously patterned substrate. The resultant structure has regions of high ($>10\%$) reflection relative to regions of low ($<1\%$) reflection. Normal-incidence reflection of soft x rays from a bulk material, such as gold, is much less than 1%.

Silicon-wafer substrates were first cleaned and then deposited with a molybdenum/silicon multilayer mirror using the dual-head magnetron sputter-deposition system described earlier. We typically used 25 layer pairs of molybdenum and silicon with a d spacing of ~ 11.5 nm, followed by a 1-nm overcoat of carbon. Next, 5 to 10 nm of chromium and 50 to 100 nm of gold were thermally evaporated onto the multilayer mirror surface, followed by a 500-nm spin coating of an AZ resist onto the gold. For gratings with periods greater than $\sim 3 \mu\text{m}$, we used optical-contact lithography to pattern the photoresist and a wet chemical etch to etch the gold. For submicron-period gratings, we used x-ray lithography, holographically produced masks to pattern PMMA, and a dry etch (argon-ion-beam milling) to transfer the pattern to gold.

Fig. 3-206. An amplitude-modulated multilayer mirror is fabricated by preparing a silicon wafer, coating it with an x-ray multilayer mirror, and patterning with an x-ray absorber (e.g., gold) or etching.

(1) Prepare substrate

(2) Deposit multilayer mirror

(3) Pattern

Fig. 3-207. Two examples of imperfections during fabrication. Sample M-2 had a thin layer of chromium overcoating the multilayer. Sample M-21 was overetched, damaging a few layers of the multilayer mirror.

- r = mirror reflection coefficient (E -field)
- ϕ_m = phase shift on reflection
- p = pattern period
- t = pattern thickness
- a = pattern attenuation per unit length
- ϕ_a = pattern phase shift per unit length

Fig. 3-208. The HDMM is modeled as having regions of high vs low reflectivity. The reflected electric-field pattern is analyzed by Fraunhofer diffraction analysis and Fourier techniques.

As shown in Fig. 3-207, imperfections in the fabrication process led to imperfect HDMMs. For example, wet chemical etching could not be used to remove the thin layer of chrome without damaging the multilayer (liquid chrome etches attack molybdenum). Larger-period HDMMs therefore had a thin layer of chromium overcoating the multilayer. Argon-ion-beam milling removed this thin chromium layer on our submicron-period HDMMs, but overetching can lead to damage on the underlying multilayer mirror. Both of these imperfections result in degraded performance of the HDMM. We are developing improved fabrication processes to obviate such problems in future HDMMs.

HDMM Modeling. Performance of an HDMM is modeled by analyzing the reflected electric-field pattern directly above the surface of the structure (see Fig. 3-208). Incident radiation is reflected at near-normal incidence. In regions where no gold lines exist (grating "spaces"), the E field is reflected with a reflection coefficient r and phase angle

ϕ_M . In regions of gold lines on the multilayer mirror, the E field is reflected with a coefficient b and phase angle ϕ_A (see Fig. 3-209). The reflection coefficient b (in Fig. 3-209, $b = 2art$) and phase angle ϕ_A depend strongly on the normal-incidence reflection of, and transmission through, gold. Using the Fresnel equation, for x rays of interest and for our gold thicknesses, the value of b is small (<0.1), whereas the value of r can be quite large (>0.5).

We can obtain coefficients for the various diffracted orders for a structure by taking the Fourier transform of the electric-field pattern. Specifically, the electric field can be broken down to its Fourier components^{357,358}

$$E(x, z) = \sum_n B_n \exp(j\vec{k}_x \cdot \vec{x}) \exp(j\vec{k}_z \cdot \vec{z}), \quad (63)$$

where $\vec{z}$ is the vector direction normal to the surface of the HDMM, $\vec{x}$ is the vector direction in the plane of the HDMM and normal to the grating lines, $|\vec{k}_x| = 2\pi n/p$,

p is the grating period, $|\vec{k}_z| = (k_0^2 - k_x^2)^{1/2}$, and $|\vec{k}_0| = 2\pi/\lambda$. The term B_n represents the Fourier components and electric-field amplitudes of diffracted orders. The orders will diffract at angles given by

$$\sin \theta_n = \frac{|\vec{k}_x|}{|\vec{k}_0|} = n \frac{\lambda}{p} \quad (64)$$

Because the electric field is periodic, B_n can be calculated by taking the Fourier transform of the electric-field pattern over one period at any convenient plane, such as at $z = 0$, as follows:

$$B_n = \frac{1}{p} \int_{-p/2}^{p/2} E(x,0) \exp(-jn2\pi \frac{x}{p}) dx \quad (65)$$

Substituting the electric-field pattern described in Fig. 3-209 yields

$$B_0 = r \left(\frac{2}{\beta} \right) + b \exp(j\Delta\phi) \left(1 - \frac{2}{\beta} \right) \quad (66)$$

and

$$B_n = \frac{\sin 2n\pi/\beta}{n\pi} (r - b) \exp(j\Delta\phi), \quad (67)$$

where

$$\Delta\phi = \phi_M - \phi_A, \quad (68)$$

and β is defined in Fig. 3-209. The term β is a variable that describes the ratio between the grating line and space. Specifically, when $\beta = 4$, the line-to-space ratio is 1:1.

Note that $\Delta\phi$ is wavelength-dependent because of different x-ray path lengths in reflections from grating lines vs grating spaces, making the diffraction efficiency wavelength-dependent. It is also important to note two additional features. First, the important phase information is the relative phase shift between "lines" and "spaces" of the grating. Second, when $b \ll r$, the diffraction efficiency becomes

Fig. 3-209. HDMM modeling. The amplitude and phase front of the electric field reflected from the surface of the HDMM is periodic. β is a constant determined by the line-to-space ratio.

Fig. 3-210. Experimental characterization of M-2, a 6- μ m-period gold grating with $d = 11.5$ nm. Note modulation in higher diffracted orders as wavelength is scanned.

independent of the relative phase shift, and the diffracted electric-field components simply become

$$B_0 = r \left(\frac{2}{\beta} \right) \quad (69)$$

and

$$B_n = \frac{r}{n\pi} \sin \left(\frac{2n\pi}{\beta} \right), \quad (70)$$

and the intensity pattern becomes

$$I_0 = R \left(\frac{2}{\beta} \right)^2 \quad (71)$$

and

$$I_n = \frac{R \sin^2(2n\pi/\beta)}{(n\pi)^2}, \quad (72)$$

where $R = r^2$.

HDMM Characterization. We performed x-ray characterization of our devices in collaboration with members of the Physikalisch-Technische Bundesanstalt at the BESSY synchrotron facilities.³⁵⁹ Monochromatic radiation was incident at near-normal incidence onto the HDMM, and the diffracted beams were measured by scanning (in angle) a channeltron detector. Absolute reflectivity into the zero order and higher orders was measured as a function of wavelength for several grating periods, ranging from 200 nm to 6 μm (see Figs. 3-210 and 3-211).

For the most part, the HDMMs performed as expected. The diffracted orders appear at the expected angles ($\sin \theta = m\lambda/p$), and the relative efficiency between orders (i.e., the ratio between the zero order and higher diffracted orders) is approximately as predicted by our model. However, the absolute diffraction efficiency is reduced considerably from calculated values. This result may be due to imperfections introduced into the HDMMs during the fabrication process, as discussed above. In addition, fine details in the absolute diffraction efficiency in the relative orders cannot be accounted for by our model. For instance, in Fig. 3-210, the diffraction efficiency into the first order seems to oscillate as a function of wavelength; in

Fig. 3-211. Experimental characterization of M-21, a 200-nm-period gold grating with $d = 11.5$ nm. Note variation in diffraction efficiency for higher diffracted orders as wavelength is scanned.

Fig. 3-211, the relative efficiencies in the second and third diffracted orders of a 200-nm-period gold grating on a multilayer also vary as a function of wavelength. However, these effects are small.

Tables 3-22 and 3-23 summarize the measured performance of two HDMMs that were characterized at the BESSY synchrotron (see Figs. 3-210 and 3-211). Assuming total absorption in, and little normal-incidence reflection from, the gold lines (i.e., $b = 0$) yields reasonable agreement between theory and experiment. For instance, in both cases, the diffracted orders occur at expected angles, and the higher-order diffraction efficiency (relative to the zero order) agrees with expected values for $b = 0$. However, two discrepancies are evident. First, the zero-order diffraction efficiency of measured HDMMs does not agree with expected values. Second, the higher-order diffraction efficiencies oscillate.

As mentioned in our description of fabrication, imperfections in the fabrication procedure led to imperfect HDMMs. For our 6- μm -period HDMMs, a 5- to 10-nm-thick coat of chromium

Table 3-22. Measured performance of a mirror with $d = 11.5$ nm, $p = 6$ μ m, $t = 100$ nm, and $\beta = 2.75$.^a

Diffracted order	Calculated performance (b = 0)	Measured performance
I_0	0.12	0.08
$\frac{I_{\pm 1}}{I_0}$	0.1	0.950 ± 0.015
$\frac{I_{\pm 2}}{I_0}$	0.05	0.450 ± 0.015
$\frac{I_{\pm 3}}{I_0}$	0.006	0.150 ± 0.005

^a Normal-incidence reflection measured from unpatterned mirror is 23% at $\lambda = 20.8$ nm.

Table 3-23. Measured performance of a mirror with $d = 11.5$ nm, $p = 200$ μ m, $t = 55$ nm, and $\beta = 3.35$.^a

Diffracted order	Calculated performance (b = 0)	Measured performance
I_0	0.08	0.02
$\frac{I_{\pm 1}}{I_0}$	0.21	0.30 ± 0.05
$\frac{I_{\pm 2}}{I_0}$	0.023	0.025 ± 0.005
$\frac{I_{\pm 3}}{I_0}$	0.011	0.011 ± 0.002

^a Normal-incidence reflection measured from unpatterned mirror = 23% at $\lambda = 20.8$ nm.

overcoats the multilayer. Calculations indicate that this layer can reduce mirror reflectivity at $\lambda \approx 20$ nm by 30 to 60%, depending on exact thickness and wavelength. For our 200-nm-period HDMM, overetching by the argon ion beam could easily reduce peak reflectivity. Both of these effects can explain the reduced reflectivity from the structure and should not affect diffraction efficiencies of the different orders relative to each other. In this case, the model agrees well with data.

On the other hand, small oscillations in higher-order diffraction efficiencies (e.g., Fig. 3-210) cannot be easily explained. If such oscillations were due to nonzero normal-incidence reflections from the top of the gold lines, we would expect to see oscillations in the diffraction efficiency as the wavelength is scanned. The period of this oscillation would depend on x-ray wavelength

(~ 200 nm) and the gold line thickness because reflections from the grating spaces add constructively or destructively with reflections from the top of the gold lines. For the 6- μ m-period HDMM (gold thickness = 100 nm), the period of calculated oscillation as wavelength is scanned should be ~ 2 nm, but the measured period is ~ 1.2 nm. For the 200-nm-period HDMM (gold thickness = 55 nm), the period of oscillation should be ~ 4 nm, but only small oscillations are apparent with a period of oscillation of ~ 2 nm. For practical purposes, the magnitude of oscillations is small; nevertheless, they cannot be adequately explained by our model.

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Scattering of X Rays from Non-Ideal Multilayer Structures. X-ray multilayer optics are increasingly useful in Nova experiments and diagnostic instrumentation. For example, an efficient soft x-ray beam splitter³⁶⁰ was successfully demonstrated in the first x-ray laser cavity³⁶¹; as in the visible analog, this device divides an incident x-ray beam into approximately equal specularly reflected and transmitted components. The performance of multilayer x-ray optics is invariably degraded due to diffusion and mixing at the interfaces and roughness of the layers replicated from the substrate or induced by processes such as polycrystalline growth.^{362,363} The non-ideal interfaces modify the specular scattering and can also scatter x rays into non-specular directions. The non-specular scattering is usually ignored, although it can be a problem in imaging applications, where it can produce undesirable halos and background noise in the image field.

We developed a comprehensive theory of the scattering of x rays from a non-ideal multilayer structure. The theory is valid for arbitrary polarization and at all angles of incidence (measured from the normal) less than the critical angle for total external reflection. Most importantly, there are no constraints on the

physical structure or spatial extent of the interfaces.

The complexity of the general theory precludes a thorough presentation in this report. Instead, we review the important special case of specular scattering from a non-ideal multilayer structure. The layered medium is described by a spatially varying dielectric function $\epsilon(\mathbf{x})$. Consider the x-ray scattering from a particular interface that represents the transition between two layers having dielectric constants ϵ and ϵ' . Let the z-direction be normal to the interface and θ be the angle of incidence of the x rays (measured from the z-axis).

The specular components of the scattered field are only sensitive to the average variation of the dielectric function across the interface. We define the interface profile function $p(z)$ to represent the average normalized value of the dielectric function along the z-direction:

$$p(z) \equiv \frac{1}{\epsilon - \epsilon'} \frac{\iint |\epsilon(\mathbf{x}) - \epsilon'| dx dy}{\iint dx dy} \quad (73)$$

In addition, we define $w(z)$ as the derivative of the interface profile:

$$w(z) \equiv dp(z)/dz. \quad (74)$$

The advantage of this formalism is its complete generality; the functions $p(z)$ and $w(z)$ can represent an interface of arbitrary structure. To illustrate the role of these functions, Fig. 3-212 shows three specific types of interfaces. Figure 3-212(a) shows the ideal interface where the transition between two media having dielectric constants ϵ and ϵ' occurs abruptly at $z = 0$. The profile $p(z)$ is a unit step function and $w(z)$ is a delta function $\delta(z)$. In a purely rough interface shown in Fig. 3-212(b), the interface profile $p(z)$ is smooth even though the transition between the media at any position (x,y) is abrupt. In particular, the boundary between the two media can be defined by a surface $z = f(x,y)$. Then the function $w(z)$ represents the normalized distribution of surface heights $f(x,y)$ about the plane $z = 0$. In comparison, Fig. 3-212(c) shows a purely diffuse interface where the dielectric function varies smoothly along the z-direction and is

Fig. 3-212. One-dimensional representations of different types of interfaces having profiles $p(z)$ and derivatives $w(z)$.

uniform in the x - y plane. In this case, $w(z)$ represents the actual composition gradient across the interface.

The amplitudes of specular reflection r^0 and transmission t^0 at an ideal interface are described by the Fresnel equations,

$$r^0 = \frac{\gamma - \gamma'}{\gamma + \gamma'}, \quad t^0 = \frac{2\gamma}{\gamma + \gamma'} \quad (75)$$

for s -polarization, and

$$r^0 = \frac{\epsilon\gamma' - \epsilon'\gamma}{\epsilon\gamma' + \epsilon'\gamma}, \quad t^0 = \frac{2\sqrt{\epsilon\epsilon'}\gamma}{\epsilon\gamma' + \epsilon'\gamma} \quad (76)$$

for p -polarization, where

$$\gamma = k_0 \sqrt{\epsilon - \sin^2 \theta}. \quad (77)$$

Here $k_0 = 2\pi/\lambda_0$, where λ_0 is the vacuum wavelength of the x rays. For a non-ideal interface it can be shown that to first order in $\epsilon - \epsilon'$ (a small quantity at x-ray wavelengths) the specular reflection amplitude is modified by

$$r = r^0 \bar{w}(-2k_0 \cos \theta), \quad (78)$$

where $\bar{w}(s)$ is the Fourier transform of $w(z)$. The specular transmission amplitude through the non-ideal interface is unchanged from the Fresnel values to first order.

Hence the evaluation of the specular scattering from a non-ideal interface requires an accurate determination of the profile function $p(z)$. Ideally, if the exact three-dimensional structure of the interface were known then the actual profile $p(z)$ could be calculated numerically by averaging the dielectric function over the x and y directions. In general, however, such detailed knowledge of the structure of the interface is unavailable, and it is expedient to model the interface profile using a simple analytical function. It is convenient (but not necessary) to assume that $w(z)$ is a symmetric function. The width of the interface σ is defined by the second moment of $w(z)$:

$$\sigma^2 = \int z^2 w(z) dz. \quad (79)$$

Table 3-24 lists several useful models of the interface profile $p(z)$ and the corresponding derivatives $w(z)$ and Fourier transforms $w(s)$. The appropriate choice of model is dictated largely by the nature of the interface. For instance, the classical diffusion of two materials produces an interface profile $p(z)$ described by an error function, for which $w(z)$ is a Gaussian. In contrast, the mixing at an interface produced by bombardment of the surface by energetic atoms during sputter deposition might be better described by a linear composition gradient, and consequently $w(z)$ is the rectangle function. In the case of a rough interface, it has been suggested that the distribution of surface heights about the mean position, which is equivalent to $w(z)$, is best described by an exponential function.³⁶⁴

As a specific example, consider an interface of width σ having a profile represented by the error function,

$$p(z) = \frac{1}{\sqrt{\pi}} \int_{-\infty}^z \exp(-t^2/2\sigma^2) dt. \quad (80)$$

Then the derivative $w(z)$ is the Gaussian,

$$w(z) = \frac{1}{\sqrt{2\pi}\sigma^2} \exp(-z^2/2\sigma^2), \quad (81)$$

with a Fourier transform given by

$$\bar{w}(s) = \exp(-\sigma^2 s^2/2). \quad (82)$$

Substitution of Eq. (82) into Eq. (78) yields

$$r = r^0 \exp(-2k_0^2 \sigma^2 \cos^2 \theta). \quad (83)$$

Thus, the specular reflection amplitude from an interface having an error function profile decreases exponentially with the dimensionless parameter $(\sigma/\lambda)^2$. If the width σ is much less than an x-ray wavelength then the interface reflects essentially as if it were ideal. However, if the width is much greater than a wavelength, then the specular reflection

Table 3-24. Several useful interface profiles $p(z)$ and the related functions $w(z)$ and $\tilde{w}(s)$.

Description of interface	$p(z)$	$w(z)$	$\tilde{w}(s)$
Error function	$\frac{1}{\sqrt{\pi}} \int_{-\infty}^z e^{-t^2/2\sigma^2} dt$	$\frac{1}{\sqrt{2\pi\sigma^2}} \exp(-z^2/2\sigma^2)$	$\exp(-s^2/2)$
Exponential	$\begin{cases} \frac{1}{2} e^{\sqrt{2}z/\sigma} & z \leq 0 \\ 1 - \frac{1}{2} e^{\sqrt{2}z/\sigma} & z > 0 \end{cases}$	$\frac{1}{\sqrt{2\sigma^2}} e^{-\frac{\sqrt{2} z }{\sigma}}$	$\frac{1}{\left(1 + \frac{s^2\sigma^2}{2}\right)}$
Linear	$\begin{cases} 0 & z < -\sqrt{3}\sigma \\ \frac{1}{2} + \frac{z}{2\sqrt{3}\sigma} & z \leq \sqrt{3}\sigma \\ 1 & z > \sqrt{3}\sigma \end{cases}$	$\begin{cases} 0 & z > \sqrt{3}\sigma \\ \frac{1}{2\sqrt{3}\sigma} & z \leq \sqrt{3}\sigma \end{cases}$	$\frac{\sin(\sqrt{3}\sigma s)}{\sqrt{3}\sigma s}$
Sinusoidal	$\begin{cases} 0 & z < -a\sigma \\ \frac{1}{2} + \frac{1}{2} \sin\left(\frac{\pi z}{2a\sigma}\right) & z \leq a\sigma \\ 1 & z > a\sigma \end{cases}$ $a = \frac{\pi}{\sqrt{\pi^2 - 8}}$	$\begin{cases} 0 & z > a\sigma \\ \frac{\pi}{4a\sigma} \cos\left(\frac{\pi z}{2a\sigma}\right) & z \leq a\sigma \end{cases}$	$\frac{\pi}{4} \left[\frac{\sin\left(a\sigma s - \frac{\pi}{2}\right)}{a\sigma s - \frac{\pi}{2}} + \frac{\sin\left(a\sigma s + \frac{\pi}{2}\right)}{a\sigma s + \frac{\pi}{2}} \right]$

amplitude is significantly reduced. Inspection of the various forms for $\tilde{w}(s)$ in Table 3-24 shows that this behavior is in fact common to all of the interface models.

A multilayer x-ray mirror is typically composed of alternating layers of high- Z and low- Z materials having a periodic bilayer thickness D . Neglecting corrections for refraction, the structure has a strong specular reflection when

$$m\pi = Dk_0 \cos \theta, \quad (84)$$

where m is an integer corresponding to the order of the reflection. Equation (84) represents the resonance condition at

which the fields reflected specularly from each interface add coherently, resulting in a total scattered field that is comparable in strength to the incident field.

The specular scattering of x rays from a multilayer structure is calculated by a matrix method.³⁶⁵ Let the multilayer structure be composed of a stack of N dielectric layers surrounded on both sides by vacuum (see Fig. 3-213). A plane wave of unit amplitude $\hat{e}^0 \exp(i\mathbf{k}_0 \hat{n}_0 \cdot \mathbf{x})$ is incident on the top of the stack. The propagation vector has components $n_x^0 = 0$, $n_y^0 = \sin \theta$, and $n_z^0 = \cos \theta$, and the polarization $\hat{e}$ is chosen to be either s - or p -type. Neglecting the scattering into

Fig. 3-213. Configuration of the multi-layer stack.

non-specular directions, the field in the i th layer away from either interface region is

$$E_j(\mathbf{x}) = \exp(ik_0 n_y^0 y) \times [E_i^- \hat{e}^- \exp(-ik_j n_z^i z) + E_i^+ \hat{e}^+ (ik_j n_z^i z)], \quad (85)$$

where $k_i = |\epsilon_i|^{1/2} k_0$ and n_z^i is a complex quantity for an absorbing medium given by

$$n_z^i = \frac{1}{|k_i|} \sqrt{k_i^2 - k_0^2 n_y^0{}^2}. \quad (86)$$

The polarization $\hat{e}^\pm$ in Eq. (85) corresponds to s - or p -type in accordance with the polarization of the incident field.

Consider the interface between the adjacent layers i and j . The interface couples the specular fields in the two layers and, if it is rough, removes some energy from the specular field. The amplitudes of the specular fields in the adjacent

layers are related by the 2×2 scattering matrix $\mathbf{S}_{ij}$,

$$\begin{pmatrix} E_i^- \\ E_i^+ \end{pmatrix} = \mathbf{S}_{ij} \begin{pmatrix} E_j^- \\ E_j^+ \end{pmatrix} \quad (87)$$

where

$$\mathbf{S}_{ij} = \frac{1}{t_{ij}} \begin{pmatrix} 1 & r_{ij} \\ r_{ij} & t_{ij} t_{ji} + r_{ij}^2 \end{pmatrix} \quad (88)$$

Here r_{ij} and t_{ij} are the specular reflection and transmission amplitudes of the interface as given by Eqs. (77) and (78).

The specular fields at the top and bottom of the i th layer are related by a propagation matrix $\mathbf{P}_i$ defined as

$$\mathbf{P}_i = \begin{pmatrix} e^{-i\phi_i} & 0 \\ 0 & e^{i\phi_i} \end{pmatrix}, \quad (89)$$

$$\phi_i = k_0 d_i \sqrt{\epsilon_i - \sin^2 \theta}, \quad (90)$$

and d_i is the thickness of the i th layer.

The amplitudes of the reflected field at the top of the stack E^r and the transmitted field at the bottom of the stack E^t are calculated through a series of matrix multiplications. Starting at the bottom of the stack, the field is scattered from each interface and propagated through each layer until reaching the top of the stack. Hence the fields above and below the stack are related by a matrix $\mathbf{A}$,

$$\begin{pmatrix} 1 \\ E^r \end{pmatrix} = \mathbf{A} \begin{pmatrix} E^t \\ 0 \end{pmatrix}, \quad (91)$$

where $\mathbf{A}$ is a product of scattering and propagation matrices,

$$\begin{aligned} \mathbf{A} &= \begin{pmatrix} A_{11} & A_{12} \\ A_{21} & A_{22} \end{pmatrix} \\ &= \mathbf{S}_{v,n} \mathbf{P}_n \mathbf{S}_{n,n-1} \dots \mathbf{S}_{i+1,i} \mathbf{P}_i \mathbf{S}_{i,i-1} \dots \mathbf{S}_{2,1} \mathbf{P}_1 \mathbf{S}_{1,0}. \end{aligned} \quad (92)$$

Here $S_{u,n}$ and $S_{i,v}$ represent the interfaces between vacuum and the top and bottom layers, respectively. The amplitudes of the field outside of the stack are

$$E^r = \frac{A_{21}}{A_{11}}, \quad E^t = \frac{1}{A_{11}}. \quad (93)$$

Finally, the intensities of the specular fields reflected and transmitted by the multilayer stack are given by $|E^r|^2$ and $|E^t|^2$, respectively.

A computer code, developed for calculating the reflectance and transmittance of multilayer x-ray optics based on the matrix method, is available on both of the LEAD group's VAX computers. In the code, the multilayer stack is represented by a sequence of interfaces. The structure of each interface is defined by a choice of profile function (from Table 3-24) and a width σ . The elemental composition of the layers is specified and the optical constants are retrieved from on-line tabulations compiled by Henke et al.³⁶⁶

To demonstrate the utility of the code, we calculate the scattering properties of a free-standing x-ray beam splitter. The beam splitter is composed of 15 alternating layers of W and C, each 2.0-nm thick, the interfaces are assumed to be ideal. Figure 3-214 shows the reflectance, transmittance, and absorbance as a function of the angle of incidence of unpolarized x rays at a wavelength of 1.0 nm. The scattering resonance is evident as a strong peak in reflectance at 82.5°. The reflectance and transmittance at this angle have values of 0.27 and 0.18, respectively. Both the transmittance and absorbance exhibit local minima near the resonance angle.

Figure 3-215 shows the effect of non-ideal interfaces on the performance of the beam splitter. All of the interfaces are modified equivalently and are described by an error-function profile of width σ . Figure 3-215(a) shows the reflectance and Fig. 3-215(b) shows the transmittance for four values of σ varying between 0 and 0.6 nm. It is apparent that the reflectance decreases with increasing σ , by as much as 50% at $\sigma = 0.6$ nm. In the vicinity of the reflectance peak, the

Fig. 3-214. The ideal beam splitter as a function of the incident angle (measured from the normal) of the x-ray beam.

transmittance increases as σ increases, indicating a redistribution of energy from the reflected to the transmitted field. In general, the reflectance is always reduced by the introduction of non-ideal interfaces. The transmittance can either increase or decrease, depending on the detailed structure of the beam splitter and the scattering parameters. The significant modification of the reflectance and transmittance seen in Fig. 3-215 illustrates the importance of accounting for the non-ideal interface structure when modeling the performance of realistic multilayer x-ray optics.

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Characterization of Multilayer X-Ray Mirrors by Small-Angle X-Ray Scattering and Transmission Electron Microscopy. The ability to design, fabricate, and characterize multilayer x-ray optics has become an increasingly important part of the laser program's diagnostic effort. Sputter deposition and sub-micron fabrication techniques produce x-ray optical components such as multilayer mirrors, beam splitters, and highly dispersive reflection gratings.^{367,368} Since the fabrication of these components requires precise control of dimensions as small as an x-ray wavelength, the reliable characterization

Fig. 3-215. Calculated performance of the beam splitter with non-ideal interfaces (error function profiles) as a function of the angle of incidence at $\lambda = 1$ nm.

of the structures is crucial. We used two important techniques to characterize multilayer x-ray optics: small-angle x-ray scattering (SAXS) and transmission electron microscopy (TEM).

A multilayer x-ray mirror is typically composed of alternating layers of high-Z and low-Z materials with a bilayer spacing between ~ 1 and 20 nm.³⁶⁹ SAXS is a well-established technique for measuring the bilayer spacing of a multilayer mirror.³⁷⁰ The coherent scattering of a monochromatic, collimated x-ray beam at grazing incidence is measured in the θ - 2θ specular reflection geometry (θ is measured with respect to the surface of the

mirror). In our case, the measurements were performed on a Rigaku RU-200 powder diffractometer using a Cu $K\alpha$ source (wavelength 0.154 nm) and a beam of divergence 0.54 mrad. Figure 3-216(a) shows data obtained from SAXS. The multilayer mirror was composed of 50 bilayers of W and C with a nominal bilayer spacing of 4.4 nm, deposited by magnetron sputtering onto a substrate consisting of a polished single-crystal Si wafer. The scattering of x rays from the multilayer mirror exhibits strong peaks at angles m , where m represents the order of the reflection. The relation between the bilayer spacing d and the angle of the m th-order reflection is given approximately by the modified Bragg equation,³⁷¹

$$\frac{m\lambda}{2 \sin \theta_m} = d - \delta \csc^2 \theta_m \quad (94)$$

Here δ is the difference from unity of the real part of the average index of refraction, which is generally small ($< 10^{-4}$) at x-ray wavelengths. In Fig. 3-216(b), we plotted $m\lambda/(2 \sin \theta_m)$ vs $\csc^2 \theta_m$ for the five measurable orders of reflection. As indicated by Eq. (94), the points define a line whose y-intercept is the bilayer spacing and whose slope yields the average index of refraction. From the data we obtain a bilayer spacing for this mirror of 4.59 nm.

High-resolution imaging using TEM is a powerful and direct tool for studying the microstructure of multilayer x-ray optics. (The TEM images were obtained at the Center for Solid State Science at Arizona State University in collaboration with M. B. Stearns, C.-H. Chang, and D. J. Smith.) We performed high-resolution imaging on multilayer structures using a JEM 4000EX TEM. The microscope operates at 400 kV and provides an image resolution of 0.15 nm. Ultrathin cross-sectional specimens of the multilayer structures are prepared by mechanical polishing followed by ion beam milling. Details concerning the specimen preparation and the procedures used to obtain the images can be found elsewhere.³⁷²

To demonstrate the utility of SAXS and TEM as characterization techniques, we

Fig. 3-216. (a) SAXS scan of a W/C multilayer x-ray mirror, showing the reflected x-ray intensity ($\lambda = 0.154$ nm) as a function of grazing incidence θ ; (b) plot of $m\lambda/(2 \sin \theta_m)$ vs $\csc^2 \theta_m$ for each angular position θ_m of the m th order reflection.

describe an experiment in which the effects of thermal annealing of a multilayer x-ray mirror were investigated. The motivation for this study was twofold: first, to determine the temperature at which the performance of the mirror degraded and to identify the failure mechanism; and, second, to test the validity of the TEM analysis. In the past there has been concern that the preparation process modifies the specimen so that the images obtained with TEM are not accurate representations of the microstructure of the multilayer mirror. In particular, it has been suggested³⁷³ that the heat generated during the thinning of the specimen modifies the structure of the interfaces between the layers. By annealing a series of samples in a relevant range of temperature, it was possible to determine if the temperature increase during specimen preparation ($<200^\circ\text{C}$) is sufficient to produce significant structural changes.

The multilayer mirror was composed of 30 bilayers of Mo and Si with a 1-nm overcoat of C, all deposited by magnetron sputtering onto a polished single-crystal Si wafer. The nominal bilayer spacing was 11.0 nm and the ratio of the Mo-layer thickness to the bilayer thickness was nominally 0.4. The mirror was cut into five samples each approximately 1 cm $\times$ 3 cm. Each sample was characterized by SAXS before annealing. All of the samples yielded at least 13 orders of reflection. The amplitude and position of the peaks were consistent between samples to within 3% and corresponded to a bilayer spacing of 11.1 nm.

The samples, labeled A through E, were annealed for 30 min in a Thermo Lyne 1400 oven. Samples B, C, D, and E were annealed at temperatures of 200, 400, 600, and 800°C respectively. Sample A, the control sample, was not annealed. To avoid oxidation, the samples were heated and cooled in an ambient atmosphere of argon. After the oven was allowed to equilibrate to the desired temperature (temperature control $\pm 5\%$), the sample was inserted for the appropriate time and then removed.

The changes of the multilayer structure induced by thermal annealing were analyzed with SAXS and TEM. Figure 3-217 shows data obtained from SAXS in the angular range of $\theta = 0$ to 5° for each of the five samples. Figures 3-217(a) and (b) show that the relative intensities of the reflection peaks are essentially identical for samples A and B. This indicates that negligible structural changes are induced by annealing at 200°C . Figure 3-217(c) shows that the first obvious changes occur at 400°C for sample C, where the relative intensities of several of the reflection peaks are altered. The reflectance of the mirror remains quite high, however, and 11 reflection orders are seen, indicating that the layered structure is still intact. By contrast, Figs. 3-217(d) and (e) show profound changes in samples D and E at 600 and 800°C . At 600°C , only the three lowest orders of reflection are observable; at 800°C , only the first order is evident. In both cases, the amplitude of the reflection is severely reduced.

Fig. 3-217. SAXS scans of Mo/Si multilayer mirrors after annealing at various temperatures for 30 min.

Fig. 3-218. Variation of the bilayer spacing d as a function of the anneal temperature.

Figure 3-218 shows the bilayer spacings of the five samples calculated from the SAXS data. The thickness of the layers is fairly constant through the 400°C anneal. Values of the bilayer spacing obtained from the 600 and 800°C data are less reliable owing to the fewer orders of reflection. However, these suggest that the spacing decreases at the elevated temperatures.

Figure 3-219 shows high-resolution TEM images of the five samples. The dark and light bands correspond to the Mo and Si layers, respectively. As anticipated from the SAXS data, samples A and B are virtually identical, exhibiting the structure previously observed³⁶⁷ for high-quality Mo/Si mirrors. In particular, the layers are smooth and separated by asymmetric interface regions of 1- to 2-nm width. The Mo layers are composed of long crystallites having a preferred (110) orientation and the Si layers are amorphous.

Sample C at 400°C shows basically the same structure seen in the lower temperature samples but has wider interfaces. This suggests that at 400°C the temperature is sufficient to drive the diffusion of the materials at the interface. However, since the interface regions remain amorphous, the layers are smooth.

Fig. 3-219. High-resolution TEM images of the Mo/Si multilayer structure after annealing.

At 600°C, the structure of sample D underwent a dramatic change. Diffusion has caused the Mo and Si to mix to the extent that no crystalline Mo remained. Instead the Mo layers were replaced by layers consisting of crystallites of the intermetallic Mo-Si compounds. The processes of diffusion and recrystallization created very rough layers separated by pockets of amorphous Si and apparent voids. The degradation of the layered structure is clearly correlated with the decrease in the amplitude and the number of orders of reflection observed with SAXS. Finally, Figure 3-219(e) shows that at 800°C the layer structure is completely destroyed. The sample is an inhomogeneous polycrystalline mixture of Mo-Si intermetallic compounds.

In conclusion, SAXS and TEM provide valuable information about the effects of thermal annealing on Mo/Si multilayer mirrors. Together these techniques present a consistent picture of the modification of the layered structure induced by elevated temperature. The close similarity of the results for the sample that was not annealed and the sample annealed at 200°C implies that heating during specimen preparation for TEM does not induce significant structural changes. The integrity of the multilayer structure is maintained through 400°C. At 600°C there are drastic structural changes. The

layers are rough and distorted, and consequently the reflectance is severely reduced. The mechanism for the degradation of the layers is diffusion of the Mo and Si leading to compound formation and recrystallization.

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Photoconductors as X-Ray Detectors.

We have explored the application of high-speed photoconductive detectors (PCDs) to the measurement of x-rays from laser plasmas. Our effort had two significant aspects: the first involved tests and calibrations of the detectors themselves, the second involved experimental applications of the detectors. We performed tests and calibrations on these devices for a variety of radiation sources in several new materials to expand on previous experiments with InP:Fe PCDs.³⁷⁴

A photoconductor is a device with two contacts and an applied electric field, as shown in Fig. 3-220. When no radiation is incident on the detector, a small leakage current flows through the detector because of its finite resistivity and ohmic

Fig. 3-220. Operation of a photoconductor. (a) No radiation-leakage current, (b) excitation-free carriers generated by radiation-current flow increase, and (c) trapping-free carriers trap and cease to conduct.

Table 3-25. Radiation sources used to characterize and test PCDs.

Source	Pulse width (ps)	Range
Y-division picosecond laser	2	0.6 to 0.2 nm
Stanford synchrotron	350	500 to 3 000 eV
Janus laser plasma	100	50 to 20 000 eV
Diode laser	55	0.84 nm

contacts. If photons with energy great enough to form electron and hole pairs are absorbed in the detector, then the carriers drift in the electric field, and current increases. The carriers soon are stopped at traps in the material, and both the number of free carriers and current decrease exponentially. The trapping time can be made very short, $t_{tr} = 10$ ps; therefore, the detectors can be made to operate very quickly. Speed is thus one advantage of PCDs. Another advantage is that PCDs are simple to operate in measuring absorbed radiation, which may permit their use as a flat-response detector or bolometer for x rays.³⁷⁵

We have studied three new materials in addition to InP:Fe, which had been

investigated earlier.³⁷⁴ The materials are neutron-damaged GaAs, thin-film GaAs on sapphire, and type-IIa insulating diamond. We studied each material on some of the radiation sources listed in Table 3-25. Each material has unique qualities that suit it for particular applications.

Neutron-damaged GaAs is a high-speed material ($t_{tr} = 60$ ps) that can be fabricated into detectors with millimeter dimensions. Neutron damaging provides uniform lattice damage that introduces trapping sites into the GaAs; the material retains relatively high mobility and, thus, high sensitivity at the same time. These detectors have been used to measure x rays from gold and gold:beryllium laser plasmas on Nova and at the Naval Research Laboratory on the Pharos III laser. In addition, these detectors have been used to detect thermonuclear neutrons. Synchrotron and laser-plasma measurements indicate that such detectors have a flat response for photon energies greater than 500 eV, but indications suggest that the response to lower-energy, less-penetrating radiation is controlled by surface mobility and lifetimes, which may differ from the bulk and result in a changing response with energy.

Thin-film GaAs on sapphire is another high-speed material due to lattice imperfections that exist in metal-organic-chemical-vapor-deposition (MOCVD) thin films. The structure eliminates the need for neutron damaging and permits discrimination against high-energy ($E > 5$ keV) photons. Electro-optic methods have been used to measure $t_{tr} = 11$ ps for a 1- μ m-thick sample. Figure 3-221 shows a sample x-ray pulse from plasma produced by the Janus laser.

Type-IIa insulating diamond has many interesting properties, including a large band gap (5.4 eV), high thermal conductivity, low electrical conductivity, and extreme hardness. Such properties indicate that a robust detector can be made. The material is also insensitive to scattered laser light because of its 5.4-eV band gap. The insensitivity is a distinct advantage for measuring x-ray conversion efficiency from laser plasmas because no filtration against scattered laser light is required to measure total x-ray yield. Experimentation is more difficult

with insulating diamond detectors because of their insensitivity to visible radiation, which requires measurements to be performed with UV light or x rays. As shown in Fig. 3-222, the trapping time of a random sample of diamonds varied from 150 to 600 ps. This variation is a result of the use of a natural material; no control can be exercised on dopants or uniformity other than by selection.

We have compared the bolometric properties (i.e., measurement of total energy) of natural diamond to measurements by the Dante and Henway systems in a variety of laser-plasma interaction experiments. Figure 3-223 shows our results. A near-linear relation exists between total radiated power, as measured by the Dante and Henway systems, and integrated photoconductor signal. Small, but significant, deviations exist for extremely soft spectral distributions (beryllium plasmas), a result of the increase in detector sensitivity as the photon energy approaches the band gap of the material. This result is obtained in diamond for photon energies less than approximately 100 eV.

In summary, we have found new materials and new applications for PCDs. High-speed, single-shot detection ($t = 100$ ps) can be performed with these detectors for x-ray signals. The detectors can be tailored to fit a variety of measurement requirements, including high speed and sensitivity or insensitivity to visible light or soft x rays. We expect that PCDs will find more applications as our understanding of their operation increases.

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Neutron Measurements.

Neutron Emission-Time Measurements. The average neutron emission time and the temporal evolution of the burn of an ICF target are good quantities with which to test models that describe hydrodynamic and implosion physics. This past year, we began measuring these quantities relative to incident laser power with fast neutron detector systems.

To record neutron production history, we need a neutron detector response

Fig. 3-221. Sample radiation pulse from a thin-film GaAs detector. Pulse width is limited by response of the single-shot recording system.

Fig. 3-222. Photo-response of a diamond photoconductor to 2-ps, 200-nm pulses.

Fig. 3-223. Total power radiated from laser-plasma interaction measured by Dante and Henway compared to time-integrated diamond PCD signal.

time that is less than the burn duration. While average neutron emission time can be determined to within ± 75 ps with a substantially slower detector system, the important requirement here is that characteristic features of detected signals be measured with an accumulated error of < 75 ps.

Neutron emission-time measurements require simultaneous recording of the incident laser power and the neutron signal (see Fig. 3-224). Laser beams heat and compress the target at the center of

the target chamber, causing the isotropic emission of thermonuclear neutrons from a small region of the fuel. An optical streak camera with a response time of 50 ps (FWHM) records the incident power for each laser beam. The neutrons travel outward from the target and interact with the neutron detector to generate a signal that is recorded with an oscilloscope. Optical fiducial pulses³⁷⁶ injected into the streak camera and neutron detector provide the information needed to cross-time the recorded signals.

The time interval between the fiducial pulses arriving at their respective detectors is a calibration constant we measure as follows. We irradiate a gold disk with an intense 100-ps Gaussian laser pulse to create a short burst of x rays. The high-energy (>10 keV) x rays emitted simultaneously with the irradiation then interact with the neutron detector to produce a signal in much the same manner that

neutrons do. From the recorded signals, we calculate the time interval between fiducial pulses.

Two separate neutron detector systems are used for measurement. The fastest system, which has a response time of 130 ps, uses a radiation-induced conductivity (RIC) device (see "Measurement of Single, 14-MeV Neutron Pulses with 130-ps Resolution," later in this section) coupled to a 6-GHz Tektronix 7250 oscilloscope. When placed 5 cm from the target, this detector can measure both burn history and average neutron emission time for deuterium-tritium (DT) yields as low as 5×10^{10} neutrons.

The second neutron detector system³⁷⁷ uses a fast plastic scintillator that is optically coupled to a microchannel plate (MCP) photomultiplier tube placed 60 cm from the target. Readout is provided by 500-MHz Tektronix R7912 transient digitizers. This type of detector records signals for targets with DT neutron yields above 10^6 and has a system response of 825 ps to the optical fiducial pulse and 1200 ps to neutrons. The additional neutron-signal broadening is caused by the exponential decay of scintillator light. Although too slow to measure burn histories, this detector system can be used to determine average neutron emission time within ± 75 ps by timing to the 50% point (constant fraction timing) on the leading edge of signal pulses. Figure 3-225 shows a signal recorded with this detector system.

Doppler broadening of the neutron energy spectra results in temporal broadening of the neutron signal at the detector. The amount of broadening depends on fuel type, ion temperature, and target-to-detector distance. At 10 cm from a DT-filled target, broadening is 40 ps for an ion temperature of 10 keV. At 60 cm, the broadening is 240 ps. Thus, corrections must be made for the effects of Doppler broadening.

The standard deviation of random timing errors for the scintillator-MCP system is about 75 ps. This deviation is associated primarily with the reading of signals from the oscilloscope trace and the streak-camera image. While the time interval between the neutron and fiducial

Fig. 3-224. Experimental setup for neutron emission-time measurements.

Fig. 3-225. Oscilloscope trace showing neutron and fiducial signals recorded with the scintillator-MCP system for a shot that produced 10^{13} DT neutrons.

pulses on the oscilloscope can be read with a precision of ± 70 ps, the laser-to-fiducial time interval recorded on the streak camera can be determined to about ± 30 ps.

The two primary sources of systematic error are the fiducial-to-fiducial calibration time, which can be < 50 ps if the average of several calibration shots is used, and the measurement of the neutron flight path. A 2-mm distance error represents a 39-ps error in the time-of-flight correction for DT neutrons and a 92-ps error for deuterium-deuterium neutrons.

Measurements made with the MCP and RIC detector systems demonstrate the quality and reproducibility of the data. Figure 3-226 shows the neutron emission time relative to the leading edge of the laser pulse (as measured with the MCP system) for two types of ICF targets—large (1020 μm) and small (350 μm) microballoons. Neutron emission occurred at ~ 850 ps after the start of irradiation for the five experiments with the large microballoon and at ~ 1250 ps for the smaller target. The data show the reproducibility of both the measurement and the target implosion. Also plotted in this figure is the average emission time determined with the GaAs detector for one of the large microballoon experiments. The agreement between the two neutron detector systems confirms that the absolute timing calibration of the slower MCP detector has been done properly.

The goal of such measurements is to relate neutron emission time to incident laser power. Figure 3-227 shows the laser power and the average neutron emission time (as measured with the MCP detector) for a high-yield target experiment. The actual neutron production rate recorded with the GaAs detector is also shown.

Neutron emission is expected to occur at the time of peak target compression. Figure 3-228 shows the target shell diameter as a function of time for a high-yield target experiment as determined with a streaked x-ray microscope (see "X-Ray Streak Camera for Hydrodynamic Instability Experiments," earlier in this section). The x rays show that the shell diameter starts at 1000 μm , converges to ~ 400 μm at ~ 900 ps, and then begins to

Fig. 3-226. MCP-recorded neutron emission times for two target designs and the average emission time (solid circle) determined with the RIC detector.

Fig. 3-227. Neutron emission time relative to the incident laser pulse for a high-yield target, using MCP to measure average time and RIC to measure the actual production rate.

expand. Superimposed on the figure is the measured neutron emission time, which comes near the time of maximum shell convergence estimated by the streaked x-ray microscope.

This past year we showed that neutron emission-time measurements made with the scintillator-MCP system have a reproducibility of ± 75 ps for yields ranging from mid 10^6 to more than 10^{13} DT

Fig. 3-228. Neutron emission time relative to maximum shell convergence for a high-yield target, as measured with a streaked x-ray microscope.

Fig. 3-229. Concept of penumbral imaging. The penumbra contains information about the shape of the source.

neutrons. Comparisons of neutron emission times measured with the MCP detector with those measured with the faster GaAs system and the radius-vs-time plots recorded with an x-ray microscope suggest the absolute timing of the MCP system is better than ± 75 ps. Our first neutron burn-width measurements made with fast RIC detectors show a burn width of >300 ps—a duration much greater than the 130-ps detector response. The signal amplitude of these measurements indicates that the RIC detectors are usable for targets with yields above about 5×10^{10} neutrons.

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Neutron Penumbral Imaging. We have obtained images of the neutron-emitting region of high-yield ICF targets with a penumbral coded-aperture imaging system. The major components of the imaging system are the penumbral aperture, neutron detector, alignment hardware, and image reconstruction software. In this article, we describe each component and present an example of the neutron-imaging results.

Penumbral imaging is a coded-aperture imaging technique that has been applied to x-ray imaging.³⁷⁸ It is conceptually similar to pinhole imaging, with the essential difference that the aperture must be larger than the source. Figure 3-229 shows the geometry of the penumbral-imaging system. The image produced consists of a uniformly bright region surrounded by a partially illuminated penumbra. All information about the source is encoded in this penumbra.

The aperture must effectively block 14-MeV neutrons while providing a sharp, isoplanatic point-spread function (PSF). To provide the necessary contrast, the aperture is constructed through a 0.63-cm-diam, 6-cm-thick gold cylinder. Neutrons are absorbed and scattered by the gold with a total mean free path of 3.2 cm. Because the aperture is far from the detector, few of the scattered

Fig. 3-230. (a) The aperture of the penumbral-imaging system has a minimum diameter of $407\ \mu\text{m}$ with a 3-D taper providing an isoplanatic PSF with a sharp cutoff. (b) Principle of the taper.

neutrons reach the detector. The aperture itself has a minimum diameter of $407\ \mu\text{m}$, with a 3-D taper that provides an isoplanatic PSF with a sharp cutoff [see Fig. 3-230(a)]. The principle of the taper is shown in Fig. 3-230(b). Lines drawn from various positions in the source plane produce self-similar cutoffs as they are swept from the edge of aperture into the torus. However, some distortion is introduced by the change in magnification associated with the different points in the source. The aperture can be regarded as a segment of a toroidal solid, and by proper choice of the segment, the distortion can be minimized. The process is a tradeoff: the larger the aperture radius of curvature R the sharper the cutoff, but at the expense of a less isoplanatic response. In our design, the magnification varies by $\pm 3\%$ across the field of view of the aperture ($\pm 200\ \mu\text{m}$), while the 10 to 90% width of the cutoff is $30\ \mu\text{m}$ referred to the source plane.

Precise alignment of the aperture to the target is critical, and an original method was used to achieve the required precision. A collimated laser beam illuminates a 2-mm-diam glass ball positioned precisely at the center of the target chamber. Light diverging from the ball is refracted by lenses mounted on each end of the aperture. The optical axis of each lens is aligned to the aperture

to within $3\ \mu\text{m}$ in a separate procedure. Spherical aberration of the glass ball and lenses causes the refracted light to converge in a region rather than at a point; within this region interference fringes are formed. The focal lengths of the two lenses are chosen to cause their refracted light to overlap, producing a moiré pattern at an alignment station just outside the target chamber. The aperture orientation is mechanically adjusted until the fringe patterns are precisely concentric, thus aligning the aperture to the target position. In addition, an interference pattern is projected many meters further along the aperture line of sight, facilitating the relatively coarse alignment of the detector to the aperture. The accuracy of this method proved to be better than $50\ \mu\text{m}$ referred to the source plane.

The neutron-detection system consists of several components, shown in Fig. 3-231. An 8-cm-diam circular array of 1240 NE102 plastic scintillators is the principal component. Each scintillator is 10 cm long with a square cross section 2 mm on a side. Within the plastic, neutrons interact primarily through elastic scattering events with the hydrogen nuclei, producing recoil protons that excite the scintillator as they slow down. The detector-element cross section is matched to the 0.7-mm maximum-transverse range of these recoil protons. About 25% of the

Fig. 3-231. Neutron-detection system.

Fig. 3-232. (a) Coded neutron image and (b) reconstructed neutron source.

incident-neutron energy is deposited in each element, producing blue light in proportion to the deposited energy. The blue-light image is too faint for direct imaging and must be amplified. The 8-cm-diam scintillator image is coupled to a 4-cm-diam image-intensifier tube with a 2:1 fiber-optic minifier. The microchannel-plate image intensifier provides a luminous gain of approximately 10^4 . Finally, the output of the image intensifier is lens-coupled to a charge-coupled-device array, which provides a digital image that is acquired by an LSI-11 computer system.

The coded image recorded on the detector must be deconvolved to determine the neutron-source distribution. In penumbral imaging, the Fourier transform of the PSF has a central lobe that is typically much narrower than that of the source, and has side lobes separated by zeros that extend out to higher frequencies. Spatial-frequency components that

lie close to these zeroes have a poor signal-to-noise ratio and must be deemphasized during the deconvolution process. This is accomplished with a Wiener filter,^{37,9} as follows:

$$G(u,v) = \frac{C(u,v)}{A(u,v)} \frac{1}{1 + \gamma \frac{\sigma^2}{A(u,v)^2}} \quad (95)$$

where A is the PSF, C is the coded image, and G is an estimate of the neutron-source distribution with all three variables in the Fourier transform domain. In this equation, u and v are spatial-frequency variables, σ^2 is the average variance of noise in the image, and γ is a filter coefficient determined through an iterative procedure. The Wiener filtering process does not cause distortion but does introduce some low-pass filtering, which can be quantified by inverting the second term in the equation. For Nova

neutron images, we use a very small value of γ , and the loss of resolution is small.

A fairly complicated neutron image demonstrates the capabilities of the diagnostic. A coded neutron image is shown in Fig. 3-232(a), with the reconstructed neutron source in Fig. 3-232(b). The image was obtained from the implosion of a directly illuminated target, a spherical, 760- μm -diam glass shell coated with a thin (1800-Å) layer of gold and filled with an equimolar mixture of deuterium and tritium at 6 atm (Ref. 380). The target produced a yield of 5×10^{11} neutrons. The pixel size is 25 μm , and the two-point resolution is calculated to be 60 μm . The neutron image shows a very asymmetric burn region with three separate lobes and two distinct peaks.

In summary, our recent experiments have demonstrated neutron penumbral imaging. The performance of our imaging system is in good agreement with predictions based on numerical and theoretical models. Our future efforts will improve the two-point resolution to 10 μm so that we can image targets that achieve high radial convergence and produce yields of 10^{11} neutrons or less. The high-resolution neutron-imaging system will be a valuable diagnostic tool for ICF experiments.

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Measurement of Single, 14-MeV Neutron Pulses with 100-ps Resolution. The laser-driven implosion of a small, DT-filled glass microshell for ICF purposes can produce in excess of 10^{13} 14-MeV thermonuclear neutrons in a single, subnanosecond pulse. Detailed knowledge of the temporal evolution of thermonuclear burn is important in understanding the physics of such implosions. In effect, the duration of a burn is a direct measure of confinement time.

In this article, we describe the development of a high-speed detector (FWHM = 130 ps) to measure the production rate of 14-MeV neutrons. Our detector is made of GaAs that has been neutron damaged to introduce trapping sites, which dramatically decreases the carrier lifetime. Because the detector exploits a bulk-material property to achieve high speed, its active volume can be significantly larger than that of solid-state diode detectors, which rely on carrier sweepout. The larger size results in a net increase in sensitivity to penetrating radiation, such as high-energy neutrons and gamma rays.

Operation of our detector is analogous to photoconductivity in the detection of electromagnetic radiation. The difference, however, is that free carriers are not created by photons, but rather, by secondary reactions following neutron collisions with atoms in the crystal lattice. We refer to this detection process by the more general term, radiation-induced conductivity. RIC detectors are capable of direct measurement of neutron pulses while having the advantage of being much simpler than neutron streak cameras and scintillator rise-time streak cameras that also have demonstrated high speed and high sensitivity during operation.

Method of Operation. In applications involving penetrating radiation (where the mean free path of the radiation is larger than the characteristic dimension of the detector), the principal advantage of a RIC detector is derived from the fact that the speed of the device can be determined by a bulk property of the material, the carrier recombination time, rather than by the geometry of the detector, as in the case of a diode. We can illustrate this advantage by comparing the speed and sensitivity scaling of a diode vs that for a RIC detector.

The signal level in volts produced by a detector is the product of sensitivity, detector volume, and number of particles per second incident on the detector. The sensitivity S of the two types of detectors scale identically, as follows:

$$S = \frac{\alpha \mu E_{\text{max}} \tau_c e R_{\text{fl}}}{L^2 h \nu}, \quad (96)$$

where α is the number of free carriers formed per incident neutron, $E_{\max}$ is the maximum electric field that can be applied to the detector, μ is carrier mobility, τ_r is carrier lifetime or sweepout time, R_0 is load impedance, h and w are dimensions of the detector transverse to the electric field, and L is the length of the detector along the electric field. For simplicity, we assume that detectors can be described by a single μ and τ_r .

One limit on the speed (fall time) of diodes is the carrier transit time

$$t_s = \frac{L}{v_s} = \frac{L}{\mu E_{\max}}, \quad (97)$$

where $v_s = \mu E_{\max}$ is the carrier velocity, or the saturated carrier velocity if the electric field is large enough (1.4×10^7 cm/s for GaAs from Ref. 381). The speed of a RIC detector is not limited by this effect because the carrier lifetime is determined by recombination time. Both types of detectors can be limited by capacitance effects. For most practical applications, the capacitive limit C_0 , assuming a parallel plate geometry, is

$$C_0 = \frac{\epsilon w h}{L}, \quad (98)$$

where ϵ is the dielectric constant of the detector material. The limiting impedance is either the transmission-line impedance in the case of a RIC detector or the series resistance of the detector for a diode. In either case, the impedance is less than or equal to 50 Ω .

The transit-time limit from Eq. (97) in diodes requires that a high-speed detector ($t_s = 100$ ps) have a length no greater than 10 μm (Ref. 381). This limitation also limits the area of the detector,³⁸² and the active volume V of the detector is restricted by the combination of the transit-time limit and capacitance. The active volume can be written as

$$V = \frac{t_s^3 v_s^2}{R \epsilon}. \quad (99)$$

The RIC detector speed is not limited by transit time, but rather, by carrier

lifetime, which is determined by t_r , a bulk property of the material. For fast RIC detectors, the length limit depends on the signal transit time. In the present case, this time is the transit of the excitation pulse (14-MeV neutrons) across the detector if radiation is parallel to the electric field or electromagnetic-signal transit time. Particles move at 17% of the speed of light, and a 100-ps transit time occurs at 0.5 cm. This length is much larger than is allowed by the sweepout time limit, and it results in a much larger active volume for a RIC detector. The active volume ratio of a RIC detector to a diode is $(v_p/v_s)^2$, where v_p is limited by incident radiation or electromagnetic-signal transit time, Ln/c , where n is the index of refraction of the detector material, and c is the speed of light. In the latter case, the detector dimension transverse to the particle flux is limited by particle transit time. The ratio of sensitivity for a RIC and a diode is t_r/t_s . Multiplying the sensitivity ratio by the volume ratio yields the ratio of output signals v_p/v_s , which is greater than a factor of 500 for both limits and represents a dramatic improvement in signal for the RIC detector.

We assume that operation of a RIC detector is independent of the primary excitation mechanism; the creation of free carriers is the only criteria. In our example, we estimated energy deposition in the detector from neutron flux by using COG, a time-dependent neutron and gamma-ray transport code. For neutrons, the dominant energy-transfer mechanism is elastic nuclear collisions with gallium and arsenic atoms. We used the TRIM code, which calculates transport of ions in matter, to estimate the fraction of energy transferred from recoil ions to free carriers. The calculated sensitivity was $1.2 \times 10^{-3} \text{ V} \cdot \text{ps} \cdot \text{neutron}^{-1} / \text{cm}^3$.

A larger detector volume has additional advantages. The larger volume implies more interactions, better detection statistics, and reduces the fraction of interactions associated with edges or regions near contacts, where material properties may not be characteristic of the bulk. In our analysis, we ignored any effects that may result from spatial

nonuniformities in the excitation of the material.

Experiments. Our experiments tested the response of a RIC detector to 14-MeV neutron pulses. The ICF targets were imploded using the Nova laser with approximately 20 kJ of 0.35- μm light in a 1-ns pulse. Data were recorded on high-speed, single-shot recording systems. The detector was a 1- $\times$ 1- $\times$ 3-mm rectangular parallelepiped of neutron-damaged GaAs mounted in a high-speed 50- Ω coaxial transmission line. We varied the distance between target and detector, neutron yield, and shielding geometry to study the performance of our detector and recording system.

We measured the impulse response of the detector system (detector, cables, bias tee, and oscilloscope) with 2-ps pulses of 0.53- μm light. Incident laser power was varied by over 2 orders of magnitude, with no change in the pulse shape and a linear amplitude. The impulse response of the detector system was 130 ps (FWHM). The FWHM of the detector alone was 60 ps using a high-speed sampling system.

We performed implosion experiments with DT-filled targets to verify that recorded signals were generated by neutrons, rather than by neutron-generated gamma rays. The detector was absolutely timed to the incident laser pulse, with a 0.53- μm timing fiducial coupled to the detector with a fiber-optic cable. The signal time of flight was measured for distances from 6 to 60 cm from the target, and in all cases corresponded to the velocity of 14-MeV neutrons. To ensure that neutron-generated gamma rays were not dominating the signal, we performed COG calculations to verify that neutron interactions within the detector dominated the signal. In addition, we observed no effect from changing the shielding around the detector, which would change the production efficiency of neutron-generated gamma rays.

Figure 3-233 shows signals from two different targets. As expected from our target design, the duration of fusion burn for target B was distinctly narrower than that for target A. The narrower pulse had a FWHM of 185 ps, demonstrating the nearly bandwidth-limited performance

Fig. 3-233. Comparison of neutron-generation rate of two ICF capsules. The FWHM of target A is 350 ps; that of target B is 185 ps.

Fig. 3-234. Normalized yield measured by the detector vs measured yield from nuclear activation measurements. The best-fit line shows nearly linear scaling.

(defined by optical excitation) of our detector system.

We also varied the excitation level of the detector by 2 orders of magnitude. Figure 3-234 shows the integrated detector signal (proportional to total neutron yield) versus the independently measured target yield. A logarithmic fit to these data yields an exponent of 1.03, which is consistent with linear operation, including errors in the experiments. The net sensitivity of the detector was $4 \times 10^{-3} \text{ V} \cdot \text{ps} \cdot \text{neutron}^{-1} / \text{cm}^3$, with a standard deviation of 40%. This sensitivity compares favorably with our simple calculated estimate of sensitivity.

Conclusions. We have demonstrated that RIC detectors can be used to measure single pulses of 14-MeV neutrons with a resolution of 130 ps. Our detector is linear and has distinct advantages over other types of solid-state detectors for penetrating radiation. The measured neutron-generation rate of ICF capsules and the trends in our data are in agreement with predictions of capsule performance. In future studies, we will attempt

to optimize both detector geometry and material and to understand the details of the excitation mechanism for particles.

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Large Neutron-Scintillator Arrays.

Our work this year has shown that secondary DT neutron measurements can be used to measure fuel areal density ($\langle \rho R \rangle$) for implosions at Nova. However, to eliminate ambiguities for the technique, which are caused by uncertainties in the triton slowing for high-density targets, it is necessary to accurately measure the secondary DT neutron energy spectra.³⁸³ For this reason, construction has begun on an extremely sensitive neutron spectrometer, known as the large neutron-scintillator array (LaNSA).

The LaNSA will consist of 960 channels of neutron detector, each consisting of a 10-cm cube of liquid scintillator, a photomultiplier tube, and associated electronics to operate in the "single-hit" detection mode.³⁸⁴ The array of detectors will be located 20-m from the target, directly under the target chamber. Each channel will have a 2-ns (FWHM) time resolution, resulting in 150 keV of energy resolution for 14-MeV, DT secondary neutrons.

In addition to providing secondary DT neutron spectra, we are designing the LaNSA to allow us to use the tertiary neutron technique. Our current estimates indicate that the secondary technique will not work well for fuel $\langle \rho R \rangle$ above ~ 50 to 100 mg/cm^2 ; the LaNSA is designed to be sensitive enough to make tertiary neutron measurements possible for targets larger than this. The tertiary neutron technique will work to fuel $\langle \rho R \rangle$ values well over 1 g/cm^2 , thus providing a method for measuring even the largest targets likely to be used at Nova.

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Areal Density Measurements of Rubidium-Doped Glass Pushers. The measurement of the neutron-averaged areal density ($\langle \rho \Delta R \rangle$) of the compressed

pusher of an ICF target by neutron activation is well-established. It is implemented by activating one or more elemental constituents of the pusher by 14-MeV neutrons produced in an implosion. Pusher debris containing these activated atoms is then collected on metal foils placed near the target. These foils are transferred to a radiation detector that measures the individual activities. The $\langle \rho \Delta R \rangle$ is determined from

$$\langle \rho \Delta R \rangle = \frac{CA}{\sigma Y N_0 f \eta_c \eta_D e^{-\lambda t_d} (1 - e^{-\lambda t_c})}, \quad (100)$$

where

- C = number of detected counts,
- A = average atomic weight of a pusher atom,
- σ = activation cross section,
- Y = neutron yield,
- N_0 = Avogadro's number,
- f = fraction of atoms of the isotope to be activated compared to the total number of pusher atoms,
- η_c = fraction of activated atoms collected,
- η_D = detection efficiency,
- λ = decay constant,
- t_d = time to load the detector,
- t_c = counting time.

Previous glass shell $\langle \rho \Delta R \rangle$ activation measurements were made on glass microshell targets by using the $^{28}\text{Si}(n,p)^{28}\text{Al}$ reaction and by using the radioactive tracer ^{24}Na to determine the collection fraction. The principal deficiency of this technique is that one must assume that the same fraction of ^{28}Al is collected as ^{24}Na .

An extension of this method that does not suffer from this difficulty was used recently on Nova experiments. This method takes advantage of the 4 at.% rubidium atoms distributed uniformly throughout the glass. Natural rubidium has two stable isotopes, ^{85}Rb and ^{87}Rb , having natural abundances of 72.17 and 27.83%, respectively. Both isotopes undergo $(n,2n)$ reactions with 14.1-MeV fusion neutrons resulting in the activation products $^{84,84m,86,86m}\text{Rb}$. The known properties of these reaction products

Table 3-26. 14-MeV (n,n) activation characteristics of rubidium.

	⁸⁵ Rb		⁸⁷ Rb	
Natural abundance (%)	72.17		27.83	
Threshold (MeV)	10.61		10.03	
Reaction product	⁸⁴ Rb	^{84m} Rb	⁸⁶ Rb	^{86m} Rb
Cross section (mb)	1447 ± 72	341 ± 21	1170 ± 59	
Half-life	32.77 d	20.5 min	18.82 d	1.02 min
Gamma energy (keV)	882	216	1077	556
Gamma intensity (%)	74	32.6	8.79	98.2
Gamma energy (keV)		248		
Gamma intensity (%)		64.5		
Gamma energy (keV)		465		
Gamma intensity (%)		32.1		

are listed in Table 3-26. Only the isomers ^{84m}Rb and ^{86m}Rb have half-lives short enough to allow reasonable counting rates.

The radioactive isotope ⁸⁶Rb ($t_{1/2} = 18.8$ d) was used as a tracer to determine the collected fraction of the rubidium isotopes. Thus the same element is used for the collection fraction measurement as for the $\langle\rho\Delta R\rangle$ measurement. The ⁸⁶Rb tracer was produced in the target by irradiation using thermal neutrons (3×10^{13} n/cm²·s) for 30 min in a small research reactor. By comparing the ⁸⁶Rb activity in the target before the implosion to the activity on the collector foils, the fraction of rubidium that was recovered can be determined. In initial experiments, between 2 and 5% of the rubidium was collected. The neutron irradiation can also be used to accurately assay the rubidium content in the glass.

A pair of high-purity intrinsic-germanium well detectors is used to count the gamma rays emitted by the collected rubidium. These detectors have an active volume of 30 cm³ each and are operated in a face-to-face geometry with the collector foils contained entirely inside the 0.5-cm diam, 1.5-cm deep wells, thus providing 4π solid angle. Absolute detector efficiencies have been estimated to be 5% for ^{84m}Rb and 7% for ^{87m}Rb. Background contributions come from natural background and from Compton-scattered photons emitted by other activities in the glass shell (predominantly ²⁴Na and ⁴²K). Therefore, the $\langle\rho\Delta R\rangle$ -yield product necessary to produce 100 ^{84m}Rb counts is approximately 5×10^6 .

Author: S. M. Lane

Fig. 3-235. Three-dimensional CADD model of the Nova ten-beam chamber.

Facilities

Target Area for the Ten-Beam Laser.

Target Chamber CADD Model. Our model for the Nova target chamber shown in Fig. 3-235 is a 3-D, wire-frame, CADD representation of the ten-beam target chamber, its beam paths, and re-entrant diagnostics. The spherical geometry of the Nova chamber and the complexity of devices occupying space within the chamber make it difficult to visualize clearances between mechanical components and beam lines. The Nova main beams and the lines of sight of other instruments must be kept clear. It is also important to keep track of unconverted laser light that enters the chamber

because this light represents as much as one-half of the total energy and can cause damage directly or through reflections. To further add to the complexity, many instruments are mounted to manipulators that must be prevented from colliding or blocking lines of sight. The Computervision computer-aided drafting and design (CADD) model has become a valuable tool for keeping track of the space available within the chamber and for identifying potential interferences among existing instruments, lines of sight, and proposed experiments.

As the complexity of the diagnostics on the target chamber increases, so does the model data base. Instruments now occupy most of the 148 ports on the chamber, with diagnostics being changed or added weekly. The model data base now contains more than 5000 entries of lines, circles, points, and curves to describe the geometry of the chamber and its instruments. It has the capacity of displaying 255 different layers, with each layer containing one or more instruments. The layers may be represented in any of 33 colors to aid in visualizing the complex system. The user can easily manipulate the model by rotating it on any axis and by placing the vantage point at any location for the best display. For instance, it is especially useful to view from chamber center and look outward

to identify problem areas or likely places that can be used for new instruments. The model can also automatically search for and identify interferences.

The Nova interior model has been in use for two years and is continuing to provide valuable design support to the Nova program. We are developing a similar model for the upgraded two-beam target chamber. These tools will serve as stepping stones to the more complex systems of the future.

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Major Contributor: M. A. Johnson

Chamber Collision-Avoidance System.

The Nova ten-beam target chamber has 20 devices that can be moved inside the chamber. We developed a programmable logic controller (PLC) system to monitor the status of these devices and to restrict their movement to prevent damaging collisions.

The system configuration is shown in Fig. 3-236. A Square D model 300 PLC was chosen for this system because of its programmable flexibility and wide use in the ten-beam target bay to control x-ray streak cameras. Two remote input/output racks were connected to the PLC with capability for 128 inputs or outputs in each rack. All

Fig. 3-236. Block diagram of the Nova collision-avoidance system for the ten-beam target chamber.

of the input/output points and a 24-V power supply were connected to the chamber devices through a central cross-connect panel. The devices that are currently interlocked by our collision-avoidance system are also shown in the figure.

An IBM personal computer is connected via an RS-422 serial communications link to the PLC. The personal computer serves as both the programming terminal for the PLC ladder-logic software and as an operator interface displaying the current status of the 20 devices and their interlocks.

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Upgrade of the Short-Pulse Laser. We have substantially extended the capabilities of our short-pulse laser facility with the addition of a new laser system. This laser will make it possible to study an entirely new area of high-intensity laser-matter interactions. We will also be able to develop and calibrate diagnostics and to perform physics experiments that would have previously been possible only on the larger Nova and Janus facilities.

Table 3-27 shows the design specifications of the new laser over two phases. Phase I was operational in December 1987; Phase II is projected to be operational by December 1988.

The design and requirements of the laser together with a schematic diagram of the system are discussed in greater detail in Ref. 385. Briefly, the laser consists of both an amplified dye laser and an amplified neodymium-doped yttrium-aluminum-garnet (Nd:YAG) laser (see Fig. 3-237). The system is unique in that the dye pulse and Nd:YAG pulse are derived from cavity-matched, mode-locked oscillators, making it possible to synchronize the outputs of the two amplifiers to within a few picoseconds.

The system begins with a continuous-wave mode-locked Nd:YAG oscillator producing 100-ps pulses at a 125.12-MHz repetition rate and an average power of 10 W. This pulse train is frequency-doubled in a 5-mm potassium titanyl

Table 3-27. Operating specifications for the short-pulse laser facility.

Wavelength (nm)	Original laser	New laser	
		Phase I	Phase II
Maximum pulse energy			
1064	700 mJ	300 mJ	3.0 J
532	320 mJ	200 mJ	1.6 J
355	—	100 mJ	1.0 J
266	—	—	0.6 J
570 to 620	2.4 mJ	5 mJ	10 mJ
285 to 310	0.6 mJ	1 mJ	3 mJ
198 to 210 ^a	0.1 mJ	0.1 mJ	1 mJ
Minimum pulse width			
1064	8 ns	100 ps	100 ps
532	5 ns	70 ps	70 ps
570 to 620 and harmonics	1 ps	2 ps	160 fs
Maximum repetition rate (Hz)			
	10	10	1 ^b
Beam quality			
1064	Multimode	Diffraction limited	Single mode
570 to 620	Single mode	Diffraction limited	Single mode

^a Using the nonlinear crystal beta-barium borate (β -BaBO₄).

^b Maximum repetition rate = 1 pulse per minute when using the 5-cm glass amplifier.

phosphate (KTP) crystal to produce an average power of 1.3 W at 532 nm. The residual 1064-nm light is used to seed a regenerative Nd:YAG amplifier system. This amplifier produces 100-ps, 10-mJ pulses at a 20-Hz repetition rate. The pulses are then double-passed through an additional 9-mm Nd:YAG amplifier producing 250-mJ pulses at 10 Hz. The 1064-nm light, or light up to the fourth harmonic, may be used directly for experiments.

The 532-nm output from the mode-locked Nd:YAG oscillator is used to synchronously pump a linear-cavity dye oscillator. The dye oscillator produces 2-ps pulses, which are continuously tunable from 570 to 620 nm (this range may be extended by using additional laser dyes). The output of the dye laser seeds a four-stage amplifier, which is pumped by 40 mJ of the second-harmonic output of the Nd:YAG amplifier system. The 532- or 355-nm output of the Nd:YAG regenerative amplifier is an ideal pump source for the picosecond dye amplifier because the short duration of the pump pulse (<70 ps) significantly reduces amplified spontaneous emission in the dye amplifier.

- DL1 through 4 = delay lines to synchronize
70-ps pump and subpicosecond dye pulse
- L1 through 7 = lenses
- R1 = 4- x 65-mm Nd:YAG rod
- R2 = 9.5- x 115-mm Nd:YAG amplifier
- SHG = second-harmonic generation
- WP1 through WP4 = Wave plates
- PC1 through PC2 = Pockels cells

Fig. 3-237. Amplified, mode-locked Nd:YAG dye laser system.

In addition, the transverse electromagnetic (TEM_{00}) output of the Nd:YAG laser system permits the use of efficient longitudinal pumping in the dye amplifier while maintaining near diffraction-limited beam quality.

By deriving both the tunable-dye and 1064-nm laser pulses from dependent, cavity-matched oscillators, we can use optical rather than electrical techniques to synchronize the output to within a few picoseconds. Such synchronization makes possible an entirely new class of physics experiments in which plasmas produced by the 100-ps output of the Nd:YAG amplifier can be probed on a picosecond time scale by the output of the dye laser. In addition, the high repetition rate of the system permits much simpler and more rapid calibration and development of diagnostics than is possible on our larger systems.

Phase II, which was scheduled to begin in October 1988, will include installation of a 5-cm silicate-glass amplifier, installation of two additional dye-amplifier stages, and development of a 100-fs dye oscillator. The final upgrades will make it

possible to produce focused intensities exceeding 10^{17} W/cm², making the short-pulse laser system one of the brightest light sources constructed to date.

Author: M. D. Perry

Nova Target Fabrication

Introduction

The Nova Targets Group is tasked with providing targets for Nova experiments and with developing new methods for producing materials unique to obtaining data pertinent to inertial confinement fusion. This year we developed some unique materials for examining laser-material interactions.

We developed a process for producing thin foils of two components that are immiscible. In the following article, we describe the use of a dual-source sputtering system to provide uniform mixtures of gold and beryllium. The mixtures are

difficult to make because the gold is the minor component. Alloys of these two elements did not occur at the required ratios; thus, we developed processes to atomistically mix the two at conditions that limit the surface mobility of the gold atoms. Using a dual-source system for magnetron sputtering deposition, we can provide gold-doped beryllium foils at ratios of less than 1:1000 to greater than 1:100 with measured gold clusters of less than 10 nm.

In the area of development for future direct-drive target requirements, we contracted with Airglass (Lund, Sweden) to provide a method for producing hollow shells of a low-density foam material known as an aerogel. The low-density foams would be used to hold liquid-hydrogen isotopes in a uniform, hollow layer. Airglass is using coaxial, dual-orifice jets to produce hollow shells of the aerogel precursor. In the article, "Cryogenic Aerogel-Foam Targets," later in this section we describe some of the chemistry and processes being developed to produce this new type of target.

In our search to find improved methods for designing and documenting Nova targets, we installed a computer-aided drafting and design system. With this system, which is discussed in the article, "CADD System for Nova Targets," we can provide each assembler and experimenter with a complete set of target drawings. The system includes the ability to produce isometric and 3-D views so that we can check assembly procedures and interferences. Because target designs change rapidly, our system has permitted much more interactive design documentation of the targets among theorists, experimenters, and target fabricators.

Author: C. W. Hatcher

Beryllium Foils Doped with High-Z Materials

Introduction. We have been developing techniques that will dilute beryllium with selected high-Z elements. As discussed in the article, "X-Ray Emission from Gold and Beryllium Mixtures"

earlier in this section, we used foils of gold-doped beryllium created from our development efforts in a study of x-ray conversion efficiency. The use of foils with variant high-Z concentrations allows the experimenter to alter the electron- and radiation-transport properties of the laser-produced plasma, while the atomic physics remains constant. Beryllium is employed as a weak radiator to dilute the strongly radiating gold.

In a supplemental study, we also examined the hydrodynamics of the targets. As the laser beam irradiates the surface, large shock waves may be generated. The rate at which the waves travel in the material is an indication of the pressure generated by the laser pulse.

Such physics experiments ideally require the production of full-density foils ($>20\text{-}\mu\text{m}$ thick) with selected high-Z elements that are dispersed on an atomic scale within a beryllium matrix. Unfortunately, the solid solubility of any element in beryllium is rather restricted.³⁸⁶ Low substitutional solid solubility in α -Be hexagonal close pack (hcp) is due to extensive electron overlap along the basal plane. In addition, interstitial solid solubility in beryllium is also restricted. The interstitial hole sizes in the hcp structure are similar to those of the high-temperature β -Be face-centered-cubic (fcc) lattice. The diameter of these sites ($0.25\text{ \AA}$) is far too small to accommodate potential interstitials.

Figure 3-238 shows that only copper, nickel, cobalt, silver, and lead display solid solubility of more than 1 at.% in the hcp structure. For the material of current experimental interest, gold, Fig. 3-238 shows that the equilibrium solid solubility is limited to 0.3 at.% in the α -Be phase and to only 2 to 3 at.% in high-temperature ($>1534\text{ K}$) β -Be. Therefore, for foils that require high dopant levels of gold ($>0.3\text{ at.}\%$), techniques must be either developed to produce metastable supersaturated solutions, or the use of a 2-phase material must be allowed. Although dispersion on an atomic scale is preferred, 2-phase material has been determined to be experimentally acceptable for the x-ray conversion-efficiency study if the minor phase is small ($<100\text{ \AA}$) and

Fig. 3-238. Solid solubility of elements in α - and β -beryllium.³⁸⁶

uniformly distributed within the major beryllium phase. Unfortunately even these relaxed requirements may be impossible to attain for high dopant concentration in beryllium. We are currently exploring the concentration limits.

A variety of techniques can be employed to create foils with extended solid solution, metastable phases, or fine dispersions. The techniques encompass a range of approaches from deposition (electrolytic, chemical, and vapor) to direct quenching from the liquid (splat, melt spinning, and directed-energy surface processing).³⁸⁷ Unfortunately, the use of beryllium as one component in the alloy excludes several of these options. Beryllium exhibits a strong affinity for oxygen and has an electrode potential higher than that of hydrogen, thereby rendering deposition processes, such as electroplating and chemical vapor deposition, ineffective for the production of even unalloyed metallic films.^{388,389} Although we are currently exploring the possible use of direct quenching from the liquid state, physical vapor deposition appears to offer a solution to our unique requirements.

Physical vapor deposition finds its basis in two principal processes: evaporation

or sputtering. Of these, sputtering is the sole, reasonable alternative. Although evaporation is a proven effective technique for the deposition of beryllium films,³⁹⁰ it does not readily lend itself to the production of alloys. Two evaporation sources can be used, but it is difficult to simultaneously maintain constant evaporation rates from both sources, thereby creating concentration gradients in the deposited foil. Our work employs cosputtering because of the inherent versatility gained by the ability to control multiple sputtering parameters. These operational parameters can be adjusted to yield the desired microstructures (full density) and are able to produce a wide range of dopant concentrations.

Foil Production. Foils were obtained using the Nova target cosputtering system, which is described in its original configuration in the 1981 Annual Report.³⁹¹ The system was designed to achieve moderate coating rates of compound materials with independently controllable deposition rates from two separate sources. Because beryllium displays one of the lowest sputtering yields of all metals³⁹² and the physics experiments require thick coatings ($>20 \mu\text{m}$), the cosputtering system required subsequent modification. We replaced one gun from Sloan, Inc., by a "high-rate" sputter gun from U.S., Inc., and moved it closer to the substrate, with a final source-to-substrate distance of 5 cm. The relative angular relationship between the two guns remained unchanged. With this configuration, we were able to obtain dc sputter-deposition rates of 15 to 20 $\text{\AA}/\text{s}$ for beryllium and up to 2 $\text{\AA}/\text{s}$ for gold. Even these high rates required coating runs in excess of 2 hr and caused excessive erosion of the targets.

The system geometry and the closeness of the beryllium gun dictates the lateral uniformity of the cosputtered deposit. A coating area of $1.0 \times 1.25 \text{ cm}$ was selected as a compromise between the need for obtaining sufficient material to produce 2-mm target disks from one coating run and an acceptable level of uniformity. From calibration runs, this uniformity was determined to vary by less than 20% of the nominal dopant levels.

Films prepared by physical vapor deposition generally exhibit a preferential growth of grains into columns normal to the substrate surface. This natural propensity for columnar growth is enhanced in beryllium coatings as a consequence of the anisotropic-atomic and self-diffusivity behavior of the hcp structure.³⁹³ Examination of scanning electron microscopy (SEM) photos of typical beryllium coatings often reveals large columnar grains with cavities between columns. Surfaces often have strongly faceted {002} crystallite grains and, to a lesser extent, show formation of platelets in the (0001) basal plane, which eventually develop into petaloid rose patterns.^{393,394} To alleviate these undesirable microstructures, the classic Thornton zone model for coatings predicts that a low total sputtering pressure combined with low substrate temperatures is required to obtain dense, smooth coatings free of these defects.³⁹⁵ Therefore for our investigation, the pressure of the sputtering argon gas was low (<8 mTorr). We gave special attention to substrate cooling not only to address microstructure considerations but also the possible effects that high substrate temperature might have on foils of metastable material. Given that the transformation kinetics of these unique materials is unknown, it is possible that temperature increases due to the natural heating effect of the coating process may transform these materials in situ.

Substrate fixturing consisted of an electrically powered solid-state refrigerator backed by a water-cooled heat sink. This configuration produced initial substrate temperatures below -10°C . We tried several substrate materials and obtained varying degrees of success. In our initial deposition experiments, we used standard glass slides with a NaCl release layer. The gold-doped beryllium films were floated off in water for target fabrication. These foils were difficult to fabricate because of their brittleness and curvature caused by internal stress. Therefore, we abandoned the method in favor of a thin copper substrate. This approach improved heat transfer between the coating and the solid-state refrigerator by removing the low-thermal-conductivity

glass and NaCl elements. An electric discharge machine was used to cut out 2-mm-diam cores with the gold-doped beryllium deposit still attached to the copper substrate. The copper was subsequently removed by chemically etching with continuously flowing, heated ($\sim 50^{\circ}\text{C}$), concentrated nitric acid. Target samples produced by this method were planar and exhibited increased strength. To date, we have produced four mixtures of gold-doped beryllium with nominal gold concentrations of 0.10, 0.85, 4.42, and 4.91 at %.

Foil Analysis. We determined composition based on profiles of thickness distribution from each gun operating alone. The calculation assumes that the interaction between the two operating sputter guns in the cosputtering configuration is negligible. Wet chemical analysis, specifically, atomic absorption spectroscopy, may be an appropriate technique for concentration determination, but our deposition area produced insufficient material for this type of analysis. Selected samples were characterized with a sputter-profiling Auger system. Although the standardless Auger analysis yielded gold levels that were higher than expected, it illustrated that the compositional variation throughout the thickness of the deposit was small.

Figure 3-239 shows measured densities along with calculated densities for the

Fig. 3-239. Measured and calculated density vs different gold dopant levels within a beryllium matrix.

compositional range of interest. The excellent agreement between the measured and calculated density for the 0.10-, 0.85-, and 4.42-at.% samples provides support for our gold concentration determination. The deviation of density of the 4.91-at.% sample from the predicted value can be attributed to porosity in the deposit. Optical microscopy of the 4.91-at.% sample revealed a darker, matt surface finish; the other samples had a silvery, milky appearance. SEM examination revealed that, although columnar growth was not suppressed for any of the mixtures, the 4.91-at.% concentration had an extreme structure.

Figures 3-240(a) and (b) compare SEM microphotographs for the 0.85- and 4.91-at.% samples and show the difference in structure. We believed that there was a loss of thermal contact between the copper substrate and the actively cooled substrate holder. The loss was confirmed by a measured reduction in "heat" loading for the solid-state refrigerator unit.

Because the nature of the dispersion of gold within the beryllium matrix is of critical importance, examination via transmission electron microscopy (TEM) was mandatory. For the two lowest concentrations, our study showed uniform

distribution of a gold-fcc phase within a beryllium matrix. The TEM microphotograph in Fig. 3-241(a) illustrates this structure for the 0.85-at.% concentration, although the results did not vary appreciably between the two samples. In this TEM dark-field microphotograph, the gold dispersion is seen as small ($<10\text{-}\text{\AA}$), bright spots within the large beryllium grain.

Examination of the high-concentration samples reveals a two-phase microstructure with a large, nonhomogeneous phase. Figure 3-241(b) illustrates this behavior with a dark-field microphotograph from the 4.91-at.% sample. Using selected-area diffraction, we determined the bright phase to be a AuBe_3 structure, with grain sizes greater than $1000\text{ }\text{\AA}$. Once again, this structure did not vary appreciably between the two high-concentration samples, although our TEM study confirmed the porosity in the 4.91-at.% sample. This microstructure compromises the x-ray-conversion data obtained from these samples. It is not clear whether this large-grain microstructure represents the limits of our computer technique or is a result of a time-temperature transformation due to in situ heating or heating during the chemical etching process.

Fig. 3-240. SEM photographs showing the different surface structures for (a) 0.85-at.% gold and (b) 4.91-at.% gold.

(a) 0.85-at.% gold

(b) 4.91-at.% gold

(a) Face-centered-cubic gold phase

(b) AuBe₇ phase

Fig. 3-241. TEM dark-field photographs showing microstructure in a beryllium matrix diluted with (a) 0.85-at.% gold and (b) 4.91-at.% gold.

To produce a finer deposition, we are currently repeating the high-concentration depositions using a substrate bias, which has been shown to help eliminate columnar growth and to improve control over the temperature history. However, observation of a two-phase microstructure for a concentration below the solid solubility limits is not a promising sign.

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Cryogenic Aerogel-Foam Targets

Introduction. As discussed by Sacks and Darling,³⁹⁶ high-gain, directly-driven ICF designs at low input energy and power require the formation of a levitated liquid DT layer a few hundred microns thick. Fabrication of targets with uniformly thick layers of liquid DT is quite difficult because the liquid layer, however uniform initially, tends to slump due to gravity. Sacks and Darling proposed a single-shell target design composed of a small-pore, low-density, low-Z,

rigid foam saturated with liquid DT, as shown in Fig. 3-242.

The foam used for holding the liquid layer has many stringent requirements, such as the following:

- Maximum pore size should not exceed 1 μm .
- Foam material should be strong enough to withstand the high capillary pressure generated in small pores by liquid DT.
- Foam should maintain its dimensional stability over a wide temperature range (from 300 to 25 K).

One such foam material appears to be silica aerogel. The chemistry of silica aerogel production is well known and is described in detail in many references.^{397,398} In this article, we briefly describe the technology of silica aerogel production.

Silica Aerogel Material. Silica aerogel and quartz are chemically identical; both are pure silicon dioxide. Unlike quartz however, silica aerogel (hereinafter referred to as aerogel) does not occur in nature, but is made synthetically.

The first step in making an aerogel is hydrolysis of an organosilicic ester, such as tetramethoxysilane (TMOS) or tetraethoxysilane (TEOS). The following two

Fig. 3-242. Use of low-density, low-Z foam to define and stabilize the DT shell (from Ref. 396).

equations represent the chemistry of the reaction:

and

where R—represents an alkyl group, such as methyl— or ethyl—. Equation (101) is catalyzed by either an acid or a base. As described in detail in Refs. 397 and 398, the properties of the resulting aerogel depend, in part, on the type of the catalyst used. Silicon dioxide molecules tend to form aggregates according to the reaction:

The product of the reaction in Eq. (103), called alcogel, is a matrix of silicon dioxide molecule aggregates filled by water and alkyl alcohol. The final step in the production of aerogel is to remove the liquid from the alcogel. Unfortunately, we cannot obtain an aerogel from an alcogel by conventional drying techniques. Any attempt to do so results in collapse of the capillary wall and severe gel shrinkage. (The final product obtained in this manner is called xerogel, which is

significantly denser than aerogel.) The phenomena of collapse and shrinkage are caused by a combination of high surface tension of water and small pores ($\sim 500 \text{ \AA}$) in the alcogel, which gives rise to high capillary pressure inside the pores and results in gel damage. Instead, we must resort to supercritical drying, where the solvent inside the alcogel is first taken above its critical temperature and pressure, thus eliminating any vapor-liquid interface. The solvent, in fluid state, is then slowly removed from the alcogel, resulting in little gel shrinkage and yielding aerogel. The principles and details of the supercritical drying process are given in Ref. 398.

Fabrication Technology. A variety of alternative approaches are possible for making hollow, spherical, ICF aerogel targets. For example, we could machine or mold the hemispheres and then bond them into a spherical shape; however, the small target sizes we require make both machining and molding quite difficult, if not impossible.

A third alternative has been used for glass and polymeric materials at LLNL and elsewhere (see, for example, Refs. 399 and 400). Airglass (Lund, Sweden), which is a commercial manufacturer of the bulk aerogel material, is currently under contract with LLNL to develop targets by techniques similar in concept (but quite different in details) to those

described by Campbell³⁹⁹ and Torobin.⁴⁰⁰ Here, we describe the work being performed at the Airglass research facility near Lund.

The chemical formulation currently used by Airglass is essentially the same (but with proprietary changes) as that described by Henning.⁴⁰¹ Figure 3-243 shows the experimental setup. The gelling solution (TMOS, water, and ammonia) is thoroughly mixed and stirred while the solution viscosity is continuously monitored. When the solution reaches the predetermined viscosity, it is transferred to the small tank on top of the nozzle assembly. The assembly consists of two concentric nozzles: an inner nozzle for gas flow and an outer for the gelling solution. The solution from the tank and gas from the inner nozzle flow simultaneously, thus the emerging liquid forms hollow droplets at the outlet.

The liquid and gas flows are adjusted to obtain a steady stream of hollow droplets, which are collected in a liquid bath below the nozzle. The time of flight and the catalyst concentration are adjusted so that the solution flows freely out of the nozzle but becomes a viscous gel during the fall. This procedure helps minimize the effect of impact of the droplet with the bath liquid. Parameters that contribute to the size and shape of the collected droplets are rates of gas and liquid flow, nozzle diameters, amount of catalyst and water relative to TMOS in the gelling solution, concentration of catalyst in the atmosphere through which the droplets fall, and time of flight. According to available evidence,^{399,400} certain combinations of these factors can yield nearly spherical droplets with uniform wall thickness. The final step is to dry the hollow microspheres supercritically.

Current Status and Future Plans. At present, most particles collected in the liquid bath using the drop-tower technique are nonspherical. They vary in terms of outer-diameter size, wall thickness, and inner sphere concentricity. Many particles appear to be hemispheres attached to irregular shapes. This irregularity may indicate that the corresponding droplets were spherical or nearly so

Fig. 3-243. Drop-tower arrangement and nozzle assembly used by Airglass.

as they approached the liquid bath, but were distorted upon impact with the hard liquid surface, thus forming a foot at the point of impact. The inner bubble appears to be spherical in most cases, suggesting that the inner shell is better protected from impact than the outer shell. This result may indicate that the inner shell is stronger because of a higher gelling rate caused by the catalyst in the inner-nozzle gas.

Our short-term plans for the future are directed toward increasing the gelation rates and softening the droplet impact. Airglass plans to increase the catalyst concentration in the liquid phase, the inner-nozzle gas, and the gaseous atmosphere through which the droplets fall. Such modifications should accelerate the gelling process in the bulk and at the outer and inner skins. Another planned design modification is to increase the residence time of the droplet in free-fall to further the extent of the gelling reaction. A "water-slide" collector is currently being designed so that gelled droplets land on the liquid surface tangentially.

Although we have not yet produced target-quality shells, the products obtained have demonstrated the possibility

of using the drop-tower technique to provide future targets. We will evaluate the improvements needed to continue progress in this difficult technology because it may well prove to be amenable to producing targets for future power reactors.

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CADD System for Nova Targets

We have implemented a computer-aided drafting and design system in the Nova targets assembly area. Our main purpose was to provide more consistent documentation of requested and constructed targets. In addition, we can now make rapid changes to a catalog of target types without the necessity of beginning a completely new drawing.

We selected a personal-computer-based system for CADD, which includes capabilities for full 3-D and shaded renderings of targets. An additional feature of the system is that it provides archival storage for all targets fabricated and shot at Nova. The output plots of drawings can now be standardized so

that our assemblers have all pertinent information in a readable format. We can provide the assemblers with a worksheet for recording the actual, as-built target dimensions and for assisting in record-keeping in future data analysis of shots. A consistent, up-to-date database of this type is essential in understanding target performance.

A catalog of targets and target components is now available for the laser experiments and advanced diagnostics (LEAD) group and assembly staff. From this catalog, we can "build" a target from a list of parts and specify changes to be made from the original specifications. Drawings for the new targets can be assembled from the parts list and the changes incorporated into the design.

The CADD system is located in our assembly area, but we also have plans to add a similar system in the vicinity of the LEAD experimenters. A fiber-optic computer link between the two systems would provide an even more rapid method for changing target designs.

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Target Materials Development

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Target Materials Development

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Target Materials Development

Introduction

S. A. Letts

We are developing foams for the direct-drive inertial confinement fusion (ICF) target that is described in the *Laser Program Annual Report 85* (Ref. 1) and in a paper by Sacks and Darling.² This target design calls for a spherical shell of foam that can be wetted with liquid deuterium-tritium (DT). The liquid DT is both fuel and ablator: the foam holds the DT in a spherical-shell configuration that is compressed to high density and ignited by laser energy.

The foam should have a density of 50 mg/cm^3 or less and be composed mainly of carbon, hydrogen, and oxygen. In addition, the foam should have a cell size of less than $4 \mu\text{m}$ to meet uniformity requirements and a pore size of less than $1 \mu\text{m}$ to stably hold the liquid DT. The foam must be machinable or moldable to the required spherical-shell configuration and tolerances. The foam must tolerate exposure to the radiation environment of tritium and to cryogenic temperatures without changing dimensions during the time needed for the target to fill and stabilize before a shot. Some target designs will require a foam with an overcoat of approximately $5 \mu\text{m}$ of a hydrocarbon or other low-Z material.

In 1986, two foam materials were under development: polystyrene (PS) foams made from an inverse emulsion system and resorcinol-formaldehyde (RF) foams made from condensation polymerization gels. Our work in 1987 continued the development of these foams with more processing and property-measurement experiments. Our goals were to reduce density and cell size, improve uniformity, develop techniques to measure uniformity, test foams for performance under target conditions, and improve foam processing reproducibility. We also started an investigation of a new foam material, cellulose acetate (CA), which is made by a phase separation technique.

In the first article of this section, "Polystyrene Foams," we discuss our continuing development of these materials. This year we concentrated our efforts on perfecting the formulation, determining the best polymerization conditions, investigating the drying process, assessing alternative homogenization techniques, and using statistically designed experiments to determine the reproducibility of foam preparation. PS foams can now be prepared with good uniformity at a density of 85 mg/cm^3 and a cell size of 2 to $3 \mu\text{m}$ using our syringe-pump mixing apparatus. Foams with densities of 50 mg/cm^3 and cell sizes of 3 to $10 \mu\text{m}$ can be made using a Brookfield counter-rotating mixer, but the uniformity is not as good.

We have also pursued the development of RF foams because they offer a much smaller cell size than PS foams, which should mean greater DT-fill stability. In last year's initial efforts, we obtained RF foams with a density of 150 mg/cm^3 only with great difficulty. We can now consistently produce RF foams with a density of 20 mg/cm^3 . The reduction in the density is a result of our better understanding of gel formulation and processing.

We have recently taken on an additional avenue of research in the RF work. In an attempt to remove the small oxygen content of the foam and improve the mechanical properties, we have investigated carbonization. When heated to 1100°C in an inert atmosphere, RF foams lose hydrogen and oxygen to form a glassy (vitreous) carbon that is totally amorphous. Some shrinkage occurs during carbonization, but a foam density of 41 mg/cm³ has been reached. All of these developments are discussed in the article "Resorcinol-Formaldehyde Foams," later in this section.

In the article "Cellulose Acetate Foams" we discuss the investigation of this new foam material, which was first developed by R. W. Pekala. Our work has concentrated on studying the preparation and properties of the CA gel and on preparing CA foams by supercritical extraction. We have made foams with densities of 30 to 150 mg/cm³ with an estimated cell size of 0.1 to 1 μ m. The reproducibility of foam preparation with respect to density is very good.

In the final article in this section, "Target-Related Foam Properties," we discuss target specifications and foam properties related to target production. We have measured foam density and density uniformity, wetting properties, hydrogen capillary pressure, machining properties, thermal contraction, and bonding properties. We have investigated molding of foams as a technique for producing materials more rapidly than by machining. We assess the advantages and disadvantages of each foam as a potential target material.

The quality of foam materials has greatly improved during this last year, and we now have three foam materials that have the potential to meet target specifications. We reduced density and improved density uniformity. We improved characterization of foams through new scanning electron microscopy (SEM) and transmission electron microscopy (TEM) techniques, thermal analysis from room temperature to 15 K, and viscosity measurements of gels and emulsions. We assessed the feasibility of producing hemishells by diamond turning and grinding. We developed molding techniques for all three foam materials. In the future, our efforts will focus on improving foam preparation, measuring foam properties, comparing the foam materials, and developing target assembly techniques.

Polystyrene Foams

Introduction

We are continuing to develop PS foams using an inverse-emulsion system originally developed by Unilever Research Laboratory.³ A water-in-oil, high-internal-phase emulsion is made by using a surfactant such as sorbitan monooleate (Span 80) and a water-in-oil ratio of 90/10 to 95/5. The oil phase is a 50:50 mixture of styrene and divinylbenzene (DVB) monomers, and the surfactant is

usually 20 wt% of the oil phase. The DVB is added to increase the strength of the foam through cross-linking. The water phase contains sodium persulfate as a polymerization initiator. After polymerization, the resulting foam is dried in a vacuum oven to yield a material with a density of 50 to 100 mg/cm³. The structure of the foams and the details of the process for making them are described in Ref. 4. Previous development work is described in Ref. 5.

We have continued our studies of the chemistry of this system by assessing alternative surfactants and initiators. We have examined what effect adding aluminum

chloride has on foam structure. We have investigated the relationships between PS molecular weight and initiator concentration and temperature, and the effects of polymerization accelerators.

The polymerization has been followed using both optical microscopy and viscometry. We have carried out several studies of the foam drying process, and we have investigated methods for removing the surfactant and initiator from the foam.

Processing methods continue to be a focus of investigation. We have assessed the use of a peristaltic pump for mixing, and we have constructed and tested a new syringe-pump apparatus.

A statistically designed response surface methodology experiment provided us with an assessment of key variables and of the reproducibility of the foam-making process. The results led to a continuing study of reproducibility, which has as its goal the production of large numbers of samples for tests that will assess potential success as a target material. The results of these tests are described in "Target-Related Foam Properties" at the end of this section.

Emulsion Chemistry

We continue to use Span 80 as our surfactant since it has proven superior to the others we have tested. We briefly investigated the use of block copolymer surfactants, which were supplied to us by D. J. Meier of the Michigan Molecular Institute. These were compounds composed of blocks of polystyrene and polyethylene oxide. We thought they might be effective at stabilizing the emulsion and reducing the cell size because of the similarity of their lipophilic components to styrene. However, emulsions were not formed, and the systems separated into two phases. It is possible that this occurred because the amount of polyethylene oxide was too large, making the surfactants too hydrophilic. Since Span 80 gives satisfactory results, we have chosen not to pursue this approach further.

Although sodium persulfate acts effectively as a polymerization initiator in this system, it would be desirable to replace

it with a material of lower atomic number. We tested a variety of organic polymerization initiators, and we also tried some polymerizable surfactants, but these initiators and surfactants produced foams of low mechanical strength that have large cells with nodular walls. It seems likely that it is necessary to have a water-soluble rather than an oil-soluble initiator to ensure that the polymerization occurs at the interface rather than within the oil phase. We will continue to use sodium persulfate and will investigate the possibility of removing it from the foam after polymerization and drying if necessary.

We have also continued our investigation of the addition of aluminum chloride to reduce emulsion cell size. When an Osterizer blender was used, the addition of an optimum concentration of 0.01 wt% AlCl_3 caused a reduction in the cell size of the resulting foam to 2 to 3 μm , compared to a control sample with a cell size of 3 to 7 μm . However, when we tried this experiment with the syringe pump as the mixing device—which produces a foam with a cell size of 2 to 3 μm without AlCl_3 —no further reduction was observed. It is likely that geometric constraints prevent any reduction beyond the 2 to 3 μm limit.⁶ We will investigate the effect of AlCl_3 addition on foams made in Brookfield counterrotating mixers to see if the effect observed in the blender occurs with this mixing device, which also produces larger cells. We have preliminary indications that the addition of AlCl_3 may improve the uniformity of foams made at the lower end of our density range.

In *Laser Program Annual Report 86* (Ref. 7) we described an experiment designed to study the effect of polymerization temperature and initiator concentration on various foam properties. Four initiator concentrations (3.0, 6.2, 11.9, and 17.6 g/L) and three polymerization temperatures (40, 50, and 70°C) were used. (The initiator concentrations were incorrectly given in Ref. 7.) These foams were made without any DVB so that the molecular weights could be determined using gel permeation chromatography. Molecular weights are important because they may be correlated with foam strength.

Table 4-1. Analysis of PS polymerization as a function of temperature and amount of initiator.

Temperature (°C)	Initiator (g)	Surfactant (g)	M_n^a	Exp. ^b DP	Calc. ^c MDP	Active initiator ^d (%)	Active initiator (g)	Increase ^e (%)
40	0.60	15.0	144 730	1390	57	4.5	0.025	—
40	1.22	15.0	96 811	930	28	3.0	0.037	48
40	2.40	15.0	74 810	718	14	1.9	0.046	84
40	3.61	15.0	61 500	591	9.5	1.6	0.058	132
50	0.60	15.0	83 600	803	57	7.1	0.043	—
50	1.22	15.0	61 100	587	28	4.8	0.059	37
50	2.40	15.0	38 100	366	14	3.8	0.091	111
50	3.61	15.0	32 000	307	9.5	3.1	0.112	160
70	0.60	15.0	30 500	293	57	19.4	0.116	—
70	1.22	15.0	22 465	216	28	13.0	0.159	37
70	2.40	15.0	18 150	174	14	8.0	0.192	65
70	3.61	15.0	15 500	149	9.5	6.4	0.231	99

^a Number average molecular weight.^b Experimental degree of polymerization = $M_n/104$.^c Calculated minimum degree of polymerization = moles styrene/moles initiator.^d Percent active initiator = (Calc. DP)/(Exp. DP).^e Percent increase in active grams of initiator, based on grams used from the 0.60-g run at that temperature.

The results are shown in Table 4-1. The molecular weights decrease with increasing amount of initiator and with increasing temperature. It is clear that the amount of active initiator increases with increasing temperature. This is most probably because the initiator breaks down into radicals faster at higher temperatures. At a given temperature, the percent of initiator that is active decreases with increasing concentration. This may be related to the fact that the initiator is in the aqueous phase, and there are a limited number of interfacial sites where it can initiate.

The mechanism of a free-radical polymerization consists of four steps, which are shown below.

The first step is the dissociation step, where the initiator dissociates into radicals. This is followed by an initiation step, in which the radical reacts with the monomer, and then by a series of propagation steps in which the polymer chain grows through propagation of the radical species. The chain growth will terminate

if two chain ends find each other. The standard steady-state treatment of this mechanism leads to the following mathematical relationship between the degree of polymerization and the initiator concentration:

$$X_n = \frac{k_p}{(k_t k_d)^{1/2}} \times \frac{[M]}{[I]^{1/2}} \quad (1)$$

The degree of polymerization should be proportional to $[I]^{-1/2}$. Figure 4-1 shows that this relationship is followed by the data from this experiment. This evidence is consistent with the free-radical mechanism. (It is likely that other forms of termination are also possible, given the large number of reactive species present, but they must be consistent with these results.)

We have assumed that it is desirable to polymerize the oil phase of the emulsion as fast as possible to minimize emulsion coalescence, which increases the cell size. The polymerization rate can be increased by raising the population of free radicals. This may be accomplished in three ways:

1. Increasing the concentration of initiator (the effects of which have just been described).
2. Increasing the temperature (which also increases the rate of coalescence).
3. Using an accelerator that increases the rate of dissociation of the initiator.

We have tried two accelerators: ferrous sulfate and N,N,N',N' -tetramethylethylenediamine. Several concentrations of both accelerators were tried at a series of temperatures. The emulsions were homogenized in a peristaltic pump. As with the previous experiment, no DVB was added because we wanted to be able to measure the molecular weights of the foams to assess the effect of the accelerator. The molecular weights of the foams were all in the range of 20 000 g/mol, regardless of the temperature or accelerator concentration. We now believe that the accelerator concentration was too high and that polymerization occurred too rapidly, resulting in low molecular weights. The emulsions became extremely viscous during homogenization, and we believe this is because the oil phase was partially polymerized. Since more recent experiments indicate that the effect of coalescence at 50°C may be minimal, this approach will not be pursued further until the extent of coalescence is clearly established.

Polymerization Studies

In order to improve our understanding of the polymerization process, we prepared an emulsion, using the Brookfield

counterrotating mixer, and removed samples at 20-min intervals after placing the emulsion in a water bath at 50°C. The oil phase of the polystyrene emulsion consisted of 40 wt% styrene, 40 wt% DVB, and 20 wt% sorbitan monooleate. The calculated density was 50 mg/cm³. We examined these samples under an optical microscope. Figure 4-2 shows the emulsion before it was placed in the water bath and the sample that was removed after 1 h 40 min, which was the time when solidification was first observed. Areas of solid polymer appear to be forming, but the emulsion shows no substantial signs of coalescence. We took samples up to 3 h 20 min, at which time the system had become too solid to sample, and we observed no substantial coalescence through this time.

Fig. 4-1. Relationship between degree of polymerization and initiator concentration for PS foams.

(a) Zero time

(b) 1 h 40 min

Fig. 4-2. Optical micrographs of polymerizing PS emulsion: (a) zero time; (b) after 1 h 40 min at 50°C.

In order to confirm these results and to determine the rate of polymerization when no initiator is present, we repeated this experiment, replacing the initiator with an amount of sodium sulfate that would give the same ionic strength. In this case, it was 4 days before the emulsion began to polymerize, showing that the rate of purely thermal polymerization is negligible. Once again, we observed no substantial coalescence even over this much longer time.

It may be that the relatively large cells of emulsions prepared with the Brookfield mixer makes them resistant to coalescence or that the surfactant forms a liquid crystalline phase at the interface that has a very high surface viscosity that stabilizes the emulsion. In order to resolve this question, we need to confirm the results using emulsions made with the syringe-pump apparatus. It is also possible that the microscope slide-making process may affect the results, although the consistency of structures seen throughout the two experiments leads us to believe that this is not the case.

We have also used viscometry to determine the time to polymerization of PS emulsions at various temperatures. Viscosity measurements lend themselves nicely to this because, during polymerization, the system changes from an emulsion with a mayonnaise-like consistency to a solid. We selected a viscometer that vibrates a glass rod at 600 s^{-1} with a vibration amplitude of $25\text{ }\mu\text{m}$ in air. When this vibrating tip is inserted in a liquid, the viscosity can be determined by the amount of damping measured by a transducer at the top of the rod.

The emulsion formula, calculated density, and mixing method were the same as for the optical microscopy experiment. We poured the emulsion into the heated reaction flask and tamped it down to eliminate voids. We immersed the glass rod of the viscometer to a mark on the rod 1 in. up from the end. We made a cover from aluminum foil to limit the escape of heat and moisture from the cell. To maintain a constant temperature, we set up a temperature control bath to pump a water/ethylene glycol solution

Fig. 4-3. Logarithm of normalized viscosity of polymerizing PS emulsions.

through a jacketed reaction flask. As the emulsion polymerized, a logging multi-meter printed the data points generated by the viscometer. Most runs were for 24 h, with typical recording intervals of 3 to 10 min. The raw data consisted of an uncorrected viscosity measurement, the temperature of the emulsion, the date, and the time.

The viscometer output is the product of viscosity and the sample density. Since the emulsion has a density of approximately 1 g/cm^3 , we read the data directly as viscosities in centipoise. In analyzing the data, we divided all the viscosities by the initial viscosity to eliminate some of the scatter. Since the viscosity changed by several orders of magnitude during a run, we took the logarithm of the normalized viscosity (η/η_0). When the data for all of the runs are plotted versus time (Fig. 4-3), the results show a trend of decreasing time to polymerization with increasing temperature. The greatest increase occurs between 30 and 40°C. Note that the inflection in each curve represents roughly a factor of 30 increase in viscosity.

To verify that the transition did signify polymerization, we made an emulsion with ethylbenzene in place of the styrene/DVB mixture. After 24 h at 50°C, no transition had occurred, and the viscosity decreased slowly with time. Each of the runs was duplicated at least twice with generally the same results. The reproducibility of the time to polymerization at 50°C among three runs was within one hour. As polymerization progresses, the viscosity increases until the material no longer behaves like a liquid, and the viscometer can no longer make useful measurements. Thus, it is not possible to determine the total time period of polymerization with this method. It is clear, however, that the time to onset of polymerization at 50°C agrees well with the time determined in the optical microscopy experiment.

Drying Studies

Once a PS emulsion has polymerized, the water must be removed from the foam structure by drying in a vacuum oven to produce the void space within

the foam. To shorten production time, it is desirable to remove the water as rapidly as possible without damaging the foam. The drying rate may also reveal something about the structure of the foam.

We have found that the water will rust out the mechanical vacuum pumps normally used on the ovens. To solve this problem, we selected a new type of vacuum pump that works on a venturi principle because it would pass the water without any harmful effects. The mechanical pumps would evacuate an empty chamber to a base pressure of approximately 50 mTorr, but the base pressure for the new pump was approximately 7 Torr. We were concerned that the rate of drying would decrease, thus slowing the production rate of PS foams, but initial comparisons between the two pumps yielded similar drying rates. This prompted a study to determine the mechanisms of the drying process and to see if its efficiency could be improved by changing the temperature and pressure.

In order to measure the rate of water removal with time, we placed an electronic balance with a remote display unit inside a vacuum oven. The remote display unit had an RS-232 output, which was fed to a computer for data logging. Logging intervals were typically 10 min.

The oil portion of the PS emulsions used for this experiment consisted of 42 wt% styrene, 42 wt% DVB, and 16 wt% sorbitan monooleate. We emulsified the samples using a counterrotating mixer; the densities were approximately 80 mg/cm^3 . We polymerized the emulsions in 50-ml plastic syringes in an oven at 50°C for 24 h. After polymerization, we sectioned the container with a band saw, leaving cylindrical sections that were 0.5- or 1.0-cm thick with the sides still enclosed in plastic. We placed the samples in the vacuum chamber on the zeroed balance, initiated data logging, and activated the vacuum pump.

Simply viewing the data as weight vs time produces a declining curve that yields little information on the mechanisms that drive the drying process, but one can determine the total amount of time it took to dry the sample. There is initially a sharp increase in the drying

Polyatyrene Foams

Fig. 4-4. Drying rates vs moisture content for 1.0-cm-thick PS foam samples.

Fig. 4-5. Drying rates vs moisture content for 3.0-cm-thick PS foam samples. ▼

rate because of the evaporation of surface water. The rest of the drying process is relatively slow. Inconsistencies in producing the samples caused slight variations in length that do not affect the area through which evaporation occurs, but do increase the volume of water removed. Since this determines the amount of time to complete the drying process, and the purpose of the experiment was to compare the rates of drying, we normalized the volume of water by plotting the drying rate as a function of the ratio of the weight of water to that of dry foam. Since all of the samples were made to the same bulk density, and the ratio of water to foam is constant, the drying rates can be compared. Figure 4-4 shows the results of three runs plotted in this manner.

We made the first run with the oven at ambient temperature and the venturi pump achieving a base pressure of

Fig. 4-6. Sample temperature vs. moisture content for 3.0-cm-thick PS foam samples: (a) room temperature; (b) at 50°C; (c) with nitrogen purge.

8 Torr. The second run was also at ambient temperature but with a mechanical pump, which achieved 200 mTorr. There is very little difference in the two rates, and the total drying time for both was approximately 8 d. The third run used the venturi pump but was made with the oven at 50°C. In this case, the rate of drying roughly doubled, and the samples dried in about 4 d.

The increase in drying rate with temperature is to be expected, because of the increase in the rate of evaporation of water, but the lack of any pressure effect was somewhat surprising. In order to get more information about the process, we attached a vacuum transducer to the oven so that pressure could also be logged with each weight reading. We measured and logged the internal temperature of the foam sample to determine the amount of temperature drop due to heat loss from vaporization.

The emulsion formula and method of preparation were the same as for the previous experiments, but we cast the emulsion in a 250-ml Nalgene container with 30-gauge type-J thermocouples arranged so that the junction was in the middle and the leads exited separately through small holes drilled on opposite sides of the container. Each sample had three thermocouples that we spaced top to bottom so that one was in the center and the other two were 0.5-cm and 1.0-cm in from their respective surfaces. We performed polymerization as before, and sectioned the container into plastic-encapsulated cylinders roughly 3 cm long with two exposed faces 6 cm in diameter. We placed the samples in the vacuum

chamber on the zeroed balance, and placed a fourth thermocouple so that it touched the center of the surface of one end of the sample. We initiated data logging and activated the vacuum pump.

The results of three runs made with this system are shown in Fig. 4-5. The shape of the curves has changed somewhat with these larger samples. The effect of increasing temperature is again observed. The sample that dried out at room temperature took 8 d, and the one that dried at 50°C took 4 d. To test the effect of increasing the flow through the vacuum oven, we ran a sample in which a dry nitrogen gas feed was attached through a needle valve to the drying oven, raising the base pressure from 8 Torr to 11 Torr. This also increased the drying rate: one sample dried in 2.5 d and another in 4 d.

More information can be obtained when the sample core temperature is plotted vs. moisture content, as in Fig. 4-6. In the beginning, the water-to-foam ratio is a little over 11:1 and the sample is still at a slightly elevated temperature. After the vacuum pump is turned on and the water begins to evaporate, temperature throughout the sample drops sharply because of the loss of heat of vaporization. The temperature remains depressed until nearly 80% of the water is removed, which occurs in only about half of the total drying time. At this point the temperature begins to rise from the outer regions of the sample towards the center.

In the case of the sample with the nitrogen purge, the internal temperatures dropped below 0°C, indicating that the water may have frozen. If this is the case,

it will be necessary to determine whether this has any deleterious effects on foam structure. In a preliminary experiment on another vacuum oven system, SEM analysis showed no changes, but this needs to be confirmed.

These temperature measurements and preliminary x-ray radiography experiments on partially dried foams suggest that the water surface is moving from the outside of the foam to the center. Our next step is to develop a model of the drying process that explains this.

Removal of Surfactant and Initiator

As a potential target material, the foam ideally should not contain any substances other than the polymerized styrene and DVB. Therefore, we have tried to remove the Span 80 and the sodium persulfate from the foams.

To remove the Span 80, we have tried two methods. In one, 30 wt% of the Span 80 was removed by heating a dry foam at 85°C in a conventional oven until a constant weight was reached. We have recently determined that the foam can probably survive heating to 115°C, so we will try increasing the temperature to remove more surfactant. In the second method, 90 wt% of the Span 80 was removed by immersing a dry foam in fresh hexane, which was exchanged daily for one week, and then by drying at 85°C. Hexane wetting and SEM tests indicated that the foam retained the same porous cell structure and wettability after treatment. In a second experiment with hexane removal, we used a Soxhlet extractor for 4 d with boiling hexane. However, only about 64 wt% of the Span 80 was removed, and there was some shrinkage of the sample, so that the reduction in density was not as great as expected. Radiography of this sample showed that the extraction was not uniform. We will do more work with both of these methods.

To remove sodium persulfate, we put dry foams in water for different periods of time, redried them, and then determined the concentrations of sodium and sulfur by elemental analysis. After 4 d in water, 99% of the sodium and sulfur was removed. (Although some sulfur should

be bound onto the polymer molecules as end groups, our results indicate that this involves very small amounts.) After 7 d, 617 ppm of sodium and 688 ppm of sulfur remained. Preliminary SEM analysis showed that the foams retained their porous cell structure but that there was a distinct increase in surface roughness. It may be necessary to tolerate the presence of sodium persulfate unless another removal method can be found.

Processing Methods

Preparation of uniform emulsions with small cell sizes requires both the proper formulation and the proper homogenization techniques. The formulation determines the volume fractions of the emulsion phases and the interfacial characteristics. The homogenization apparatus provides both the necessary mechanical energy to break the coarse emulsion into fine droplets and the mixing to eliminate density variations. We have found that streamlined shear is best for our emulsions. Turbulence causes coalescence or inversion of the emulsion by disrupting the cell structure. Low shear produces an emulsion with a large cell size.

A second desirable feature of an homogenizer is the ability to produce material either continuously or in large batches. Our syringe pump is limited to 25-cm³ batches. Random mixing of the emulsion mixture in the syringe pump is not achieved because of the reciprocating nature of the syringe. For these reasons, we continue to look for improved mixing methods, even though the syringe pump currently gives us the best means of achieving target specifications.

Previously we reported on the use of a peristaltic pump to homogenize an emulsion.⁸ The early design, which consisted of a test tube with a side-arm inlet and a bottom exit, trapped air within the emulsion and did not provide random mixing. We improved both the equipment and the procedures for operating the pump this year. To assure uniform density, we first prepared a low-viscosity, large-cell emulsion using a low-shear homogenization technique. We poured the emulsion into a storage vat from which it

was fed by gravity into the peristaltic pump. The vat has a conical shape that is efficient for forcing all the emulsion to flow into the pump; it is made of polyethylene, which is not wetted by the emulsion, allowing the emulsion to flow to the exit. We then returned the output of the pump to the top of the vat where the emulsion flowed smoothly over the contents of the vat without entraining air.

We used this technique of mixing for preparing all the samples in the accelerator experiment. Unfortunately, mixing was hindered because the rates of polymerization were so high that the emulsion began polymerizing inside the pump. We need to assess this method using the standard formulation.

In addition to investigating the peristaltic pump, we also constructed a new syringe-pump device. We did this because the old pump was needed to begin experiments on molding PS and because we felt that several improvements could be made to the design. Figure 4-7(a) shows the apparatus with a load cell to measure the pressure on the syringes, and a thermocouple, which is taped to the outside of one of the syringes, to measure the temperature. This data is recorded on a strip chart recorder during a run. One syringe is filled with the water phase of the formula and the other with the oil phase. After all the air bubbles are removed, the syringes are connected with a plastic fitting and placed in the cradles of the pump. The fit is adjusted and the run started by turning on the air flow to the hydraulic pump. The pressure can be controlled, within limits, by regulating the air flow. A counting device on the carriage is connected to a control box so that the desired number of strokes can be carried out. Fig. 4-7(b) shows the two syringes in place before the start of a run. After just a few strokes, a creamy white emulsion is formed, as shown in Fig. 4-7(c). The new pump has been designated PUSH-ME-PULL-YO-TOO (PMPY2). The original one, now dedicated to molding, is PMPY1. The concept for the syringe pump came from J. M. Williams at the Los Alamos National Laboratory.

Statistically Designed Experiments

In the *Laser Program Annual Report 86* (Ref. 9), we described an experiment that was designed to optimize the formulation of the PS foam and to assess the reproducibility of foam preparation. This experiment was a three-variable, Box-Behnken response surface methodology (RSM)

Fig. 4-7. Syringe-pump mixing apparatus (PMPY2): (a) with load cell, thermocouple, and recorder; (b) syringes filled with oil and water before mixing; (c) syringes showing emulsion formation after mixing.

design with the concentrations of surfactant and initiator and the ratio of styrene to DVB as variables. After preparing the 15 samples in random order, as called for in the design, we looked at several emulsion and foam properties.

Several important decisions had to be made about the factors and their ranges and about certain parameters that had to be fixed. The units and ranges for the factors were chosen empirically as weight percents based on the way the foams are actually formulated. Although it is likely that molar concentrations and more strictly chemical factors may actually be controlling the system, our understanding of the basic chemistry is not sufficient to identify these.

The percents for each factor are on three different bases. For the percent styrene, the range is 0 to 100% of the monomer used, which means that we have foams that are made with 100% DVB, 100% styrene, and a 50:50 mixture. For the initiator, the percent is based on the amount of water phase, and the percent for the surfactant is based on the amount of oil phase. Hence, the percentages for the three factors do not sum to 100.

The size of the batch was fixed by the 25-cm³ capacity of the syringes, but in the case of density, a choice had to be made. We wanted to keep a constant target density of 50 mg/cm³, but, since the initiator and surfactant are not removed from the foams, the variation in these factors would have to be compensated for, particularly in the case of the surfactant, which constitutes as much as 35 wt% of the oil phase. This meant, however, that the amount of monomers had to vary in order to keep the total weight of the oil phase constant. In the case of the initiator, which amounts to a maximum of 1.5 wt% of the water phase, the decision was made to accept the relatively small variation in the density caused by not compensating for differences in the amount of initiator because this compensation would have led to a variation in the ratio of water to oil phases. Since the surface properties of the interface are presumed to be very important in the behavior of this system, we judged this to be the greater risk.

We measured the emulsion droplet size by taking optical micrographs and measured the foam cell and pore sizes from scanning electron micrographs. We measured foam density and qualitatively assessed density uniformity through x-ray radiography. We attempted to measure BET surface areas, but the samples submitted turned out to be too small to give meaningful results, and there was not enough foam remaining to repeat the tests. We had also planned to measure the hexane wetting rate, as an indication of how hydrogen wetting would proceed, but discovered that the samples made with 100% styrene shrank substantially when put into the hexane; we have not found another solvent to replace it. Finally, we measured the compressive moduli of the foams. We analyzed the responses using the program X-STAT,¹⁰ which runs on an IBM PC computer. The model was the standard quadratic polynomial, and the coefficients were determined by a linear least-squares regression.

The most important information obtained from this experiment was that the structures of the three center-point foams, as determined by SEM, were widely different. One foam had the expected 2- to 3- μ m cells and was reasonably uniform; one had cells ranging up to 20 μ m; and the third had regions of apparent coalescence up to 1000 μ m across and was very nonuniform.

This was the first time that the reproducibility of the foam process had been checked in any systematic way, and the results indicated that it was very far from ideal. Our continuing efforts to investigate this problem are described in the following subsection on foam production. Unfortunately, lack of reproducibility meant that the statistics of the regression model were not particularly good, but we have drawn some qualitative conclusions that will need verification in future experiments.

Figure 4-8 shows that there appears to be a good correlation between the emulsion and foam cell sizes and the amount of surfactant used, as expected from the surface chemistry of the system and as indicated by earlier experiments. The

Fig. 4-8. Box-Behnken plot showing the effect of formulation on emulsion and foam cell sizes for PS foams.

Numbers in circles = emulsion cell size (μm)
 Colored numbers = largest foam cell size (μm)
 *Range of 2-5 μm

other two factors do not seem to have a significant effect. There also appears to be a qualitative correlation between cell size and density uniformity. The statistics for the compressive modulus data were not really good enough to draw firm conclusions.

Foam Production

Our attempts to use the new syringe pump (PMPY2) were initially very frustrating. We were attempting to reproduce the formula that had been the center point of the RSM, and we consistently got foams with regions of coalescence or phase separation—as shown by the scanning electron micrographs—although the cell size was only 2 to 3 μm in the regions where this did not occur. When we raised the density to 85 mg/cm^3 , this problem immediately disappeared. At about the same time, we remeasured the density of the original 50- mg/cm^3 sample that had been the basis for the center point selection in the RSM. We discovered that the density of this foam had been incorrectly measured and is in fact not 50 but 82 mg/cm^3 . Unfortunately, this result was incorrectly reported in the *Laser Program Annual Report 86* (Ref. 11) and

was the basis for the conclusion that we could produce good 50- mg/cm^3 foams. [A scanning electron micrograph of this foam appears in Fig. 5-1(b) of that report.] Apparently, the amount of water needed for a 50- mg/cm^3 foam is too large to be stably incorporated into the emulsion, at least with the current formula. These results were confirmed by making several 50- mg/cm^3 foams using the old syringe pump (PMPY1). The same regions of coalescence were observed.

Another problem in producing consistently good-quality foams appeared when we looked at the x-ray radiographs of foams made with PMPY2, which showed concentric rings of high density. In this case, the problem was traced to differences between the two syringe pumps. We used the load cell to measure the pressure on the syringes in both pumps, and found it to be substantially higher in PMPY2. This is because improvements had been made in the design that reduced the drag of the carriage. We lowered the air pressure until the pressure on the syringes was the same in PMPY2 as it is in PMPY1, and the radiographs showed that the problem had almost disappeared.

Our next steps will be to attempt to optimize the operation of PMPY2 with respect to the pressure and the number of strokes, and to try to gain a better understanding of the mixing process. We will also be running experiments to lower the density and still maintain uniformity, either by increasing the amount of surfactant, which should stabilize the incorporation of more water into the emulsion, or by removing the surfactant after the foams are made.

Conclusion

During 1987 we substantially increased our understanding of the PS system, particularly in the area of reproducibility and in identifying key variables that control the system. We are now able to consistently produce good-quality 85-mg/cm³ foams with cell sizes of 2 to 3 μ m. We have promising leads for improving quality and lowering the density. We are able to supply foam materials suitable for many of the tests that need to be carried out for target properties, as described in the final article of this section.

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Resorcinol-Formaldehyde Foams

Introduction

The development of low-density foam materials from a resorcinol-formaldehyde system was initiated in 1986 by Pekala at LLNL.¹² Results obtained by his group are very similar to ours. Development work is described in the *Laser Program Annual Report 86* (Ref. 13). The foams are promising as target materials because of their very small cell size, estimated at about 0.1 μ m, which offers potentially greater DT-fill stability than PS foams.

The synthesis of RF foams involves the condensation reaction of the monomers resorcinol and formaldehyde to form a sol-gel in a slightly basic aqueous solution at elevated temperatures, followed by supercritical extraction from liquid carbon dioxide to form a foam.

In the early stages of development, we could not control foam shrinkage, and foam densities were high and unpredictable. We have resolved these problems by improving the formulation and processing. We have also investigated carbonization of RF foams as a means of improving mechanical strength. We can now make foams with densities of 20 to 50 mg/cm³ that are promising in their quality and reproducibility as they apply to target design specifications.

We have employed additional techniques to characterize the foams in detail. To gain a basic understanding of the reaction kinetics of the gels, we have performed dynamic mechanical analysis studies. We are investigating transmission electron microscopy as a technique to resolve the very fine microstructures of the foams. We have also reduced the production time by a factor of 2. Molding of RF foams is discussed in the article "Target-Related Foam Properties" at the end of this section.

Foam Preparation

RF foam properties depend upon the degree of cross-linking and the morphology of the network structure. To identify key variables that may affect these two features, we performed a screening experiment,¹⁴ which showed that the factors that suppress foam shrinkage are low concentration of sodium carbonate initiator, high concentration of reactants, high molar ratio of formaldehyde to resorcinol (F:R), and long curing times. Results from another experiment show that a post-curing acid treatment also enhances cross-linking. We investigated the effects of formulation, curing temperature and duration, and different acid treatments on foam properties to further our understanding of the significance of each factor.

For the formulation studies, the first step was to conduct a three-variable Box-Behnken response surface methodology similar to the one performed for PS foams. We varied the reactant concentration from 4 to 8%, F:R from 1.6:1 to 2:1, and the concentration of Na_2CO_3 from 0.025 to 0.08 wt%. We prepared the foams in random order.

First we mixed and stirred the reactants at 70°C for 2 h. Then we cured them at 70°C for 7 d inside sealed ampules before washing them in 5% acetic acid at 50°C for 1 d. We then put the gels through a series of solvent exchange steps: they were placed in methanol, amyl acetate, and then liquid carbon dioxide for 7 d each. The CO_2 was taken above its critical temperature and was bled off slowly to avoid passing a liquid meniscus through the material.

The densities of these foams are shown in Fig. 4-9. It is apparent that Na_2CO_3 plays a dominant role in controlling foam shrinkage. For a fixed amount of reactants, all foams having the lowest densities were obtained from formulations with the lowest concentration of Na_2CO_3 . The effect of F:R appears less significant. The lowest density obtained from this study was 57 mg/cm^3 , but this foam fractured easily when being cut or touched.

In the curing study, we fixed the formulation at 6 wt% reactants, F:R at 2:1, and Na_2CO_3 at 0.05 wt%. We cured the mixtures at 70, 90, and 110°C for 7 to 14 d. We found that foam density is affected only by the curing temperature. The densities were in the range of 90 mg/cm^3 after curing at 70°C, and 80 mg/cm^3 after curing at 90 and 110°C. The compressive modulus of the foam, on the other hand, is affected by both curing temperature and time. We measured the moduli in the elastic region and found values between 1 and 2 MPa for foams cured at 70 and 90°C. Foams cured at 110°C had many cracks, and the moduli decrease as the curing time increases. All moduli were measured by compressing 1 × 1 × 1-cm cubes to a maximum of 10% strain at 0.1 mm/min. Since we were more concerned about lowering the density—as long as the foam had enough

Numbers in circles = density (mg/cm^3)

structural integrity to sustain machining and tritium radiation at cryogenic temperatures—we selected 90°C and 9 d as the optimum curing conditions. To verify these results, we cured the formulation with the lowest density from the formulation study (57 mg/cm^3) at 90°C for 9 d. The resulting density was lowered to 48 mg/cm^3 , and the foam was not as fragile as before: it could be vibratomed into blocks with smooth surfaces.

The purpose of an acid treatment in RF foam processing is to assist in further condensation of hydroxymethyl groups remaining in the gels to form ether bridges between resorcinol molecules. We tried to enhance this effect by using stronger acids. One advantage of using a stronger acid is that a lower concentration achieves the desired pH, and this can prevent side reactions. We investigated 0.125-wt% trifluoroacetic acid and acetic anhydride. Their pH values at room temperature were 1.9 and 1.05, respectively, as against 2.19 for 5-wt% acetic acid. After they sat in the acids for 3 d at 50°C, we compressed the gels to a 5% maximum strain at 0.1 mm/min in an attempt to determine the extent of cross-linking by comparing their compressive moduli with those of the gels that had not been treated with acid. We found that the modulus increased by 50% after treatment with acetic anhydride, 75% after treatment with acetic acid, and 170%

Fig. 4-9. Box-Behnken plot showing the effect of formulation on foam density in RF foams.

after treatment with trifluoroacetic acid. However, we did not observe any significant improvement in foam modulus or foam density. Therefore, we chose to keep 5% acetic acid at 50°C for 1 d as our standard acid treatment since it is much less hazardous.

Key findings from these studies have enabled us to produce foams of very low density. By lowering the Na_2CO_3 concentration to 0.0125 wt% and using the optimum curing conditions and an acetic acid treatment, we can reduce foam shrinkage to less than 25% and can consistently produce foams with densities in the range of 20 to 50 mg/cm^3 . All of these foams appear uniform in x-ray radiographs. While their low densities are attractive in that they would allow a lower target ignition temperature, these foams sometimes fracture during measurements of their thermal contraction. Details are given in "Target-Related Foam Properties" at the end of this section.

Thinking that the foams might contain internal stress because of cross-linking and/or vibratoming, we tried to eliminate this problem by annealing the foams, but without success. But an alternative technique that greatly improves the mechanical strength of RF foams is carbonization.

Carbonization of RF Foams

Carbonization is a process in which polymers are heated in an inert atmosphere to temperatures above 300°C. Reactions take place initially within the C-C chain: Depending on the chemical structure, the chains can either degrade into small molecules, collapse to form aromatic lamellae, or remain intact and coalesce with neighbors.

We have been using the carbonization facility in the Chemistry and Materials Science Department to carbonize RF. The procedure involves heating the foams in argon from room temperature to 1050°C in 12 h, maintaining 1050°C for 4 h, and then cooling down to room temperature in 1 d. RF generally retains its shape, but its shrinkage is greater than its weight loss, which results in an increase in density. This leads us to believe that the mechanism of RF carbonization is similar to that of many other organic materials: at temperatures below 300°C, RF turns black and loses excess monomers and solvents. Between 300 and 500°C, it undergoes a rapid weight loss in which oxygen and nitrogen are removed, and beyond 500°C, hydrogen is gradually eliminated. The C-C chains then coalesce to form a large network of aromatic rings. Elemental analysis reveals that an RF foam contains 62 wt% carbon and 38 wt% oxygen, nitrogen, and hydrogen, whereas its carbonized counterpart (CRF) contains 93 wt% carbon and 7 wt% oxygen, nitrogen, and hydrogen. We do not yet know what the source of nitrogen is in the foams.

The most striking result of carbonization is a tremendous increase in compressive modulus. An early study showed that an RF foam with a density of 48 mg/cm^3 has a modulus of 0.1 MPa, whereas its CRF foam has a density of 77 mg/cm^3 and a modulus of 8.7 MPa. To bring the density of the CRF foams to less than 50 mg/cm^3 , we carbonized RF foams with densities less than 40 mg/cm^3 .

We discovered that the increase in density as a result of carbonization depends on the Na_2CO_3 concentration and on the foam density before carbonization. Generally, foam density increases by 50% after carbonization. If the Na_2CO_3 concentration is high, or the foam density before carbonization is in the range of 20 mg/cm^3 , the density increases by more than 100% after carbonization. At present, we can produce CRF foams with densities less than 40 mg/cm^3 . Like RF foams, CRF shows a uniform density distribution in x-ray radiographs (Fig. 4-10). Other target-related properties appear

Fig. 4-10. X-ray radiographs of cylindrical samples of (a) RF foam, (b) CRF foam. The dents are surface defects caused by mishandling.

promising and are discussed in the final article in this section.

Other Formulations

We have investigated other resorcinol-based materials to see if we can achieve a network with increased and more uniform cross-linking. To increase cross-linking, we substituted catechol (1,2 dihydroxybenzene) and hydroquinone (1,4 dihydroxybenzene) for resorcinol (1,3 dihydroxybenzene). Since each of these isomers has four potentially active sites compared to the three possessed by resorcinol, tetra-functional cross-linking may occur. However, an analysis of their electronic resonance structures shows that, unlike resorcinol, in which all three sites are potentially active, in these isomers only two sites have significant activity, and the other two are less active. The reaction in tetra-functional systems would, at best, be slow.

Although we made a gel with catechol, the foam obtained had a density of 440 mg/cm³; a longer reaction time might have yielded better results. In the case of hydroquinone, a gel was never formed; we obtained only a precipitate, regardless of pH.

To increase uniformity, we tried phloroglucinol (1,3,5 hydroxybenzene) because it has three equally active sites. Preliminary studies showed that low concentrations of Na₂CO₃, high F:P molar ratios, and low curing temperatures contributed to lower-density foams.

We also set up a two-stage reaction with phenol-resorcinol-formaldehyde. The object was to first make linear prepolymers of phenol with formaldehyde by using an ortho-directing catalyst and then react these prepolymers with resorcinol and formaldehyde to make a cross-linked network. Such a procedure has the possibility of controlling network structure. We produced a crumbly, pink foam with a density of 55 mg/cm³ using triethanolamine as the catalyst.

Of all the alternatives we have looked at, phloroglucinol is the most promising candidate. We may resume our research effort in this area, depending

on the outcome of RF and CRF foam development.

Dynamic Mechanical Analysis of RF Gels

To increase our basic understanding of the RF system, we used several methods of mechanical testing to follow the changes in the visco-elastic properties of RF gels as they undergo condensation polymerization. The goal of this study was to characterize the rheological properties of the gel to determine whether it would yield a good foam at the early stages of gelation. This would allow a reduction of 2 to 4 wk in the processing time.

We used a mechanical testing machine to measure the change in compressive modulus with polymerization: (cure) time. For this experiment, a formulation of 2:1 F:R, 6 wt% reactants, and 0.05 wt% Na₂CO₃ was used. A batch of this mixture was sealed into 40 ampules and put in an oven at 70°C. Data was taken over a 40-d period by removing three ampules from the oven at various intervals and compressively testing the gels.

Figure 4-11 shows the results. The modulus reached a plateau after 20 d, and then continued to rise slowly. Extrapolating to infinite time, the maximum compressive modulus was calculated to be 0.0594 (±2%) MPa. To reach, for example, 70, 80, and 90% completion, we calculate that the gels would have to be cured for 9.4, 15.0, and 31.7 d, respectively. In our current processing, gels are

Fig. 4-11. Compressive modulus of RF foams vs cure time.

cured for 5–9 d, which should produce gels that have only 70% of their maximum modulus. Although the acid treatment improves foam strength, the mechanical testing results imply that a considerably longer cure time may produce gels with even higher moduli, and this needs to be confirmed.

Dynamic mechanical spectroscopy was the second technique we used to follow the polymerization of the RF system. In this experiment we used a rheometer equipped with a Couette (concentric cylinder) viscometer. The outer cylinder is sinusoidally oscillated at a constant frequency and amplitude while the magnitude and phase angle of the force transmitted to the inner cylinder is recorded as a function of the time as the polymerization reaction proceeds.

We determined the shear storage modulus, shear loss modulus, and viscosity as the gel polymerized. We used the same RF formulation as in the compressive testing experiment, but we changed the temperature in different runs. We prevented water evaporation from the cell by placing a layer of mineral oil on the surface of the RF solution.

Figure 4-12 shows the shear storage modulus G' as a function of time for four RF gels polymerizing at temperatures ranging from 59.2 to 81.7°C. Increased temperature accelerated the rate of polymerization, as evidenced by the earlier increase in modulus at higher temperatures. At long times the modulus of the gels approached a limiting value independent of cure temperature, indicating that the degree of cross-linking is the same for all cure temperatures.

Fig. 4-12. Shear storage modulus of RF gels vs time and temperature.

As discussed above, the strength of RF gels was shown to be a function of the formulation. To quantitatively compare gels prepared with different initiator concentrations or at different temperatures, we needed to define a point in each run that is specific to that run and that is easily recognizable.

We have defined the gel point as the time at which G' vs time has the maximum slope. Using the gel-point data, we compared the reaction rates of gels that have initiator concentrations of 0.025, 0.05, and 0.1 wt%. We found that reducing the concentration by a factor of 2 slowed the rate by 57%, while doubling the concentration doubled the reaction rate.

Using compressive and dynamic mechanical testing, we followed the polymerization of RF gels. The rate of reaction was found to be directly proportional to the initiator concentration. We found that increased temperature accelerates the reaction as expected but does not produce a stronger gel. We plan to use this dynamic mechanical technique to study the effects of other formulation variables on gel properties and reaction rate.

TEM Analysis of Foam Structure

Transmission electron microscopy (TEM) has been used on RF and CRF foams in an effort to identify their basic structures. By using a scanning electron microscope equipped with a field-emission gun at low voltage, we were able to tell that RF has a fibrous structure with a cell size of less than 0.1 μm . However, we were never able to resolve the structure fully; preliminary TEM results revealed that both RF and CRF are actually composed of interconnected beads, as in the case of silica aerogel. The bead size is on the order of 100 Å, with a pore size roughly five to ten times larger. BET surface area analysis reveals that RF foams have surface areas of 555 m^2/g , and CRF foams have surface areas of 705 m^2/g .

Production Time Reduction

The standard procedures for making RF foams are long and tedious. It takes 31 d to obtain a foam with a 1.5-cm diameter (from a 10-ml ampule): 9 d for

curing, 1 d for acid treatment, and 21 d for solvent exchange and drying.

To make production more efficient, we have attempted to minimize the time in the solvent exchange steps for both sizes of foam. We have tried direct supercritical extraction of water, and have tried exchanging water with methanol, isopropyl alcohol, or acetone before exchanging with liquid CO_2 . We also tried direct extraction from isopropyl alcohol after the exchange with water. Only isopropyl alcohol and acetone followed by CO_2 were found to give good foams. To accelerate diffusion, we have increased the temperature from room temperature to 45°C in the first step, and have flushed the supercritical extractor more frequently during the exchange with liquid CO_2 . Preliminary studies show that a gel with a 1.5-cm diameter takes 3 d in the first solvent exchange and 3 d in liquid CO_2 . This means we have reduced the production time of the foam to 16 d from 31 d.

Conclusion

The development of RF foams progressed rapidly in 1987. We have investigated the effect of formulation and process variables on foam properties, and have reduced production time. We are now able to produce foams with densities as low as 20 mg/cm^3 in 16 d. We are utilizing carbonization to improve the mechanical strength of the foams; the lowest density CRF foam has a density of less than 40 mg/cm^3 . Both the RF and CRF foams appear to have uniform density distributions and cell sizes of less than $0.1 \mu\text{m}$. We are now examining their behavior with respect to target specifications, as described in the final article of this section. Based on the information we have collected, we think RF and CRF foams are very promising materials for direct-drive ICF targets.

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Cellulose Acetate Foams

Introduction

Cellulose acetate foams hold promise for direct-drive ICF targets because of their extremely small cell size, which appears to be in the range of 0.1 to $1 \mu\text{m}$. Current SEM techniques make it difficult to verify this cell size since it is below their resolution, but we plan TEM work similar to that done for RF foams.

The idea for developing CA foams as low-density materials originated with R. W. Pekala, who based the idea on the thin membrane technology of CA and on some work indicating that benzyl alcohol would be a very effective solvent for this process.^{15,16} We began our development of CA foams this year and are making good quality foams with densities of 40 to 50 mg/cm^3 and cell sizes less than $1 \mu\text{m}$.

The structure of the foam is determined by a phase separation technique similar to the one used to make polyethylene foams.¹⁷ The foams are machinable, appear to be moldable, and wick liquid hydrogen. We have conducted gelling studies to correlate the cooling rate and gel composition to the structure and strength of the dried foams. We have studied CA foams from gel concentrations of 2 to 15 wt%. The compressive moduli of the foams range from 0.3 to 28 MPa for 3 to 15 wt% gels, respectively. The densities of the foams are predictable as a function of gel concentration.

Gel Preparation

The foams are made by a phase separation technique based on the solubility behavior of CA in benzyl alcohol. At temperatures greater than 45 to 50°C , CA has a very high solubility, but below 45°C the solubility drops sharply. The cooling is done in a water bath at programmed rates of 20 to 35°C per hour. This allows the foams to be made in the following manner: The CA is dissolved in benzyl alcohol at 110°C and then cooled to 30 to 45°C , at which point gelation takes place.

Fig. 4-13. Scanning electron micrograph of CA foam.

Fig. 4-14. X-ray radiograph of CA foam.

A series of solvent exchanges from benzyl alcohol to carbon dioxide is required to allow supercritical drying from carbon dioxide. First, benzyl alcohol is exchanged for toluene, then the toluene is exchanged for liquid carbon dioxide, and finally the carbon dioxide is supercritically extracted. The entire process takes 5 to 7 d.

Foam Properties

The densities of the CA foam materials, which can be cast in very large bricks, range from 28 to 225 mg/cm³, corresponding to gel concentrations of 2 to 15 wt%, respectively. The densities are higher than we calculated because of some shrinkage resulting from the drying process. ∴ scanning electron micrograph of the foam structure is shown in Fig. 4-13. As with the RF foams, it is very difficult to extract reliable structural information because the resolution is poor. An accurate assessment

of foam structure will be made from a TEM study similar to the one done for RF foams. The x-ray radiograph in Fig. 4-14 shows that the material is very uniform.

The compressive moduli of the foams range from 0.3 MPa for 3 wt% gels to 28 MPa for 15 wt% gels. Brunauer-Emmett-Teller (BET) surface area analysis yielded values of 600 to 800 m²/g, which is similar to the values for RF and silica aerogels. We have measured the thermal contraction of the foams from 300 to 20 K and determined it to be a 0.6% contraction in linear dimension. Additional foam properties are described in "Target-Related Foam Properties" at the end of this section.

Alternative Chemistries

We looked at other cellulose esters to see if the mechanical properties of the foams would be improved. These have the basic cellulose structure,

but have different types of esterification of the hydroxyls on the ring. The materials used were cellulose triacetate (CTA), where R, R', and R'' are acetate; cellulose acetate butyrate (CAB), where R is acetate, R' is butyrate, and R'' is hydrogen; and cellulose propionate (CP), where R is propionate and R' and R'' are hydrogen. We prepared all of these using the same method, and they behaved similarly except for some differences in physical properties—mainly their solubility in benzyl alcohol. The CP dissolved in the benzyl alcohol, but did not separate out when cooled. The CTA and CAB dissolved, and phase separation occurred when they were cooled; these gels were processed and dried to foams. A 3.8-wt% gel of CTA yielded a foam with a density of 80 mg/cm³ and a compressive modulus of 0.56 MPa. The CTA foam was more rigid to the touch than CA foams because

of its larger percent of crystallinity. CTA also requires a higher temperature to dissolve than the other cellulose esters and is more difficult to get into solution. The processing of the CAB gel was the same as for CA and CTA through the solvent exchange steps, but the CAB gels collapsed almost immediately when they were pressurized in the supercritical extractor with liquid carbon dioxide, making CAB unsuitable for foams made by this process. We do not have an explanation for this result.

We are also investigating a solvent/nonsolvent system for producing a CA gel. In this process the CA is dissolved in a solvent and the solution is placed in a permeable membrane container in which the solvent is exchanged with a nonsolvent. The solvent and nonsolvent we used were acetone and methanol, respectively. A gel is formed by precipitation as the solvent is exchanged with the nonsolvent. The gel is then placed in methanol for a more thorough exchange of the acetone, and finally the methanol is exchanged with liquid carbon dioxide, which is supercritically extracted. We are still evaluating this approach.

Viscosity Measurements, Cooling Rate Effects, and Thermal Analysis

We undertook viscosity and cooling rate studies to determine the gelation points of the CA/benzyl alcohol solution and to find an optimum cooling rate to control foam morphology to improve mechanical strength. We made measurements of viscosity vs temperature as a function of time to determine the gel point of the CA solution. We measured the viscosities with a vibrating rod viscometer. We prepared the gels in 3- to 15-wt% concentrations, placed them in a jacketed reaction vessel, and cooled them from 90 to 20°C during 3-, 10-, and 48-h periods. We controlled cooling rates by a refrigerated circulator bath and temperature programmer. We placed additional samples in the bath to be cooled at the same time as the viscosity sample so that the gel point could be observed visually.

Figure 4-15 shows that all the viscosities change at approximately the same rate but have different values. The gelation point is seen as a sharp increase

Fig. 4-15. Viscosity vs temperature in CA gels for various concentrations.

Cellulose Acetate Foams

Fig. 4-16. Densities of CA foams vs gel concentration.

Fig. 4-17. Compressive modulus vs density for CA foams.

in viscosity at temperatures ranging from 33 to 45°C, depending upon gel concentration. The gel point increases with increasing concentration as expected, and the rise in viscosity matches the observed gelation points of the solutions.

In Fig. 4-16, the densities of dried foams made from the gel samples are shown as a function of gel concentration. The densities are very reproducible and are essentially independent

of the cooling rate. There is a linear correlation between density and gel concentration.

Figure 4-17 shows preliminary measurements of the compressive modulus. There is an increasing trend with increasing density as expected, but the cooling rate appears to have little effect. We submitted the 3-wt% foams for x-ray crystallography analysis, which indicated that all the samples were amorphous; there were no observable differences in the structures of the 3-wt% foams cooled at the different rates. We may have to use small-angle x-ray scattering to observe any crystalline junctions.

A comparison of differential scanning calorimetry results for the CA starting material and the dried foams showed no differences, which indicates that there is no chemical change in the CA during the foam-making process.

Conclusion

During our first year of making cellulose acetate foams we have produced a promising material for target foam. Next year we will concentrate on improving

our formulation and processing methods and will make additional tests of foam properties.

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Target-Related Foam Properties

Introduction

We are conducting investigations of foam properties that are directly related to the target specifications described in the introduction to this section. We are developing techniques for quantifying density uniformity, for measuring wetting properties with both liquid hydrogen and liquid DT, and for measuring thermal contraction from 300 to 15 K. Radiation stability is assessed as part of the DT wetting experiments. We are evaluating various machining techniques, and tests are under way to identify an agent suitable for bonding hemishells together. Because machining is not an efficient method of producing large numbers of targets, we are also investigating molding of foams.

Density Uniformity

The specification for target density uniformity is somewhat ill-defined, but it is generally given as uniformity of the projected or radial density to within 1% variation between areas $30\text{ }\mu\text{m}$ in diameter. As the earlier articles in this section report, we have used x-ray radiography to make a qualitative assessment of density uniformity and to locate defects such as voids and high-density particles. However, to verify that the foams meet target specifications, we need a quantitative technique.

Since the foams are opalescent and transmit light—although with considerable scattering—the question arose as to

whether this light could be used as a quantitative method for determining foam density uniformity. We conducted two types of experiments: contact printing from a point source of illumination, and scanning densitometry with a photometer.

In the first experiment, we placed a piece of foam on top of a photographic plate, which we exposed by switching on a point source of light. We then developed the plate to reveal the light transmitted through the foam. Comparison of the transmitted-light photograph and an x-ray radiograph shows similar patterns within the nonuniform foam. To quantify the light transmission, we tried digitizing the photograph using a scanning microdensitometer, but this approach took much time and seemed to lack intensity resolution.

We took a more direct approach in the second experiment by using a stepper-motor-driven stage on a microscope equipped with a photometer. We scanned a piece of foam with a $25\text{-}\mu\text{m}$ void, and, from measurements of the change in intensity, we calculated the void size to be $25\text{ }\mu\text{m}$. After this first experiment, which was operated manually, we interfaced the equipment to a computer that controlled the stage motion and collected the photometric intensities. We analyzed the data and plotted them using the VIEW program on the LEAF computer system. Figure 4-18 shows an automated scan through a step wedge of foam. The step heights are 0.6, 0.8, 1.0, 1.2, and 1.5 mm. The intensity was sampled at $120\text{ }\mu\text{m}$ intervals. On the second step from the top, two small density increases are apparent as depressions in the intensity.

We are also developing a method of scanning an x-ray beam over the surface of the foam and measuring the transmitted intensity. This is called point-by-point radiography. We have assembled the equipment and written the computer programs needed to analyze the data. We plan to begin testing on foam samples shortly.

In addition to our work on experimental measuring techniques, we have

Fig. 4-18. Photometric scan of PS foam step wedge.

developed a well-defined computer model that closely resembles the features of PS foams. The objective here is to study the relationships among cell size, cell-size distribution, foam density, and radial density uniformity.

The model is constructed as follows: A box with dimensions of $40 \times 40 \times 20$ is set up and filled with spheres that represent the water droplets of the emulsion. In order to fill the box, we start by placing spheres of unit radius randomly in the box in such a way that they do not intersect; the dimensions of the box are defined in terms of the radius of these largest spheres. When all spheres of this size that will fit within the box have been inserted, the box is scanned and the interstitial locations are filled with the largest spheres that can be accommodated without having two spheres intersect. This placing of spheres is continued until the radius of the largest sphere that can be placed is smaller than some predetermined size, in this case 0.1. In order to avoid concern with the ordering effect of flat surfaces, the box is constructed with periodic boundary conditions. This means that the bottom of the box is actually in the same plane as the top of the box, so that a sphere can be placed at a distance less than its radius from the bottom, and the part of the sphere that leaves the bottom of the box reenters at

the top. The same conditions are applied to the sides. As long as the box dimensions are large compared with the sphere radii, this represents an efficient method for accurately modeling the interior of a piece of foam.

Since the spheres are meant to represent the cells in the foam, they are said to be empty, and the PS fills the interstitial spaces between the spheres. The density ρ of the system is then

$$\rho = 1 - \frac{\sum_i \frac{4}{3} \pi r_i^3}{V_{\text{box}}}, \quad (2)$$

where the sum is over the spheres of radius r_i , and V_{box} is the volume of the box. Normally, the sum is taken from the largest to the smallest spheres; in this way, we can look at systems of different densities by truncating the sum when the desired density has been reached. (Here and throughout this discussion we are assuming a polymer density of unity. Actual bulk densities can be inserted without difficulty.)

As expected, as the spheres get smaller, more can be fitted into the structure. In Fig. 4-19, we plot both smallest sphere radius and system density as a function of the number of spheres added.

As noted above, our primary interest is in the density uniformity projected through the sample. For our model the projected density is easily calculated as follows: In setting up the model, the stored information includes the coordinates a , b , and c of each sphere along with its radius r . Thus the equation of sphere i is

$$(x - a_i)^2 + (y - b_i)^2 + (z - c_i)^2 = r_i^2. \quad (3)$$

If we want to project the mass (the space between spheres) onto the (x,y) plane, and if in the box the z direction is represented by the box height of 20, it is easy to see that the projected mass m at the point (x,y) is given by

$$m(x, y) = 20$$

$$- 2 \sum_i [r_i^2 - (x - a_i)^2 - (y - b_i)^2]^{1/2}. \quad (4)$$

In Eq. (4) only those spheres that intersect the line parallel to the z -axis that passes through the point (x,y) are included in the sum. The projected density is simply $m(x,y)/20$. Thus, a grid of projected densities of arbitrary mesh size can be set up. In the work reported here, we used a mesh of 256×256 . One can easily take the projected densities at these points and calculate the average density in the square areas defined by the points by taking a simple average. In a similar fashion, average projected densities over larger square areas can also be computed, allowing an examination of fluctuations in density from area to area as a function of area size. In addition, we can look at the projected density as a function of slab thickness by simply projecting only some predetermined fraction of the distance through the sample.

To summarize, we have developed a model that allows us to look at projected density homogeneity as a function of sample density, sample thickness, and the size of the projected area. The results from this model follow.

Fig. 4-19. Model of PS foam density. Sphere radius and system density are plotted vs the number of spheres added.

Let us first examine the effect of overall density on homogeneity. In Fig. 4-20(a) we have plotted the frequency of deviation from the mean density against the percent difference from the mean density for projected areas of four grid squares (of size $40/256$ on a side in units of largest sphere radii) for systems with overall bulk densities of 0.2, 0.4, and 0.6. Note that as the density drops, the distribution becomes somewhat more narrow. The actual differences are more striking if we remember that the abscissa is in terms of percent difference from the mean. Thus 5% for the sample with a density of 0.6 represents an actual density deviation of 0.03, while the same percent deviation for the system with a density of 0.2 is only a deviation of 0.01. The more narrow distribution at lower density is, of course, a result of the broader distribution of spheres needed to lower the density to 0.2, which results in a more homogeneous distribution of the remaining mass.

The effect of sample thickness is shown in Fig. 4-20(b), where data for $\rho = 0.4$ is shown for both the full thickness of 20 and for the two halves of 10 each. The plot shows the expected result, namely a narrower distribution for the thicker sample resulting from the collective effects of randomly placing mass throughout the sample. The curves are roughly Gaussian, a fact easily checked by noting that the ratio of their peak heights is roughly $\sqrt{2}$. This fact allows us to estimate the distribution for samples of arbitrary thickness. The Gaussian nature of the model also

Fig. 4-20. PS foam density homogeneity effect of: (a) overall density; (b) sample thickness; (c) grid square size.

manifests itself in Fig. 4-20(c), where the frequency of deviation is plotted for projected areas of 4, 16, and 64 grid squares.

We ultimately expect to acquire the foam homogeneity specifications in terms of the Fourier transform of the projected density as obtained from a radiograph. We will use our model to explore the nature of the transform for a system with a well-defined and adjustable structure. For example, we can easily create a system with a large defect and see how the Fourier transform changes with defect size. Work along these lines is in progress.

Foam Wetting Properties

Polymer foams used in laser fusion targets must be wetted with liquid DT. To help understand the dynamics of wetting and to characterize the foam, we use liquid hydrogen, which does not pose a radiation hazard. Hydrogen wetting measurements were made at the National Bureau of Standards using a cryogenic apparatus equipped with a balance. The foam samples were equilibrated at 20 K before starting the wetting experiment. To start the wetting, the foam was lowered until it just touched the liquid hydrogen. Foam weight was then recorded as a function of time.

Figure 4-21 shows the experimental liquid hydrogen wetting behavior of the foams we are investigating. Silica aerogel and polyethylene (PE) were also included for comparison.

The rate of wicking in increasing order is aerogel < PE < CRF < RF < CA < PS, which corresponds approximately with increasing pore size but may also depend on contact angle and foam morphology. For laser fusion targets a smaller pore size is desirable because it should provide a higher capillary pressure. This makes the target capable of sustaining a higher internal vapor pressure and allows it to tolerate variations in temperature without suffering a liquid blowout. Thus, the materials having the lowest wicking heights are the most desirable from this standpoint.

We have already carried out a preliminary capillary pressure test on a PS foam sample. The value obtained was 0.75 psid with hydrogen, which should correspond to ~ 1.5 psid with DT. Since this result

Fig. 4-21. Liquid hydrogen wetting behavior of low-density foams.

is acceptable according to our calculations, we believe PS foam will work for our target design, although CA, RF, and CRF offer potentially improved target stability.

We are just beginning experiments to measure the rate of DT wetting in PS foams. The first step was to develop an annealing procedure that reduces the amount of residual water in the foam to a safe level, i.e., one that precludes formation of any hazardous amounts of T_2O . This has been accomplished: Heating the foams to 115°C for 24 h is sufficient. Another objective of these tests is to assess the radiation stability of the foam to the tritium. In one preliminary test, we exposed a piece of PS foam to DT for 72 h with no visually observable changes, but no quantitative data are yet available. Other foam materials will be tested after the PS work is completed.

In addition to our experimental measurements, we have continued our efforts to provide a mathematical model for wetting behavior. In the *Laser Program Annual Report 86* (Ref. 18), we made use of the rate of solvent wetting to estimate

pore size in foam materials. Briefly, the rationale involves identifying the driving pressure P at any time as the difference between the pressure due to capillary rise and gravity:

$$P(t) = \frac{2\gamma \cos \theta}{r} - \rho h(t)g, \quad (5)$$

where γ is the surface tension, θ the contact angle, r the radius of the capillary, ρ the density of the fluid, $h(t)$ the fluid height as a function of time, and g the acceleration due to gravity. The rate of laminar fluid flow in a pipe is

$$\frac{dh}{dt} = \frac{r^2 P}{4\mu h}, \quad (6)$$

where μ is the fluid viscosity. We have a factor of 4 in the denominator rather than 8 as previously reported because in our application the rate of advance of the fluid height is twice the rate of advance of the center of mass of the fluid, which is actually what the equation should

address. Using Eq. (5) for P in Eq. (6) we get

$$\frac{dh}{dt} = \frac{\gamma r \cos \theta}{2\mu h} - \frac{r^2 \rho g}{4\mu}. \quad (7)$$

If one is concerned only with the initial rate of rise, the second term can be dropped and the resulting expression integrated to give

$$h(t) = \sqrt{\frac{\gamma r \cos \theta}{\mu}} t. \quad (8)$$

Thus a plot of $h(t)$ vs $\sqrt{t}$ allows the determination of r , if γ , μ , and θ are known.

This development is strictly valid only for laminar fluid rise in cylindrical capillaries, for which r is constant and well-defined. In our PS foam materials the structure more closely resembles 1- to 5- μm polyhedral cells randomly connected by small ($<2\text{-}\mu\text{m}$) pores. In

addition, issues of permeability and two-phase (fluid and air) flow in the experimental situation, long recognized as important by oil engineers, are ignored in the simple capillary model. Because of these problems, the approach would probably not have been pursued, except that the wetting data in fact show an excellent and reproducible $\sqrt{t}$ behavior. However, even if we accept the equations as they stand, we must still address the question of proper interpretation of capillary radius r from Eq. (8). In order to probe this issue, we solved a similar set of equations for a model, shown in Fig. 4-22, that includes both cells and pores. The cells are diamond-shaped with truncated height and diagonal length d , and they are connected at their corners with pores of diameter p .

As in the capillary problem, we will assume Eqs. (5) and (6) and start with the truncated Eq. (7),

$$\frac{dh}{dt} = \frac{\gamma r \cos \theta}{2c\mu h}, \quad (9)$$

where we have replaced the h in the denominator of Eq. (7) with the actual side length subject to drag, ch , where c is a geometric factor that approaches $\sqrt{2}$ as p/d approaches zero. Also, r is now a function of the fluid level in the cell and this depends upon h , as well as upon d and p . We note again before proceeding that Eq. (6) is strictly valid only for flow in cylindrical pipes, a condition violated by the present model.

Integration of Eq. (9) leads to the result

$$h = \sqrt{\frac{\gamma(d-p)\cos \theta}{2c\mu \ln(d/p)}} t \quad (10)$$

for $h \gg d$. It is interesting to note that the $\sqrt{t}$ dependence is maintained with this modified model. We see that r of Eq. (8) can be identified as

$$r_{\text{cap}} = \frac{d-p}{2c \ln(d/p)}. \quad (11)$$

Our conclusion, then, is that the capillary radius r obtained from plots of h vs $\sqrt{t}$ is at best a complicated function of both cell diameter and pore size even for

Fig. 4-22. Model of PS foam structure for wetting data analysis.

Fig. 4-23. Apparatus for measuring thermal behavior: (a) overview; (b) close-up of sample holder, cryostat, and displacement measuring device.

this simple model, and does not specify either of these quantities. Thus, although our rate-of-wetting data may be useful in certain characterizations of our foams, it cannot give us detailed information about the dimensions of local structure as was previously reported.

Measurement of Thermal Behavior

It is important to characterize the thermal behavior of foam materials at cryogenic temperatures because the foams will be prepared at room temperature and then cooled to approximately 20 K for filling with liquid DT. Our goal was to characterize the dimensional change of foams over a range of 300 to 15 K.

We started the characterization with an existing thermal mechanical analyzer that was designed to operate from 90 to 400 K. The sample is held in a quartz fixture that transmits changes in sample length to a linear variable differential transformer (LVDT). An adjustable force is applied to the sample through an electromagnet. Our modifications to the system included extending the low temperature range to 15 K, which we did by building a new cryostat with simultaneous feedback control of both the cryogen flow and the cryostat heater. We also improved the sensitivity of the length-sensing transducer by substituting an eddy-current transducer for the LVDT. The new transducer has a sensitivity of $1\ \mu\text{m}$.

Photographs of the apparatus are shown in Fig. 4-23. Length changes are

recorded on a chart recorder as well as on a computer, which also controls the cryogen flow. The apparatus can be operated in two different modes: as static length measurement vs temperature, or as dynamic mechanical spectroscopy. In the static length measurement mode, a constant load is applied to the sample and the length is recorded as a function of temperature. In the dynamic mode, a varying force, either square wave or sinusoidal, is applied to the sample in the frequency range of 0.05 to $20\ \text{s}^{-1}$, and the change in length is recorded as a function of temperature. In this mode, changes in the elastic modulus of the foam can be determined throughout the temperature range of the apparatus.

Our first experiments involved measurements of thermal contraction in the temperature range of 300 to 15 K. We prepared the foams by vibratoming them into rectangular prisms with dimensions of $5 \times 5 \times 7.5\ \text{mm}$. The foams tested were PS, RF, CRF, CA, and silica aerogel.

The fractional dimensional change as a function of temperature is shown in Fig. 4-24. The contraction of PS foam to 15 K was measured to be 1.4%. We observed exactly the same behavior on samples of full-density PS, which were used to test the operation of the apparatus. Literature values of the thermal contraction of PS are listed as 1.5%. With values of 0.6 and 0.55%, respectively, CA and RF foams have very similar thermal contraction behavior. Silica aerogel and CRF expand on cooling by 0.1 and 0.15%, respectively.

Fig. 4-24. Results of thermal measurements for low-density foams.

Fig. 4-25. Development PS hemishell for proposed direct-drive laser ICF target.

The cooling-related expansion of materials having a plate morphology, such as polyethylene, has been observed before.¹⁹ The overall volume of the material decreases with temperature, but the dimensions of the material do not change uniformly: the material contracts strongly in the thin dimension of the plate. This strong contraction causes the length to actually increase. For the purposes of target construction it would be desirable to have no thermal contraction; however, having accurate measurements of the contraction allows us to predict the final dimensions.

From these results, we can conclude that the RF and CRF foams are potentially superior to PS in this respect. However, as mentioned previously, RF foams sometimes fracture during these measurements, which causes problems. The CRF foams do not fracture. Targets can probably be made from PS foams, but the larger thermal contraction may present significant challenges for the target design.

Foam Machining

An assessment of machinability for PS, RF, CRF, and CA foams is under way. We have produced several PS hemishells—such as the one shown in Fig. 4-25—by simply turning them on a lathe. We are investigating both diamond-point turning and grinding as machining methods, and we have prepared samples of all three foam materials. Methods of quantifying the surface finish using the scanning electron microscope (SEM) are being developed and preliminary results indicate acceptable values. PS foams are by far the easiest to machine, but so far there is nothing conclusive to indicate that the other foams cannot also be suitably machined.

Foam Bonding

We are conducting experiments to identify a suitable bonding agent for PS and RF foams. Several adhesives bond PS foams; these include the GR-650 resin used for silica aerogel, urethane and resorcinol glues, and a plastic resin. It is desirable to use a bonding agent that is mostly C, H, and O or N, rather than the GR-650, which contains silicon and creates a bond of substantially higher density than the hydrocarbon foams. These bonds are being assessed by x-ray radiography and with the SEM.

We have found that only the GR-650 bonds RF foams; none of the other agents mentioned above does. The experimentation is still in its very early stages, however, and we plan to expand the testing to our other foam materials and to test other types of adhesives.

Foam Molding

The required shot repetition rate for a commercial laser ICF facility is estimated to be on the order of 10 s^{-1} . If machining is selected as the method of choice for making the target hemishells, the number of spherical surfaces needed to be machined per day will be well over three million, which appears to be an impractical proposition. Molding offers an acceptable alternative: although molding will not eliminate the need for machining, it limits it to the flat surfaces that will be

bonded. We have therefore undertaken to develop a molding process for PS foams.

Molding of PS foams for ICF targets imposes two apparently conflicting requirements:

1. The material to be molded should have a low viscosity so that it can be poured into the die without trapping air bubbles.

2. The emulsion to be molded should be uniform and have a small cell size.

The way to satisfy the second requirement is to mix the emulsion well, which, unfortunately, increases its viscosity. An immediate consequence of this situation is that simple molding techniques, proven to be useful for many systems, would not work in this case. A low-viscosity emulsion, which fills the die readily without any air bubbles on the

surface, yields a large cell-size, nonuniform foam. A well-formed emulsion, having relatively high viscosity, almost always traps small air bubbles near the surface if one tries to fill a die with it. Injection molding using flow-through dies is also problematic because the high viscosity of the emulsion requires pressure drops well beyond those capable of breaking—at least partially—the well-formed emulsion. A more far-reaching consequence of this dilemma is that any attempts to separate the issues of foam processing and hemishell fabrication are not likely to be very rewarding.

This situation led us to develop an integrated processing/molding scheme, in which the emulsion is created and molded simultaneously. In this technique, the syringes are filled with the

Fig. 4-26. Schematics of molding apparatus for PS foams: (a) mixer/mold assembly; (b) die assembly.

desired amounts of aqueous phase and organic phase; the die assembly and a specially designed mixing element (described below) are placed between the syringes, and mixing is initiated as described in the earlier articles in this section. Figure 4-26(a) is a schematic of the mixer/mold assembly. The components of the die assembly are shown in Fig. 4-26(b).

The original syringe mixer, the PMPY1 described in "Polystyrene Foams" earlier in this section, had to be modified for the molding application. Whenever we assembled the various components of the system air would get trapped alongside the process liquids, leading to bulk and surface imperfections. To eliminate these air bubbles, we modified our operating procedure so that the entire mold-mixer-syringe assembly was fitted together under water and tested for leaks. We also modified the syringe supports so that the completely assembled system could be placed and secured inside them.

To reduce the pressure drop across the mixing orifice and thereby shorten the mixing time while still maintaining

emulsion uniformity, we developed a new mixing element to replace the single orifice. This element (Fig. 4-27) consists of a perforated plate containing holes of a single diameter. We tried different combinations of hole number and size, and chose a plate with 12 holes of 1-mm diameter.

The die material also imposes some special requirements. The ideal material should be inert toward the emulsion, easily machinable to a smooth surface, and rigid enough to maintain the design dimensions during the molding operation. Metals tend to interact with the emulsion; in the initial experiments with metal dies, the foam surfaces were invariably pitted, pointing to localized emulsion breakup near the die surfaces. Gold-plating the die surfaces in contact with the emulsion proved only marginally useful. Teflon is an ideal material in its inertness, but it is too flexible to maintain dimensional stability. One way around this problem is to use Teflon impregnated with ceramic or graphite, which we have done successfully.

Figures 4-28(a) and (b) show the hemispherical parts made in the ceramic-impregnated Teflon die, using the new mixer element and the modified syringe pump. The flat surface of the hemisphere has been vibratomed. Figure 4-28(c), the SEM picture of the same hemisphere, shows foam uniformity and cell size. Figure 4-28 shows that the surface of the part, although mostly uniform, has identifiable nonuniformities. Also, the surface is still not within the target specification of $5\text{ }\mu\text{m}$. The surface finish of the die, which is similar to that of the foam, appears to be the limiting factor here, but

Fig. 4-27. Mixing element for PS foam molding apparatus.

Fig. 4-28. Molded PS foam part: (a) photograph of inner surface; (b) photograph of outer surface; (c) scanning electron micrograph of foam structure. The hemisphere is 1 cm in diameter. ▼

localized breakdown of the emulsion may also cause larger surface defects.

Thus the major problem in molding PS foams at the present time appears to be surface finish. We will attempt to improve surface quality by investigating the causes of local emulsion breakdown, improving the die surface finish by techniques such as diamond machining, grinding, and lapping, and/or by changing the die material to glass.

We have also looked at the feasibility of molding RF foams into hemishells. During the preliminary study, we found that glass molds yield smoother surfaces than metal molds. We have also found that proper sealing is essential to avoid water evaporation during curing. (One way to ensure a good seal is to apply silicone sealant in all the openings of the mold.) We have successfully made hemishells of RF foam from glass tubes and a prototype multi-cavity mold. The surface finish of these hemishells appears reasonably smooth, but they are bigger than the real target size. If molding is chosen as the technique for making hemishells, we will then make a precisely machined glass mold.

Conclusion

At this point, we can compare our foam materials as potential targets. The big advantages of PS over the other types of foams are its superior strength and

machinability and its greater efficiency of production. The fact that it has a larger cell size than the other foams does not seem to be a significant problem in light of the capillary pressure test. However, its relatively large thermal contraction is a significant disadvantage.

RF foams seem to have problems with fracturing under various conditions, but this problem is apparently overcome with carbonization.

None of the foams has actually been quantitated in terms of density uniformity, but since this is almost certainly related to cell size, RF and CRF probably have an advantage here.

Although CA is not as far along in development as the other foams, no insurmountable hurdles have been encountered. Needless to say, we have many further tests to perform to confirm that target specifications will be met, but all the foams under development are still promising candidates.

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Laser Research and Development

5

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Laser Research and Development

Introduction

W. H. Lowdermilk

The principal objective of the Laser Research and Development Program is to identify and develop laser technology necessary to meet near-term and long-term Laser Program goals. Two laser systems, with the performance requirements shown in Table 5-1, will be needed to meet these goals.

The first system will be used to further develop the physics of laser-driven implosions, with the objective of producing the high-density, high-temperature plasmas necessary for fusion reactions. The goal of this research is to achieve an energy gain of up to 100 from a laser-driven fusion capsule. Experimental results obtained during 1987 from Nova and the Halite Program, and computational analysis of those results, lead us to conclude, with high confidence, that a laser meeting the performance requirements in the column labeled "high gain" can indeed drive a capsule to high gain. During 1987, the Inertial Fusion Division of the Department of Energy initiated a study of requirements for this high-gain facility, now called the Laboratory Microfusion Facility (LMF); the goal, as stated in the Inertial Confinement Fusion (ICF) Program 5-Year Plan, is to have LMF operational within 10 years.

The technical and cost requirements for an LMF driver are achievable with credible extensions of the state of the art of Nd:glass laser technology. In 1986, we completed a design study of a glass laser system that meets the LMF technical requirements using technology already in existence or judged to be achievable within 10 years. The estimated cost of that system was \$1 billion. This study also identified the technological developments with the greatest potential for reducing the cost of LMF:

1. High-efficiency solid state amplifiers.
2. High-damage-threshold optical materials and coatings.
3. Advanced nonlinear optical materials.
4. Advanced crystalline laser materials.

In 1987, we focused our resources on these key technologies. The results of those efforts are summarized in this section. On the basis of the

Table 5-1. Characteristics of laser drivers needed for ICF military and civilian applications.

Characteristic	Laboratory high gain (~100) fusion driver	Inertial fusion reactor driver (1 GW _e)
Pulse energy	10 MJ	2-6 MJ
Peak power	1000 TW	200-600 TW
Pulse duration	0.03-100 ns	~ 10 ns
Pulse shape	Diverse, flexible	Single, fixed
Pulse repetition rate	Single shot	Few hertz
Efficiency	Unimportant except for cost impact	≥10%
Wavelength	250-1000 nm	Fixed, <500 nm
Cost goal	\$25-50/joule	\$30-60/watt \$100-150/joule

results achieved, we expect that the LMF, with a Nd:glass laser driver, can be operational within 10 years at a cost of \$750 million.

The second laser system, whose requirements are also shown in Table 5-1, would be used to drive an ICF reactor to generate commercial electric power. The technical requirements for this system, shown in the column labeled "Inertial fusion reactor," are beyond the state of the art for solid state lasers or for any other driver. In previous years, we carefully examined the ability of solid-state lasers to meet these technical and cost requirements. Our conclusion was and still is that it is feasible to meet all ICF reactor requirements using a solid-state laser driver. The most challenging requirement is for a driver efficiency of at least 10%, while maintaining system cost of \$30 to \$60 per watt. The requirement that the driver produce an average power of ten or more megawatts can be more easily achieved using crystalline laser host materials; their advantage is a thermomechanical strength roughly 10 times higher than that of glass.

In our study we identified the key technological developments for ICF reactor driver applications. During 1987, we pursued research (with funding from the Department of Energy) on two of these technologies:

1. Identification of advanced crystalline laser materials.
2. Development of advanced methods for growth of crystals with the high optical homogeneity and large dimensions required of an ICF reactor driver.

We continued basic materials research in 1987 (with funding from the Office of Basic Energy Science) on three areas of fundamental, long-term importance for laser applications:

1. The magnitude and wavelength dependence of the nonlinear index of refraction of transparent laser materials.
2. Spectroscopic and laser properties of impurity ions in solid state laser host lattices.
3. Basic properties of damage-inducing interactions of laser light with optical surfaces.

We also continued theoretical atomic and molecular physics research for ICF applications.

Solid State Laser Components

Introduction

The ultimate goal of our efforts in laser components is to design, build, and test hardware for use in a next-generation fusion laser system. During 1987, however, our efforts were largely expended in developing a long-lived edge cladding for the new Nova laser disks. This successful project, reported in "Amplifier Development for Nova" in Sec. 3, has direct application to our multimegajoule (MMJ) laser goals.

To optimize the design of laser disks for a MMJ system, it is necessary to have a reliable quantitative model of amplified spontaneous emission (ASE). We have therefore developed computer codes, described in the next article, "Calculations of Amplified Spontaneous Emission Rates in Laser Disks," to calculate the disk-averaged ASE decay rate and the spatially resolved ASE decay rate for laser disks with arbitrary edge conditions and gain distributions. We have compared these predictions with experimental results obtained with elliptical disks of three sizes. The predicted ASE decay rates are lower than the measured rates by about 33%, for reasons that we do not yet understand.

We are also developing the capability to measure very small optical losses in laser components, as discussed in "Attenuation of New Nova Laser Glass at 1 μm ." Such losses will play an important role in determining the energy extraction efficiency in a MMJ laser system. Although we report measurements on the new Nova disks, our long-term goal is to obtain quantitative understanding of energy extraction in a MMJ system.

Author: H. T. Powell

Calculations of Amplified Spontaneous Emission Rates in Laser Disks

Spatially Averaged Decay Rates. During 1987 we extended the Monte Carlo ray-tracing analysis for ASE, originally developed by Trenholme,^{1,2} so we could treat laser disks with imperfect edge claddings (i.e., claddings that do not absorb all ASE radiation incident upon them). The updated model includes the effects of a nonuniform gain profile in the disk and of Fresnel reflectivity at the disk faces, and it treats polygonal disk geometries such as those used in the composite-clad disks for Nova and those anticipated for use in multimegajoule laser designs. The general aspects of the model are discussed here; its application in the design of the new Nova disks is described in "Calculations of Effects of Imperfect Claddings," in Sec. 3.

The spatially averaged ASE decay rate can of course be calculated by evaluating the sum of the spontaneous and stimulated decay rates at each point in the gain medium, as determined by the incident flux coming from all other points, and then averaging this rate over the laser volume. But as noted previously,¹ when the absorption of the gain medium at the laser wavelength is negligible (as is the case for the Nova laser disks), the average ASE decay rate for the disk can be computed directly without requiring a calculation of its spatial dependence. This is done by noting that the instantaneous power loss P_{out} corresponding to radiation that leaves the gain volume through all surfaces is related to the total

instantaneous spatially averaged decay rate k_{tot} due to radiation by

$$k_{\text{tot}} = \frac{P_{\text{out}}}{E_{\text{stored}}}, \quad (1)$$

where E_{stored} is the total energy stored in the disk. In this way the calculation of the average ASE decay rate is reduced to the simpler calculation of the total power output from a given spatial distribution of gain, edge conditions, etc.

In the limit in which the gain in the medium is negligible, k_{tot} is simply the spontaneous radiative decay rate k_{spont} for the transition of interest. When the gain (and hence the ASE) is significant, it is convenient to define a decay rate multiplier $\bar{M}_{\text{ASE}}$ such that

$$k_{\text{tot}} = \bar{M}_{\text{ASE}} k_{\text{spont}}. \quad (2)$$

One can easily show that $\bar{M}_{\text{ASE}}$ is an average, over wavelength, space, and angle, of the effective gain seen by the spontaneously emitted radiation as it leaves the disk, having the form

$$\bar{M}_{\text{ASE}} = \frac{1}{4\pi} \int d\lambda \int dV \times \int d\Omega g(\lambda) \rho(\mathbf{r}) G_{\text{eff}}(\mathbf{r}, \Omega, \lambda), \quad (3)$$

where $g(\lambda)$ is the normalized emission line shape [$\int g(\lambda) d\lambda = 1$], $\rho(\mathbf{r})$ is the normalized excited-state concentration in the disk [$\int \rho(\mathbf{r}) dV = 1$], and G_{eff} is the instantaneous effective gain, which represents the multiplicative factor by which the number of photons leaving the disk in any "ray" (group of photons) exceeds the number that were spontaneously emitted.

The calculation of G_{eff} for a particular ray must take into account amplification in the laser medium and reflection and transmission at the boundaries. Figure 5-1 shows the first few steps of a typical ray path. We designate the point of

Fig. 5-1. Cross section of a laser disk, showing the first few steps of a typical ASE ray path.

emission as P_0 , and the successive intersections of the ray with the disk boundaries as P_1, P_2 , etc. In traveling from P_{n-1} to P_n , the ray experiences a gain equal to

$$\exp \left[\int_{P_{n-1}}^{P_n} \alpha(s) ds \right], \quad (4)$$

where $\alpha(s)$ is the gain coefficient at each point s on the line connecting the two points. At the boundary point P_n , a fraction R_n (equal to the Fresnel reflectivity at the interface) of the photons in the ray is reflected back into the disk, while the rest of the power leaves the gain volume. With these definitions, the effective gain for any ray is

$$G_{\text{eff}} = \sum_{n=1}^{\infty} (1 - R_n) \exp \left[\int_{P_{n-1}}^{P_n} \alpha(s) ds \right] \times \prod_{m=1}^{n-1} \left\{ R_m \exp \left[\int_{P_{m-1}}^{P_m} \alpha(s) ds \right] \right\}. \quad (5)$$

Fig. 5-2. (a) 31.5-cm and (b) 46-cm polygonal Nova disk geometries.

(a) 31.5-cm Nova laser disk

(b) 46-cm Nova laser half-disk

In numerical simulations the sum in Eq. (5) is truncated when the power remaining in the ray in question has been attenuated to a negligible value. The spatial, angular, and wavelength integrations are carried out numerically.

An application of the computational model used in our ASE calculations begins with the specification of the amplifier disk geometry. In what follows, we consider only the 31.5- and 46-cm laser disks shown in Fig. 5-2, but the techniques and approximations used apply equally well to other geometries, such as that of a high-average-power zig-zag slab laser. In all the calculations reported here we assumed that the two faces of the disk were parallel to one another; the orientations and positions of the edges could be set arbitrarily.

To investigate the effects of a non-homogeneous gain profile through the disk thickness, we assumed that inside the gain region (delimited by the elliptical masks in Fig. 5-2) the gain coefficient varied with distance from the disk faces, and we ignored any transverse spatial dependence. It is important to note that the gain profile determines not only the amplification of a ray but also the amount of spontaneous emission originating from each point. For most calculations we used a stepwise approximation to the predicted gain profile derived from calculations of the single-pass flashlamp pumping of 2-wt%-doped Nd:glass.³ The true gain profile is probably somewhat flatter than this because of multipass contributions to the pumping and because of radiative effects (e.g., diffusion caused by trapping plus the spatial dependence of the ASE).

We assumed that a fraction of each ray given by the average of the s - and p -polarization Fresnel reflectivities was reflected back into the laser volume from the disk faces. We ignored any translation of the ray in the cladding and assumed that any reflected components reentered the laser glass at the intersection of the incident ray and the edge.

In the computation of the ASE multiplier we parametrized the integrals over volume and angle in terms of the dimensionless product of the wavelength-dependent spatially averaged gain

coefficient $\bar{a}$ and the characteristic dimension D of the gain volume. We took D to be the major axis of the gain ellipse for the 31.5-cm disk and the minor axis of the half-ellipse for the 46-cm disk. The spatial and angle integrals were calculated using a Monte Carlo algorithm. Depending on the problem geometry, an integration for a single value of $\bar{a}D$ tracked between 2×10^4 and 2×10^6 rays.

In most cases the non-wavelength-averaged ASE multiplier M_{ASE} was sufficient to determine an optimum design point. When wavelength-averaging was required, we used the measured value⁴ of the Nd⁺ fluorescence line shape for the LHG-8 Nd:glass used in the disks. The Monte Carlo results determined at a representative set of values of $\bar{a}D$ were fitted to an exponential function of the form

$$M(\bar{a}D) = \exp \left[\sum_{n=1}^N C_n (\bar{a}D)^n \right], \quad (6)$$

where $N = 3$ or 4 gave a good fit. The completely averaged ASE multiplier $\bar{M}_{\text{ASE}}$ was then obtained as a function of the product $\bar{a}_0 D$, where $\bar{a}_0$ is the spatially averaged gain coefficient at line center, by numerically convolving this fit with the line shape function.

To see clearly the effects of a nonuniform gain distribution, we consider the case of a perfect edge cladding, which absorbs all radiation incident upon it. In this case the sum for G_{eff} is truncated when the ray hits the disk edge. Figure 5-3 shows the variation of the non-wavelength-averaged ASE multiplier M_{ASE} in a 31.5-cm disk with $\bar{a}D$ for disks with homogeneous or layered gain profiles. At the lowest values of $\bar{a}D$ the results of the two calculations converge, since in this limit the gain seen by each ray approaches a linear function of the distance traveled. The increase in the ASE decay rate for the layered gain profile at higher values of $\bar{a}D$ is caused by the higher gain coefficient seen by rays that spend more time near the surface. In the exponential gain regime these rays more than compensate for the reduced gain seen by rays that spend more time near the center of the

disk. All the results that follow were obtained assuming the non-homogeneous gain distribution.

Figure 5-4 shows the calculated ASE multipliers for the 31.5- and 46-cm disks, with and without wavelength averaging. The wavelength-averaged results are plotted vs $\bar{a}_0 D$. Averaging over the line

Fig. 5-3. Variation of the non-wavelength-averaged ASE multiplier M_{ASE} with $\bar{a}D$ for a 31.5-cm disk with a perfect edge cladding.

Fig. 5-4. Calculated ASE multipliers for disks with perfect edge claddings.

Fig. 5-5. Calculated dependence of peak gain coefficient on $\bar{M}_{ASE}$ at time of peak gain.

shape of the transition reduces the ASE multiplier to 1/3 to 1/2 of its value at the peak value of the fluorescence. Under typical operating conditions the maximum values of $\alpha_0 D$ in the 31.5- and 46-cm amplifiers approach 3.5 and 2.5, respectively. For both these values the calculation indicates that the maximum radiative decay rate at the peak of the gain would be about 2.6 times greater than the spontaneous rate for a perfect edge cladding.

To determine the effects of this increased decay rate on the predicted peak gains in the amplifiers, we must average the wavelength-averaged ASE multiplier over the time-dependent gain coefficient in the amplifier disk. To do this we used Trenholme's GAINPK amplifier model.⁵ This model requires two cavity radiative transfer parameters, which were fixed by requiring that the model, when used with the ASE multipliers in Fig. 5-4, reproduce the peak gains measured with fused-glass claddings (which we assume are perfect). The effect of a possible increase in the ASE multiplier was then modeled by scaling $\bar{M}_{ASE}$ by a constant factor.

The results of these calculations for both disks are presented in Fig. 5-5, where we plot the calculated peak gain coefficient vs $\bar{M}_{ASE}$ at peak gain. The solid lines (monomial fits to the calculated points) indicate an approximate power-law dependence of the peak gain coefficient on $\bar{M}_{ASE}$ of the form

$$\alpha_{\text{peak}} \propto \bar{M}_{ASE}^{-1/4} \quad (7)$$

for the Nova amplifier disks. A more accurate calculation, taking into account any change in the shape of the ASE curve

vs $\alpha_0 D$, would result in a slightly weaker dependence, since the ASE rates for perfect and imperfect claddings must converge at the lowest values of disk gain.

We also modeled the effect of removing the cladding from the disk-split edge of the 46-cm laser disks. Experience on Nova and off-line measurements show that considerable optical distortion occurs within 1 to 2 cm of these edges. We presume that this distortion is caused by the cladding at the split, which absorbs ASE, heats up, and deforms the disk. By removing the cladding on these surfaces, we might eliminate this distortion and thereby make it possible to use the added beam area in the region near the split. This would lead to a decrease in the spatially averaged gain coefficient in the disk, however, because of the increase in the ASE decay rate caused by rays that are reflected from the unclad surface.

We assumed in the model that the cladding of the remaining edges was perfect, and used an average total Fresnel reflectivity for each ray at the disk split having the functional form discussed in "Calculations of Effects of Imperfect Claddings," in Sec. 3. The predicted non-wavelength-averaged ASE multiplier in this case is shown in Fig. 5-4(b). The calculations indicate that removing the cladding from the disk split would increase the ASE decay rate by about 20% at the standard operating point. Coupled with the scaling law of Eq. (7), this leads us to expect that the peak gain in the disk would be reduced by about 5% in disks with this configuration. This loss in gain is probably enough to cancel the potential increase in beam area.

Spatial Variation of the ASE Decay Rate. If there are paths in the laser medium along which the small-signal gain of a spontaneously emitted ray is very high, the ASE decay rate can vary significantly throughout the medium. For example, one might expect a significantly greater ASE enhancement at the edge of an elliptical disk along the major axis than at the center because of the longer gain paths available. Such an effect would produce spatial inhomogeneities in the peak gain in an otherwise uniformly pumped disk.

We have calculated the expected spatial variation in the local ASE ratio R (defined as the ratio of the stimulated and spontaneous radiative decay rates, rather than as a multiplier of the spontaneous rate) using a method similar to that used to predict the spatially averaged rate. By an approach analogous to that employed in the derivation of Eq. (1), one can demonstrate that $R(\mathbf{r})$ at any position $\mathbf{r}$ in the laser medium is given by an integral over the gain distribution of the form

$$R(\mathbf{r}) = \frac{1}{4\pi} \int d\lambda \int dV' \frac{1}{l^2} \alpha(\mathbf{r}', \lambda) \times G_{\text{eff}}(\mathbf{r}, \mathbf{r}', \lambda), \quad (8)$$

where $\alpha(\mathbf{r}', \lambda)$ is the wavelength-dependent gain coefficient at the point of emission $\mathbf{r}'$ of the ASE ray, l is the length of the path taken by the ray in traveling from $\mathbf{r}'$ to $\mathbf{r}$, and $G_{\text{eff}}(\mathbf{r}, \mathbf{r}', \lambda)$ is the effective gain of the ray, including the effects of loss due to partial reflection at the interfaces. Using the same formalism as in the spatially averaged case, we obtain for the effective gain for the local decay rate model the product

$$G_{\text{eff}}(\mathbf{r}, \mathbf{r}', \lambda) = \exp \left[\int_{P_M}^{P_0} \alpha(s) ds \right] \times \prod_{m=1}^{M-1} \left\{ R_m \exp \left[\int_{P_{m-1}}^{P_m} \alpha(s) ds \right] \right\}, \quad (9)$$

where the starting point P_0 is $\mathbf{r}'$, the terminal point P_M is $\mathbf{r}$, and the remaining quantities are as defined for Eq. (5).

We used this model to predict the spatial variation in a perfectly clad elliptical laser disk. To permit comparison with experimental results, we modeled a 4-wt%-doped LHG-750 disk with the same dimensions as the 31.5-cm Nova disks. The gain profile calculated for this Nd ion density has a surface-to-average ratio of 4.5 (as against 3.0 for the 2-wt%-doped glass in the Nova disks), resulting in a larger overall ASE decay multiplier than those calculated above. We used the same model for the Fresnel reflectivity in the calculation of the local ASE multiplier as in the spatially averaged case.

Fig. 5-6. Calculated values of the spatially averaged ASE multiplier M_{ASE} for an elliptical disk geometry.

With the above assumptions, and taking into account the symmetry of the disk geometry, we calculated the local value of M_{ASE} for values of αD from 1 to 4 over a one-eighth segment of the disk volume on a mesh of evenly spaced points at five radial, axial, and azimuthal positions (100 points total) that extended from the center to the face and edge of the elliptical boundary. To obtain an estimated error of several percent, the Monte Carlo volume integral at each point tracked 10^4 rays. We checked the results of this model by averaging the local values (weighted by the normalized gain distribution) over the volume of the disk and comparing the results with a direct spatially averaged calculation.

Figure 5-6 demonstrates the agreement between the two calculational techniques. The calculation of M_{ASE} using local values required the tracking of 4×10^6 rays; the direct calculation gave the same results with the same accuracy by tracking only 8×10^4 rays, reducing the computational cost by well over an order of magnitude.

The variation of the local ASE decay rate with position in the disk is illustrated in Fig. 5-7, which shows the thickness-averaged ASE multiplier (without wavelength averaging) as a function of position along the major and minor axis of the ellipse for several values of αD . The increase in the decay rate at the edges of the disk at the higher gains is caused by the longer ASE gain paths

Fig. 5-7. Variation of calculated thickness-averaged ASE decay ratio R in an elliptical disk for $\bar{\alpha}D = 1, 2, 3$, and 4: (a) along major axis; (b) along minor axis.

Fig. 5-8. Experimental and calculated values of the thickness-averaged ASE decay rates at the center of an elliptical disk.

available at these positions, which is further enhanced by the relatively large stratification of the gain profile. The corresponding increase in the wavelength-averaged results is considerably smaller, because the effective value of the gain coefficient is less than the value at line center.

We have compared the results of this model with measured values⁶ for the instantaneous excited state loss caused by ASE at the center of the laser disk. A thickness- and wavelength-averaged ASE ratio was calculated at the center of the disk and converted into an effective ASE decay rate using the value $k_{\text{spont}} = 1.28 \times 10^3 \text{ s}^{-1}$ (Ref. 6). The results of this calculation, and the measured ASE decay rate, are presented in Fig. 5-8, which also shows the predicted variation of the

spatially averaged decay rate. The predicted center-line decay rate more closely matches the experiment than do the spatially averaged results, but the predicted ASE decay rate is still about 50% less than the measured value. We have no explanation for this discrepancy as yet.

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Attenuation of New Nova Laser Glass at 1 μm

We are developing loss measurement capabilities to improve predictive performance models for Nova and future lasers. In 1987 we made sensitive optical loss measurements on laser disks and witness samples obtained from Schott Glass Technologies and Hoya Corp. melts that were pilot runs for the Nova glass replacement project. All witness samples and disks met the Nova specification of $0.15\% \text{ cm}^{-1}$ or lower total absorption loss at $1.053 \mu\text{m}$. For future systems, we would like to reduce this figure by a factor of 2 to 3. We used three complementary loss measurement techniques—photometry, calorimetry, and cavity ringdown. We describe these techniques and the results of our measurements in this article.

Photometry. As part of the Nova project, a photometric facility was constructed to measure the optical transmission and reflection (OTR) of large optics. The OTR facility employs laser sources

that give primary measurement capability at both 1.06 and 0.35 μm . In 1987 we added a Nd:YLF laser source operating at 1.053 μm to supplement the Nd:YAG source operating at 1.064 μm . Measurements can be made at either wavelength by a simple exchange of laser heads. The probe laser beam is chopped at 510 Hz and the transmitted signal measured in a 1-Hz bandwidth by a temperature stabilized silicon photodiode. A single amplitude determination consists of 160 samples (14-bit digitization) of the chopped transmitted probe beam.

The absorption in the laser disks was determined by alternately sampling the probe beam with and without a disk in the path. This was facilitated by OTR hardware that transports laser disks rapidly and accurately in and out of the beam. The absorption was measured before edge cladding, with the beam incident normal to the disk edge and passing straight through the long dimension (48- or 63-cm path) of the 46- or 31-cm polygonal disks. The effect of Fresnel reflection was removed in the data analysis. The coherence length of the laser light was short enough that interference of the reflections by the two surfaces did not occur.

We investigated some possible systematic errors in the photometric measurements. The linearity of the detectors is extremely good over a large dynamic range, so that this does not pose a problem. However, the detectors show a slight sensitivity to the position of the probe beam in the detector aperture and to the detector's rotational orientation. These two factors combine to produce a statistical uncertainty of 0.2% in signal level. Another systematic error arises from scattering losses at the input and output optical surfaces, but the importance of this effect is greatly reduced with a long optical path.

As an overall check on system accuracy, we measured the normal-incidence transmission of well-polished laser glass samples 4.2 and 46 cm thick. Extrapolated to zero thickness, the observed transmission was within 0.2% of that expected from Fresnel losses calculated using the index of refraction of the samples at the laser wavelength.

For the photometer measurements on the large disks, the disk edges were not polished but were instead machined to a reasonably specular surface by Zygo Corp. on a Maho milling machine with a 5- μm grit tool. This was done to avoid the expense of edge polishing, which is not required in the cladding operation. We originally planned to "oil-on" well-polished thin plates of laser glass using index-matching fluid to minimize surface scattering. We found it simpler just to spread a thin layer of the fluid on the disk edges. The surface tension of the oil produced a scatter-free (but somewhat optically distorting) surface. The scale size of the distortion was large enough that the detector was able to intercept the full beam. This was ascertained by transmission measurements on a sample having opposed regions that were polished and opposed regions that were finished with a 9- μm grind. When the index-matching fluid was applied to the ground surfaces, the transmission increased to within 0.3% of the transmission through the polished surfaces.

Calorimetry. Calorimetric measurements were performed by heating a thermally isolated sample with a cw 1.064- μm Nd:YAG laser beam.

For highly transparent samples the rate R_1 of heat generation due to absorption is

$$R_1 = \alpha_{\text{abs}} L P_0, \quad (10)$$

where L is the sample length and P_0 is the incident laser power. The rate R_2 of heat leakage from the sample is

$$R_2 = h S \Delta T, \quad (11)$$

where h is the surface heat transfer coefficient, S is the surface area, and ΔT is the sample temperature rise. At thermal equilibrium these rates must be equal, so that we have

$$\alpha_{\text{abs}} = \frac{h S \Delta T_0}{L P_0}. \quad (12)$$

After the input laser beam is turned off, the rate of heat leakage from the

sample is equal to the negative of the rate of decay of the sample heat content:

$$hS \Delta T = - \frac{d}{dt}(CV \Delta T), \quad (13)$$

where C is the sample heat capacity and V is the sample volume. Equation (13) has the solution

$$\begin{aligned} \Delta T &= \Delta T_0 \exp[-(hS/CV)t] \\ &= \Delta T_0 \exp(-t/\tau), \end{aligned} \quad (14)$$

where $\tau \equiv CV/hS$ is the time constant of the sample cooling curve. By replacing CV/hS with τ in Eq. (12), we arrive at the following expression for absorption coefficient:

$$\alpha_{\text{abs}} = \frac{CA \Delta T_0}{\tau P_0}, \quad (15)$$

where A is the face area of the (cylindrical) sample. The absorption coefficient can now be determined from a measurement of P_0 and ΔT and knowledge of C . This technique does not require knowledge of h , a quantity that is often only known to marginal accuracy.

In our measurements the laser glass samples were roughly $1 \times 1 \times 2$ cm and had carefully polished and parallel surfaces. A test sample and a similar reference piece were held in an insulated container that was temperature-ballasted by a 50-L water bath. The temperature difference between the sample and the reference was monitored by thermistors placed in a Wheatstone bridge circuit.

Fig. 5-9. Typical laser calorimetry data. Sample is raised to a new equilibrium temperature $T = T_0 + \Delta T_0$, then laser irradiation is terminated and τ is determined by monitoring sample return to T_0 .

The temperature drift of the system was typically 2×10^{-4} K/hr. A typical measurement requires several hundred milliwatts of laser power and takes 45 min to achieve thermal equilibrium. An example of a calorimetric measurement is shown in Fig. 5-9.

Cavity Ringdown. The cavity ringdown technique has been previously described.⁷ Briefly, the apparatus is a resonator cavity consisting of two highly reflective mirrors into which a short, tunable probe laser pulse is introduced on-axis. The transverse mode of the probe pulse is matched to the lowest-order Gaussian mode of the cavity. The pulse energy in the cavity decays as it is dissipated by scattering, absorption, diffraction, and transmission. By comparing the loss rate (or the "ring-down time") of an empty cavity with that of a cavity containing a transparent sample, one obtains a highly sensitive measure of the total optical loss of the sample.

Results and Discussion. Tables 5-2 and 5-3 summarize the results of absorption measurements using the three techniques just described. The photometer and ringdown measurements agree within 0.01% cm^{-1} on average. The ringdown measurement is consistently higher; this is to be expected, because the long-path measurement is less sensitive to surface scattering losses.

The calorimetric loss coefficients α_{calor} agree very well with the photometric results and are in some cases even lower. However, these values must be corrected for the effects of Nd absorption and fluorescence, using

$$\alpha_{\text{calor}}^* = \alpha_{\text{calor}} + (1 - \epsilon/h\nu)\alpha_{\text{Nd}}, \quad (16)$$

where α_{Nd} is the Nd absorption and ϵ is the energy deposited as heat in the sample by Nd absorption of a photon of energy $h\nu$.

In determining α_{Nd} , we assumed that the Nd^{3+} absorption cross section is $1.7 \times 10^{-24} \text{ cm}^2$ at line center,⁸ and that this value is reduced by a factor of 0.4 at $1.064 \mu\text{m}$ (Ref. 9). Values for ϵ (approximately 0.3 eV) were estimated from the spontaneous emission rate, branching ratios, and concentration quenching rates for the laser glass. After adjustment, the

Table 5-2. Comparison of three loss measurements on Hoya pilot-melt laser glass doped with 3 wt% Nd₂O₃. All values in % cm⁻¹. (Here α_{calor}^* is corrected for fluorescence.)

Melt No.	$\lambda = 1.053 \mu\text{m}$			$\lambda = 1.064 \mu\text{m}$			
	α_{phot}	α_{ring}	α_{Hoya}	α_{phot}	α_{ring}	α_{calor}	α_{calor}^*
18	0.130	—	0.09	0.102	—	—	—
20	0.137	0.133	0.09	0.102	0.105	0.098	0.112
23	0.110	—	—	0.103	—	—	—
25	0.080	0.111	0.09	0.080	0.083	0.072	0.086
27	0.080	—	0.05	0.073	—	—	—
28	0.083	0.108	0.05	0.068	0.081	0.068	0.082
34	0.090	—	0.05	0.057	—	—	—

Table 5-3. Comparison of three loss measurements on Schott pilot-melt laser glass doped with 2 wt% Nd₂O₃. All values in % cm⁻¹.

Melt No.	Disk No.	$\lambda = 1.053 \mu\text{m}$			$\lambda = 1.064 \mu\text{m}$			
		α_{phot}	α_{ring}	α_{Schott}	α_{phot}	α_{ring}	α_{calor}	α_{calor}^*
LB-714	SP4-306	0.092	0.118	0.10	0.084	0.093	0.085	0.096
LB-716	SP4-322	0.087	—	0.08	0.080	0.088	0.080	0.091
LB-723	SP4-310	0.128	0.153	0.13	0.125	0.129	0.128	0.139
LB-735	SP4-314	0.122	—	0.15	0.109	0.112	0.113	0.124
LB-736	SP4-319	0.122	—	0.15	0.112	0.118	0.116	0.127
LB-738	SP4-320	0.139	0.122	0.12	0.103	0.102	0.096	0.107
LB-742	SP4-325	0.085	—	0.12	0.062	0.101	0.086	0.097

calorimeter loss values are in good agreement with the ringdown data although again systematically higher.

As a cross-check on the assumed Nd absorption, we made spectral scans of the loss in each sample using the ring-down apparatus. We found well-resolved Nd absorption peaks on top of what appears to be a baseline loss that increases toward short wavelength (see Fig. 5-10). The estimated Nd absorptions were in approximate agreement with the assumed cross section and Nd concentrations.

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Fig. 5-10. Lossmeter scan of Hoya melt 28 (sample length 2.15 cm). Vertical line indicates estimated loss attributable to Nd ion absorption.

2. Preparation of more durable (i.e., more abrasion resistant) sol-gel anti-reflection (AR) coatings.
 3. Preparation and evaluation of the damage resistance of chemical vapor deposited (CVD) coatings.
 4. Investigation of the source of laser damage in potassium dihydrogen phosphate (KDP) harmonic conversion crystals.
 5. Development of methods for the rapid growth of KDP.
- Clearly much of our research and development effort continues to be focused on improving the damage resistance of optical materials. In fact, optical damage in coatings continues to be a major technical problem in the development of high-peak-power lasers.

Optical Materials and Coatings

Introduction

Our work on optical materials and coatings in 1987 focused on five main areas:

1. Development of damage-resistant high-reflectivity (HR) coatings using sol-gel processes.

The excellent damage resistance of sol-gel-derived AR coatings¹⁰ prompted us to try to extend this technology to HR coatings (see the next article, “High Index Sol-Gel Coatings”). During 1987 a number of oxides were surveyed for the ease with which they can be applied as sol-gel layers. We found that Al₂O₃ gave good single-layer coatings with damage thresholds comparable to those of SiO₂ coatings. We prepared high-reflectivity coatings using alternating Al₂O₃ and SiO₂ layers. Unfortunately, the damage threshold of the resulting coating was less than that of either of the individual materials. Organic-derived impurities may be responsible; we are testing coatings prepared under conditions designed to reduce the level of organic contaminants.

Work to improve the abrasion resistance of sol-gel-derived AR coatings was more successful (see “Abrasion-Resistant Antireflective Coatings”). The addition of small amounts of tetraethyl silicate as a binder in the sol-gel SiO₂ coating increases abrasion resistance without reducing the damage resistance of the coating.

We investigated chemical vapor deposition (see “Chemical-Vapor-Deposited Coatings”) as an alternative means of preparing damage-resistant optical coatings. A reactor was built and operated for preparing CVD coatings at moderate temperatures (250 to 400°C) by thermal decomposition of metal-organic alkoxides. Films of good optical quality were prepared, but the damage threshold is very sensitive to the deposition environment. We are continuing work on the use of oxygen plasma cleaning to remove impurities that may cause damage.

In 1987 we correlated damage in KDP with the presence of organic impurities¹¹ (see “Role of Organic Contaminants in Optical Damage of KDP”). Our work

agrees well with work simultaneously reported by workers in Japan.¹² These studies show that part-per-million concentrations of organic impurities in the solution growth materials and in the KDP crystal product cause low damage thresholds. The classes of organic materials that cause damage have been identified. We are investigating the use of oxidizing process conditions to remove organic species from the crystal growth system and are designing growth tanks and materials that do not produce organic contamination.

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High-Index Sol-Gel Coatings

Introduction. Our major effort on sol-gel coatings in 1987 was directed towards the preparation of high-reflectivity dielectric coatings suitable for use at 1064 nm and consisting of alternating layers of high- and low-index materials.

We used silica as the low-index component. Coatings of this material have very high laser damage thresholds, and we are quite familiar with their fabrication because of our experience in their use as an antireflection coating on Nova optics.¹³

We evaluated TiO₂, ZrO₂, HfO₂, and Al₂O₃·H₂O for suitability as the high-index components. We included TiO₂ despite preliminary results¹⁴ that suggested that its multishot damage threshold was low. We prepared single coatings of these oxides to develop suitable fabrication procedures and to determine coating properties, particularly damage thresholds and refractive indexes.

We then made HR coatings with Al₂O₃·H₂O, the high-index candidate with the highest damage threshold. The coatings had disappointingly low damage thresholds; we are seeking the reasons for this failure.

New Coating Technique for Metal Oxides. In the standard method for preparing oxide sol-gel coatings, a solution of a suitable precursor material (usually a metal organic compound) is applied to a substrate. After the solvent is evaporated, the precursor is converted to the metal oxide by exposure to water and

Table 5-4. 1064-nm damage thresholds and refractive indices of single-layer oxide coatings.

Oxide	Index	Damage threshold (J/cm ²)	
		10-ns, single shot	16-ns, 120 Hz
SiO ₂	1.20	30-40	30-40
Al ₂ O ₃ ·H ₂ O	1.43	20-30	30
ZrO ₂	1.55	10-20	10-15
HfO ₂	1.52	15-20	15-20
TiO ₂	1.75	15-2	2-5

heat. Shrinkage brought about by loss of organic material formerly attached to the metal gives rise to stress in the coating and leads to crazing and peeling.

Dissatisfaction with this method led us to develop an alternative method, in which the precursor material is converted to an oxide before application to the substrate and is applied in the form of a colloidal suspension of particles in a volatile liquid medium. The liquid is evaporated at room temperature, and a stress-free coating is obtained.

We developed techniques for the preparation of suitable colloidal suspensions using high-purity starting materials. To prepare TiO_2 and $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ we hydrolyzed the appropriate alkoxides and then resuspended the precipitated hydrated oxides, adding a small amount of organic base in the case of TiO_2 (Ref. 15) and of mineral acid in the case of $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ (Ref. 16).

The primary hydrolysis reactions are

We prepared suspensions of ZrO_2 and HfO_2 by hydrolyzing the oxychlorides in boiling water¹⁷:

In all cases we obtained oxide suspensions that consisted of discrete monodispersed particles in the 100 to 300 Å size range.

Coating of Candidate High-Index Oxides.

We spin-coated single layers of the colloidal suspensions onto fused silica substrates at room temperature. Table 5-4 shows the refractive indices and laser damage thresholds for these coatings at 1064 nm. The corresponding SiO_2 properties are listed for comparison. Several points are immediately apparent:

- Refractive indices are low relative to e-beam deposited layers because of porosity.
- $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ has a significantly higher damage threshold than ZrO_2 , HfO_2 , or TiO_2 , and it is almost as good as SiO_2 .
- The low multishot damage threshold of TiO_2 is confirmed.

HR Coatings with $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$. We prepared HR coatings using the best high-index oxide, $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$. We spun

Fig. 5-11. 1064-nm damage thresholds of $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}/\text{SiO}_2$ HR coatings subjected to 10-ns single laser pulses.

alternating layers of colloidal suspensions of the two materials onto fused silica substrates. Coating was done at room temperature, with a few minutes' drying time between coats. Layer thicknesses were chosen to give maximum reflection at 1064 nm; about 35 layers were required to give reflectivities of 99%. Figure 5-11 shows single-shot damage thresholds with 10-ns pulses. These thresholds show a wide spread and average about 12 J/cm^2 , a disappointingly low figure in view of the thresholds of the individual layer materials.

One possible explanation for these low thresholds is that the porosity of the coatings allows absorption of detrimental species, either during synthesis or from aerial contamination later. We are investigating potential "cleanup" processes such as the use of UV light with oxygen or hydrogen peroxide.

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Fig. 5-12. Reaction between sol particles and tetraethylsilicate.

Table 5-5. 1064-nm refractive index and damage threshold (for single 10-ns pulses) for sol-binder coatings.

Binder concentration (%)	Refractive index	Damage threshold (J/cm^2)
0	1.22	30-35
10	1.22	32
20	1.22	32
30	1.25	38
40	1.38	37
50	1.40	42

Abrasion-Resistant Antireflective Coatings

The porous silica used as an antireflective coating on Nova optics and as a component in the highly reflective coatings described in the previous article has low abrasion resistance because it is held together only by point contact between particles. In 1987 we attempted to increase the abrasion resistance of such silica coatings by improving particle-to-particle and particle-to-substrate adhesion.

Two methods were successful. In the first, a soluble silicate "glue" was incorporated into the coating suspension; in the second, the standard coating was "stitched" by bombardment with 200-keV helium ions.

Silicate "Glue." This method is based on the reaction of the surface hydroxyl groups on colloidal silica particles with tetraethylsilicate, as shown in Fig. 5-12. The tetraethylsilicate binder, which is soluble in the suspending medium (ethanol), was introduced at various concentrations. The properties of the resulting coatings are shown in Table 5-5.

The refractive index (and thus the AR efficiency) of the coating was unaffected up to a binder concentration of 20 to 30%. Above this concentration the binder was in excess and began to fill in the space between colloidal particles, thereby increasing the refractive index. Coatings produced at 20% binder concentration and above could be drag wiped and survived the adhesive tape test. Laser damage thresholds increase with binder concentration; this may be related to the increase in coating strength.

Ion "Stitching." We bombarded a standard AR silica coating with 200-keV helium ions for various durations. Total ion fluences varied from 1×10^{14} to 3.3×10^{15} ions/ cm^2 . In all cases adhesion between particles was greatly improved, and the treated coatings passed the adhesive tape test. This was the first particulate coating of any type stitched in this manner. At the lowest ion fluences there was little change in the optical properties, but at higher fluences there

was considerable increase in absorption below 300 nm. This was probably due to the formation of absorbing by-products in the high-energy ion beam. Damage thresholds for single 10-ns, 1064-nm laser pulses were reduced to about 15 to 20 J/cm² from an initial 30 to 35 J/cm².

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Chemical-Vapor-Deposited Coatings

Introduction. Chemical vapor deposition is an established technique for growing thin films in the semiconductor industry, but has seen little application in optical coatings. The reviews by Gribov et al.¹⁸ and Jacobson¹⁹ and the experimental data of Verhijen²⁰ suggest that metal oxide formation by CVD may be useful for making optical components such as reflecting dielectric mirrors and polarizers. Films made by CVD are reported to have a high index of refraction, which is an advantage in the fabrication of mirrors because fewer layers are needed.

The damage thresholds of CVD optical films have not been studied. Our interest is to prepare by CVD thin films

suitable for optical coatings and test their damage resistance. These films must be uniform and free from scattering centers, which arise from atmospheric sources or are formed during film growth.

We prepared thin single layers of alumina, hafnia, and zirconia on silica substrates, with layer thicknesses varying about the $\lambda/4$ quarterwave point, and determined their damage thresholds for various preparation conditions.

Chemical Vapor Deposition. We constructed a highly flexible reactor (Fig. 5-13) that allows us to vary source materials and processing conditions without changing the reactor design or configuration. Films were prepared using thermal decomposition at high (400°C) or low (250°C) substrate temperatures or using pulsed plasma excitation to assist the decomposition of the source materials to their oxides.

The silica reactor tube allows visual inspection of the film during growth and provides a chemically clean chamber. Mass flow controllers assure reproducible gas metering. Internal heating (so-called cold wall) methods reduce wall deposits that may contaminate the surface of the growing film.

We developed a new chemical method to speed otherwise sluggish CVD reactions at low temperatures by introducing

Fig. 5-13. Metal-organic CVD reactor.

tertiary butanol into the low-pressure gas stream. Two reactions are involved:

where OR represents an alkoxy group. The tertiary butanol acts as a water transport agent. Simply adding water vapor will result in premature decomposition of the highly water-sensitive alkoxides. Forming water assists the decomposition only at the point of reaction (the substrate surface). The best results have been obtained with the tertiary butoxides of aluminum, hafnium, and zirconium, which have the volatility needed for CVD processing while providing a basis for comparison between the three metal oxide precursors. We are surveying other possible source materials such as mixed alkoxides and chlorides.

The CVD technique produces scatter-free films with high refractive indexes (see Table 5-6). The indexes are highest with high temperature processing and decrease as the temperature is lowered. In hafnia films the index of refraction is essentially independent of process

temperature. Even with the reduced index obtained at lower coating temperatures, fewer layers would be required to prepare mirrors with this technique than with porous-layer sol-gel processing.

The refractive indexes were calculated using a rearrangement of a standard formula²¹ for determining the index of refraction of a single film covering a substrate in air:

$$n = n_s \left(\frac{1 + R^{1/2}}{1 - R^{1/2}} \right)^{1/2}, \quad (23)$$

where R is the reflectance and n_s is the substrate index. The reflectance was determined from transmission minima at or near 400 nm, corrected for the reflectance of the rear surface of the substrate. Optical transmission spectra of films prepared at 400°C show a decrease in UV transmission. Films prepared at 250°C have a higher UV transmission, and the transmission maxima correspond to those of the bare substrate, indicating that the index is homogeneous throughout the film.

Table 5-6 gives the laser damage thresholds for these films. The damage threshold for alumina is most sensitive to growth conditions. The damage threshold is 3 to 5 J cm⁻² for 400°C deposition, independent of thickness. At low temperature, the damage threshold depends on thickness and is 6 to 8 J cm⁻² for a 1060-nm quarterwave coating. Films prepared by the pulsed-plasma process (in which successive 70-Å layers were deposited at low temperature, followed by oxygen plasma cleaning) had damage thresholds of 18 to 19 J cm⁻², independent of thickness.

The damage thresholds of zirconia and hafnia are less sensitive to deposition conditions. The highest damage thresholds for both materials are within the range of thresholds of similar films made by sol-gel processing. We are confident that optimization of the CVD process will produce optical coatings with much higher damage thresholds.

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Table 5-6. Characteristics of alumina, hafnia, and zirconia thin films deposited using high-temperature (400°C), low-temperature (250°C), and pulsed-plasma decomposition.

Method	Refractive index	Thickness ^a (nm)	Threshold ^b (J cm ⁻²)
Alumina			
High temperature	1.58	1040	5.6
High temperature	1.57	2400	4.0
Low temperature	1.50	820	11.4
Low temperature	1.50	1455	3.1
Pulsed plasma	1.52	840	18.6
Pulsed plasma	1.54	1200	19.1
Hafnia			
High temperature	1.79	750	11.5
Low temperature	1.76	920	17.8
Pulsed plasma	1.81	1060	17.8
Zirconia			
High temperature	1.99	525	6.6
High temperature	1.89	1230	9.0
Low temperature	1.82	970	12.8
Low temperature	1.78	1300	15.1

^a Optical thickness based on quarterwave reflection point.

^b Laser damage: single shot, 10 ns, 1.06 μm.

Rapid Growth of KDP

The three-zone system for rapid growth of KDP crystals, described in the 1986 Laser Annual,²² was tested by using supersaturated KDP solutions in the temperature range of 60 to 70°C with and without crystal growth. Each test led to hardware and software improvements; a few more changes are yet to be made. The current version of the system is shown in Fig. 5-14.

Direct Supersaturation Control. The saturator/superheater assembly (at the left in Fig. 5-14) performed as designed with one exception. It had been assumed that the concentration in the saturator is close to its equilibrium value and is thus uniquely determined by the saturator

temperature. It was found, however, that this is so for only a few hours after fresh KDP salt is added to the saturator, while salt particles are suspended. Eventually a crust of KDP forms at the bottom of the saturator (a similar observation has been reported elsewhere²³) and the surface available to dissolution is sharply reduced. Consequently, even with a high interchamber circulation rate, when concentration in the growth chamber approaches that in the feed, the concentration in the feed is not a unique function of temperature but rather depends on the shape of the crust and on the intensity of agitation.

Hence, instead of monitoring the difference between the growth and saturation temperatures, we found it necessary to

Fig. 5-14. Three-zone system for rapid growth of 10 × 10 cm crystals from aqueous solution at constant temperature and supersaturation.

control the supersaturation in the growth chamber directly by allowing the saturator temperature to float. This was accomplished by in situ recalibration of the Foxboro conductivity probe for temperatures from 60 to 70°C and supersaturations from 0 to 5% and incorporating the probe into the feedback control loop. A total of 36 solution samples were taken for gravimetric analysis; six data points were obtained for each temperature. Linear regression equations derived for the above temperature and concentration ranges predicted the concentration to within 0.13%. Thus, supersaturations between 1 and 5% could be controlled with accuracies of 13 to 2.6%, respectively.

Self-Unclogging Return Line. As mentioned in the 1986 Laser Annual,²² the pipeline that returns the supersaturated solution and spurious crystals from the growth chamber to the saturator was prone to clogging. (All other valves that control solution flow are inside the tanks and are thus clog-proof.) This problem was solved in two steps. First, an overflow line was installed so the operator could unclog the return line without upsetting the process. Later, a stainless steel heat exchanger (shown in Fig. 5-14) was added at the entrance to the return line below the growth chamber, which made the system self-unclogging. The heat exchanger and return line are placed in a hot water tank in which the temperature is typically 10 to 15°C higher than that of the solution. As soon as the heat exchanger gets clogged, the temperature inside it goes up and the crystals that clogged it are dissolved, while circulation proceeds through the overflow line.

Conical Growth Chamber Bottom. Replacement of the flat growth chamber bottom with a conical or sloped bottom is an important design improvement still to be made. The flat bottom is the only exception to our design rule that horizontal surfaces in contact with supersaturated solution must be avoided. This rule, and a few similar ones regarding sharp corners, narrow gaps, and use of materials least "sticky" for KDP crystals, express the same idea—minimization of the probability of attachment and accumulation of spurious crystals inside the

growth chamber. This exception, made to simplify design, was based on the observation that the axial jet deflected by the seed holder creates a spot on the bottom towards which spurious crystals are swept; this determined the location of the opening to the return line for that flow arrangement. Later on, as we continued experiments with different combinations of flows, shields (to control x - y growth), and crystal holders, and as it became more and more evident that fluid flow plays a decisive role in growth stability, crystal quality, and the eventual outcome of the entire program, it became clear that solution flow should be determined solely by considerations of growth kinetics. Accordingly, the removal of spurious crystals accumulated at the bottom should be treated as a totally independent problem.

Another consideration favors a conical or sloped bottom, even if a flow that was optimum for crystal growth and for tank bottom cleaning could always be arranged. Because sweeping is possible only when the fluid velocity in the vicinity of the bottom is high enough, some particles may be resuspended, entrained into the overall flow inside the growth chamber, and carried through the axial pump, and may then strike the surface of the growing crystal. Although this has never been observed in any of our growth runs in which crystals were grown upward and flow impinged downward, the possibility of a crystal being activated by the impact of a spurious crystal particle is a real concern^{24,25} and is analyzed below.

Particle Bombardment of a Growing Face. In the situation shown in Fig. 5-15, a KDP particle in the vicinity of a (101) face has two velocity components. It approaches the face at the velocity u_z , rapidly decelerating from the nozzle velocity u_0 to a terminal velocity u_t . At the same time it travels along the face at the velocity u_x , which decreases from approximately u_0 as the one-seventh power of the distance $z/\sqrt{2}$ to the surface within the turbulent boundary layer (of thickness δ), and directly with $z/\sqrt{2}$ in the viscous sublayer (of thickness δ_0).

Any change in the nozzle velocity u_0 affects both velocity components u_z and

u_x in the same way, but deceleration in the z -direction is much more rapid than deceleration in the x -direction, especially for smaller particles. On the other hand, the distance from the point where the particle enters the boundary layer and begins to decelerate is much greater along the face than perpendicular to the face. Thus, depending on the face size x_0 , large enough particles will always strike the surface regardless of the nozzle velocity. For smaller particles there should be a range of nozzle velocities below which (and down to $u_0 = 0$) particles will settle on the surface at their terminal velocity u_t , above which particles will strike the surface at a velocity greater than u_t , and at any velocity within that range particles will clear the surface.

The assumption that a particle acquires its x -velocity component only after it enters the boundary layer is probably too conservative, because the deflection of the flow from vertical occurs smoothly and well above the boundary layer. This assumption is balanced, however, by the assumption that there is no vertical velocity component in a fluid flow within the boundary layer. The problem is therefore reduced to computation of the distances $2\delta/\sqrt{2} > z > D/2$ to the surface (in our flow arrangement the particle crosses the boundary layer at a 45° angle) and $0 < x < x_0$ from the leading edge. If x approaches x_0 while z is still greater than the particle radius $D/2$, the particle will not strike the crystal surface; otherwise it will strike the surface.

The velocity u_z necessary for the computation of z can be obtained from the simple force balance equation

$$\frac{\pi}{6} D^3 \rho_s \frac{du_z}{dt} = \frac{\pi}{6} D^3 \rho_s - \rho_l g - 3\pi \rho_l \nu u_z D (1 - w) - \frac{\pi}{8} C_D \rho_l u_z^2 D^2 w. \quad (24)$$

During deceleration the net gravitational force on a particle (the first term on the right) is not fully balanced by the friction and pressure drag forces (the second and third terms, respectively), thus producing a net force on the particle. For a Reynolds number $Re = Du_z/\nu \leq 1$, the weight factor w in Eq. (24) is zero: w

Fig. 5-15. Motion of a particle entrained in the flow near the crystal surface.

increases roughly as $w = \log Re^{1/3}$ for $1 < Re < 10^3$ until it becomes unity at $Re \geq 10^3$.

When steady state is approached, the net force acting on the particle becomes zero, and u_z becomes the terminal velocity u_t . Then Eq. (24) yields Stokes's formula (for $Re < 1$).

$$u_t = \frac{D^2(\rho_s - \rho_l)g}{18\rho\nu}, \quad (25)$$

and Newton's formula (for $Re > 10^3$),

$$u_t = 1.74 \sqrt{\frac{D(\rho_s - \rho_l)g}{\rho}}, \quad (26)$$

where ρ_s and ρ are the solid and liquid densities, ν is the kinematic viscosity, and g is the acceleration due to gravity; in Eq. (26) we have taken the drag coefficient $C_D = 0.44$ for spheres.

The velocity u_x required for computation of x is given by

$$u_x = u_0 \left(\frac{z\sqrt{2}}{2\delta} \right)^{1/2}, \quad \frac{2\delta_0}{\sqrt{2}} < z < \frac{2\delta}{\sqrt{2}}, \quad (27)$$

$$u_x = u_0 \frac{z\sqrt{2}}{2\delta_0}, \quad \frac{2\delta_0}{\sqrt{2}} < z < \frac{D}{2}. \quad (28)$$

The turbulent boundary layer thickness δ , viscous sublayer thickness δ_0 , and

velocity u_0 at the upper boundary of the viscous sublayer are given by

$$\delta = \frac{0.376x}{\text{Re}_x^{1/5}}, \quad (29)$$

$$\delta_0 = \frac{67x}{\text{Re}_x^{0.9}}, \quad (30)$$

and

$$u_0 = \frac{2u_n}{\text{Re}_x^{0.1}}, \quad (31)$$

where $\text{Re}_x = u_n x / \nu$.

Fig. 5-16. Variation of impact velocity and impact point with the size of large spurious crystals.

Computation for $x_0 = 10 \text{ cm}$ indicates that KDP particles with $D > 250 \mu\text{m}$ will strike the crystal surface at any nozzle velocity. Typical nozzle velocities in the three-zone system are 1 to 2 m/s. Figure 5-16 shows the impact velocity vs D for $u_n = 1 \text{ m/s}$. As D increases, particles strike the surface closer to the leading edge, and their impact velocity increases much faster than their terminal velocity.

To become entrained in the nozzle flow, these large particles must be kept in suspension or be resuspended from the bottom. Given their relatively high terminal velocities, resuspension seems unlikely. Nevertheless, this could be an additional argument in favor of a conical or sloped growth chamber bottom, because such shapes would virtually eliminate resuspension.

Terminal velocity decreases rapidly as particle size decreases, in accordance with Eq. (25), and the likelihood of entrainment in the flow increases. At the same time, Fig. 5-17 shows that it becomes increasingly difficult for a particle to strike the surface. All particles in Fig. 5-17 clear the surface over a certain range of nozzle velocities, but for smaller particles there is essentially no limit to the velocities that allow safe passage. In

Fig. 5-17. Variation of minimum clearance between the particle and (101) face with solution velocity in the nozzle jet and the size of small spurious crystals.

addition, depending on particle size, some velocities yield maximum clearance between the particle and the trailing edge of the (101) face, where closest approach (in the absence of impact) is usually achieved. For all practical purposes, nozzle velocity need not be greater than 2 m/s, and the interval of velocities from 1 to 2 m/s that has been used for growth practically coincides with that required by particle bombardment considerations.

Solution Stability. By stability we mean an absence of spontaneous precipitation (what we call "solution collapse") over a prolonged period of time. Solution collapse is a rapid process that must be distinguished from the slow breeding of spurious crystals in an otherwise stable solution. Fast, clear growth requires uniform surface concentration, which is best achieved with turbulent flow. But turbulent flow induces collapse. It is possible, in principle, to achieve conditions in which the surface is equally accessible for diffusion at all points with a laminar flow perpendicular to each crystal face. But the laminar diffusion boundary layer is roughly three times thicker than the turbulent boundary layer, so a significant increase in bulk supersaturation would be required; the result would be a solution just as unstable as if turbulent flow were used.

Solution stability improves with purification and with the elimination of insoluble particles. This has been accomplished in the three-zone system by continuous filtration. Further improvements in solution stability should come as a result of continued theoretical and experimental study of fluid flow and growth kinetics as well as from solution purification efforts related to laser damage.

In the meantime, we developed a solution stability map that shows safe combinations of supersaturations and nozzle velocities. During our tests, the solution was considered stable if it did not collapse overnight. An example of the map for 60°C is given in Fig. 5-18. With the decrease of solution velocity, stable supersaturation approaches a limit of about 5%. This is close to the 5.4% (at 29.5°C) reported for the three-zone system with laminar flow.²³ It follows from Fig. 5-18 that a practical range of supersaturations

Fig. 5-18. KDP solution stability in the growth chamber.

is 1 to 3.5% for axial flow velocities of 0.5 to 2 m/s. This translates into growth rates of 5 to 20 mm/day, depending on seed plate condition.

Growth Experiments: Control of Crystal Shape and Cracking. KDP crystals were grown from (101) seed plates measuring 10×7.5 cm at 5 to 15 mm/day in the z-direction at 60 and 70°C. We used a second-generation axial pump with a slot-shaped nozzle cross section that gave the required average nozzle velocity with lower rpm.

Initially, the longer axis of the nozzle cross section was aligned with the long dimension of the seed (see Fig. 5-19). In the course of growth, the pyramidal apex drifted to the right until it was directly under the center of the jet, where the solution velocity should have its maximum. Pitot tube measurements confirmed the velocity profile shown in Fig. 5-19. They also confirmed that the velocity was constant along the greater part of the longer axis of the nozzle cross section. Clearly the (101) face, being in the zone with lower solution velocity, was growing more slowly than the (101) face, and the drift of the apex stopped only after normal growth rates for the two faces became equal.

With a high overall growth rate, x-y growth is inevitable even in the presence of Cr^{3+} ions.²⁶ This is a major concern, not only because the growth is usually flawed (as long as z-growth is clear, the center of the boule is usable) but because the internal stresses that accompany this type of growth lead to cracks.²³

Fig. 5-19. Longitudinal flow arrangement and stages of growth of an unshielded KDP crystal from a (101) seed.

Fig. 5-20. Transverse flow arrangement and stages of growth of a shielded KDP crystal from a (101) seed.

In Fig. 5-19, crack areas are shown at the edges between the (100) and (010) faces.

For the next growth run, the crystal holder was redesigned to include two parallel shields in contact with two opposite (100) and (100) faces. It was impossible to shield the remaining two prismatic faces without creating a trap for spurious crystals and ruining the entire flow concept. We also turned the pump nozzle 90°, as shown in Fig. 5-20.

No cracks developed in this growth run, even though the unshielded prismatic faces were growing about as fast as in the previous run. As long as prismatic growth is one-dimensional it is stress-free and thus acceptable. The overall crystal shape in Fig. 5-20 was much closer to the desired one.²⁷ Both (101) and (101) faces grew at the same rate, so the relative position of the apex did not move. The apparent increase in size of the (101) face is merely the result of (010) face growth. If it is desired to minimize the size of the (101) face, the solution velocity there must be higher than at the other faces. In general, the largest faces are those with the lowest normal growth rates. Thus, in principle, a great variety of growth shapes can be obtained by manipulating the flow.

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Role of Organic Contaminants in Optical Damage of KDP

In 1987 we significantly advanced our understanding of laser damage in KDP crystal, a nonlinear optical material used for frequency conversion in high-power laser systems. The discovery of part-per-million concentrations of organic impurities in growth materials and in "best available" KDP single crystals led us to look for a correlation between the presence of these contaminants and low laser damage thresholds. We established this correlation, and identified and quantified a number of organic compounds in KDP feedstocks and single crystals.^{28,29}

Reduction of the organic impurities in KDP growth materials and development of growth tanks that resist further contamination are the goal of our current research efforts. We believe the result will be consistently high-damage-threshold crystals that can perform satisfactorily in single-shot and high-average-power laser systems.

Over the last several years we have tested more than 300 KDP crystals, from commercial sources and research laboratories, using a single-shot Nd:YAG laser (1-ns pulse length, 1064-nm wavelength). Damage thresholds consistently fall in the range of 3 to 8 J/cm², and damage sites are randomly distributed in the bulk material.^{30,31} Models describing the laser damage phenomenon suggest that solution-wetted absorbing species are concentrated at specific sites in the crystal, and that the damage sites (micron-size fractures) arise from an overpressure resulting from temperature excursions at these sites during laser irradiation.^{32,33} The identification of trace amounts of absorbing materials in KDP therefore became the subject of our investigation.

We had previously investigated the possibility of bacterial contamination in our moderate-temperature (50 to 75°C) growth solutions. We were unable to verify the presence of viable bacteria as a source of contamination in KDP grown in these experiments, although other researchers^{34,35} had reported the presence of microbes in KDP crystals. We carried this investigation further in 1987 by analyzing a variety of KDP samples using the *Limulus* amoebocyte lysate (LAL) assay, an extremely sensitive test for the presence of viable or nonviable gram-negative bacteria.³⁶ The LAL assay detects endotoxin, a breakdown product from the cell wall of this type of bacteria. The tests were run for us by Cape Cod Associates, which produces the assay reagents.

Samples tested included KDP powder (growth material) from three manufacturers and single crystals from a variety of sources. In all eight samples there was no significant bacterial contamination; in seven samples the contamination was

less than 1 pg endotoxin per gram of KDP. A great deal of effort was made to eliminate any interference of KDP with the LAL end point, and three different end point methods were compared to validate these tests. In spite of the absence of significant bacterial contamination in these growth materials and single crystals, the laser damage thresholds remained in the 3 to 8 J/cm² range. This led us to look for other sources of absorbing inclusions.

With our discovery that the concentration of total oxidizable carbon (TOC) was several ppm in both KDP crystals and growth materials, we began a systematic search for a correlation between TOC and laser damage threshold. Identification and quantification of specific organic impurities was made possible by computer-controlled gas chromatography/mass spectrometry (GC-MS) analysis. Methylene chloride extracts of growth media and of redissolved KDP crystals yield residues that can be analyzed using GC-MS. With the addition of an internal standard, it is possible to estimate the amount of total extractable

material and the amount of individual compounds present at concentrations of 10 ppb or more.

Figure 5-21(a) shows a representative total ionization plot (chromatogram) of organic impurities extracted from a KDP growth solution from our laboratory. The concentration of extracted organic material was approximately 5 ppm. The isolated compounds, which were identified by their characteristic mass spectra, included alkyl amines, short- and long-chain fatty acids, alcohols, phthalates (plasticizers), and phenols. With minor differences, these types and quantities of compounds were found in all KDP supplied as starting material for growth solutions.

Figure 5-21(b) shows the chromatogram of organic impurities extracted from a KDP single crystal grown from the solution of Fig. 5-21(a). Comparison of the two chromatograms shows that the crystal concentrated specific compound types into its matrix, typically long-chain fatty acids, phthalates, and phenols.

The ameliorating effect of laser annealing on the damage threshold of KDP crystals is a well-established phenomenon³⁷ closely related to the problem of laser damage. Laser annealing is accomplished by irradiating the same site on a crystal starting at fluences below the single-shot "1-on-1" damage threshold and increasing the fluence until the "*n*-on-1" damage threshold (*n* shots on one site) is reached. The *n*-on-1 threshold is generally much higher than the 1-on-1 threshold.

We investigated the effect of laser annealing on the makeup of the organic contamination by annealing half of a 5 × 5 × 1 cm crystal and then analyzing both halves separately by GC-MS. Annealing of this large volume was accomplished with a 1064-nm high-repetition-rate scanning laser. Figure 5-22 shows the total ion chromatograms of extracts of the two halves. The concentrations of several compounds, particularly the long-chain fatty acids, phthalates, and phenols, are significantly lower in the extract from the annealed crystal. This is the first direct evidence of a change produced in a KDP crystal by laser annealing.

Fig. 5-21. Chromatogram of methylene chloride extract from (a) a KDP growth solution, (b) a KDP crystal grown from that solution.

Fig. 5-22. Chromatogram of methylene chloride extracts from two halves of a $5 \times 5 \times 1$ cm KDP single crystal. (a) Laser annealed at 1064 nm with a rastered laser scan; (b) not annealed.

We compared the extractable organic contamination in a number of KDP samples with the corresponding laser damage densities and thresholds. Figure 5-23(a) shows the damage thresholds in a crystal with a moderate (~ 1 ppm) concentration of extractable organic contamination, as identified in the chromatogram shown in Fig. 5-23(b). In contrast, Fig. 5-23(c) shows the highest damage threshold of any of over 300 crystals that we tested. The organic analysis of this crystal [Fig. 5-23(d)] indicates no measurable organic contamination (< 10 ppb) by comparison with an internal standard. Nominal damage thresholds in Fig. 5-23 refer to n -on-1 tests.

Having established that certain classes of organic compounds are incorporated

into growing KDP crystals, and that some of these compounds are photochemically changed by laser annealing, we measured the extinction coefficient at $1 \mu\text{m}$ of the extracted material from KDP crystals. We found extinction coefficients of between 900 and $1500 \text{ cm}^2/\text{g}$. Models of thermomechanical fracture^{32,33} indicate that these extinction coefficients are sufficient to cause damage if the organic compounds are concentrated by about a factor of 10^4 in the crystal. The concentration of these materials is consistent with the observed morphology of damage and with the behavior of fatty acids in aqueous salt solution. Fatty acids are known to condense into cell-like structures called micelles.³⁸ The micelles themselves can aggregate and, by a

Fig. 5-23. Laser damage thresholds and chromatograms of methylene chloride extracts from two KDP crystals with very different extractable organic contamination. (a), (b) Contamination ~1 ppm; (c), (d) contamination <10 ppb.

detergent-like action, can concentrate particulate matter.

We are investigating methods for removing organic impurities from KDP growth solutions. We have found ozone treatment extremely effective in oxidizing these impurities to carbon dioxide and water, reducing their concentrations to below detection limits (10 ppb). We have tested several absorbing materials and found that activated charcoal removes significant amounts of organic compounds from KDP solutions. We are working on making the cleaning procedure simple and economical for commercial application to large quantities of material.

Although we have established a strong case for the role of organic contaminants as a source of laser damage in KDP, we should point out the importance of optimizing all the growth conditions for production of high quality KDP crystals. The benefit of filtering the solution through a 0.2- μ m filter (or even multiple-pass filtering) cannot be overemphasized. A KDP solution that was filtered three times produced crystals with decreasing damage site densities after each

filtration. The effects of other parameters we have studied are discussed in previous reports.^{39,40}

Designs and materials for growth tanks that produce consistently high quality single crystals are under development. Most plastics are considered to be undesirable if we wish to eliminate organic carbon compounds in growth solutions. We are therefore conducting materials compatibility studies to determine if any plastics, especially Teflon or Lucite, can be tolerated in long growth runs. Ideally the tank materials should be impervious to strong oxidizing agents so that solutions can be "cleaned" inside the growth tank, thus avoiding recontamination during transfer of cleaned solutions. The development of appropriate tank designs should result in large, high-damage-threshold KDP crystals that are commercially available in the near future.

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Advanced Nonlinear Materials

Introduction

Over the last few years, experiments in laser-driven ICF have shown that targets use drive laser energy more efficiently at short wavelength. The optimum wavelength will be a compromise between target efficiency and laser technology, and will almost certainly be between 250 and 500 nm. The most efficient laser architecture to reach this wavelength range uses harmonic conversion of light from an efficient near-IR laser source.

Potassium dihydrogen phosphate (KDP), the harmonic conversion material used in present ICF lasers, is not ideal for future ICF drivers because, as currently grown, it has a low optical damage threshold and because its conversion efficiency is compromised by the trade-off between its acceptance angle and its nonlinear coefficient. To be economically viable, future ICF lasers (which will be very large) will require efficient, cost-effective harmonic generators. We are therefore working to develop and eventually deploy a material that is more efficient and less expensive than KDP. One such material (l-arginine phosphate, or LAP) is under development.

Materials that improve on KDP are likely to be an inexpensive choice for many other applications in which high-power lasers are involved. We therefore expect that despite the relatively narrow near-term focus, progress in the ICF application will also result in inexpensive new materials for other applications.

The most promising material for multi-megajoule (MMJ) ICF lasers is deuterated l-arginine phosphate, or d-LAP. The next article describes our work in 1987 on this material. We have continued growth scaling studies, and have grown large, high-optical-quality crystals of over 100 cm³ useful volume. Only about one-third as much d-LAP as KDP would be required for the laboratory microfusion facility (LMF); therefore, despite the slightly higher cost of the raw materials, the cost

of frequency conversion based on d-LAP is likely to be less than that of a KDP-based subsystem. The damage threshold of d-LAP is 2 to 3 times that of standard KDP.

We are looking for materials even better than d-LAP for the ICF application. Crystals grown from aqueous solution or with low-temperature Bridgman methods are likely to be the most economical; we have identified several classes of molecular crystals in these categories. In "New Organic Materials for Harmonic Generation" we report our work on molecular crystals based on amino acids. These molecules are particularly attractive because they include both a chiral center and a relatively strong harmonic-generating unit,

and because (being ionically bonded) *they are mechanically more robust than hydrogen-bonded crystals*. Most are also more nonlinear than KDP, and they possess a wide range of birefringence (0.01 to 0.15) and are optically biaxial.

We have developed a rapid method, based on white-light refractive index measurements, for estimating the noncritical wavelengths of nonlinear materials. We have used this technique, described in "Estimating Noncritical Wavelengths in Organic Salts," to select 25 candidates for more precise and time-consuming microcrystal measurements.

In "Dispersion Measurements Using the Spindle Stage Refractometer" and "Phasematching Measurements in Biaxial Crystals" we describe new characterization techniques for nonlinear optical materials. These techniques are designed to be carried out on irregular microcrystals with typical dimensions of 0.05 to 0.25 mm. Using microscopic goniometers to hold and orient the microcrystals, we can measure refractive indices from 400 to 1100 nm and can directly determine the phasematching locus and the nonlinear coupling and angular acceptance along that locus. We can also measure the temperature bandwidth. By using such easily obtainable samples we can quickly characterize a large number of materials, and can concentrate on growing only promising materials.

Using these methods, we have identified several amino acid salts that are better than KDP. These include LAP and

other arginine salts such as the fluoride, chloride, bromide, and acetate. There are in fact many crystals in this class, and many of them are efficient nonlinear materials for high-power lasers.

We are also interested in developing nonlinear materials for high-average-power applications. (Average power considerations are relevant to the LMF, but not to near-term MMJ experiments designed to demonstrate laboratory micro-fusion.) Under average-power conditions, thermal distortions arise from the linear optical absorption of the crystals. Thus high average power calls for nonlinear materials with very low absorption, high resistance to thermal fracture, and tolerance for thermo-optic effects and for imperfect beam quality. For the average-power application, we have characterized the thermomechanical properties of d-LAP.

This subsection reviews our progress in new nonlinear materials for LMF applications, including progress in d-LAP, a description of the microcrystal characterization techniques, and the application of our strategy for choosing candidates for the fusion application.

Author: D. Eimerl

Deuterated L-Arginine Phosphate

We recently reported⁴¹ a significant new nonlinear material, the deuterated analog of l-arginine phosphate monohydrate, known as d-LAP. We have continued

to characterize the optical and mechanical properties of d-LAP, and continued the program to increase the size of d-LAP crystals.

L-arginine phosphate is one of the chiral organic materials we are studying for laser fusion applications (see the next article). Both LAP and d-LAP are easily grown from aqueous solution, are monoclinic (space group $P2_1$), and are phase-matchable for all processes for which KDP is phase-matchable. With the possible exception of harmonic generation to wavelengths shorter than about 270 nm, their threshold powers⁴² are a factor of 2 or 3 lower than those of KDP or barium borate. In the absence of optical absorption, then, these materials are considerably more efficient harmonic converters, for a given laser source, than KDP. They are relatively inexpensive, exhibit high damage thresholds, and achieve higher efficiency than KDP using smaller crystal volume. Table 5-7 summarizes the linear and nonlinear optical properties of LAP and d-LAP; Fig. 5-24 shows LAP phasematching curves for second, third, and fourth harmonic generation.

Crystal Growth. When LAP is dissolved in D_2O , the NH and OH protons are replaced by deuterium; the CH_2 protons are unaffected. After three recrystallizations from 99.5% D_2O in covered flasks, the OH and NH sites are 95.5% deuterated, as determined by NMR. The deuteration of these sites can be brought arbitrarily close to 100% by repeated recrystallization. Using deuterated starting material and D_2O , the crystal growth proceeds as for LAP. We have obtained high optical quality $2 \times 2 \times 0.7$ cm crystals of d-LAP by this method.

A more thorough study of d-LAP growth and growth scaling was conducted by Cleveland Crystals, Inc., under contract to LLNL. The starting material was prepared by reacting l-arginine with P_2O_5 in 95% D_2O , and purifying by recrystallization. The solubility of d-LAP in D_2O was determined for various values of pH and temperature. At the stoichiometric pH, d-LAP grows as plates, but the growth habit can be controlled by adjusting the pH. At lower pH, the habit is elongated,

Table 5-7. Optical constants for LAP and deuterated LAP.

a. Sellmeier parameters (λ in μm) for d-LAP					
$n^2 = 1 + s_1 + s_2\lambda^2 + \frac{s_3}{(\lambda^2 - s_4^2)}$					
	s_1	s_2	s_3	s_4	
α	2.2352	-0.00683	0.0118	0.1208	
β	2.4313	-0.0143	0.0151	0.1463	
γ	2.4484	-0.0115	0.0172	0.1513	
b. Nonlinear optical constants (pm/V) for LAP					
	$\alpha\alpha$	$\beta\beta$	$\gamma\gamma$	$\beta\gamma$	$\alpha\gamma$
α	0	0	0	-0.58	0
β	0.4	0.92	-0.84	0	-0.58
γ	0	0	0	-0.84	0

Fig. 5-24. Empirical phase-matching loci for LAP. Solid lines are loci calculated from the Sellmeier formula; points on the curves are experimental data; asterisks denote the locations of the two optic axes.

Fig. 5-25. Crystals of d-LAP grown by Cleveland Crystals, Inc.

primarily along the b -axis. At pH 4.4 and above, the habit is prismatic and the growth rates in the b and c directions are about equal. The large d-LAP crystals shown in Fig. 5-25 were grown by this method.

Damage Threshold. The damage threshold of d-LAP remains high as sizes increase. We test each sample of the larger crystals grown at Cleveland Crystals. Samples are tested using 1-ns pulses at 1064 nm; most show thresholds between 10 and 13 J/cm². In earlier samples, sites were occasionally found at

which the threshold was lower, but the material has become more consistent. Some uncoated samples exhibited rear surface damage at about 22 J/cm² with 10-ns pulses at 30 Hz.

UV Properties. As currently grown, d-LAP exhibits a weak blue fluorescence under irradiation with 355-nm (3ω) light. Extensive tests at this wavelength indicate that the samples do not experience damage or solarization. Under irradiation with light below about 270 nm they darken, however, apparently undergoing a photochemical process. If this process

is intrinsic, it would make the crystal generally unsuitable for UV applications below 270 nm.

The fluorescence depends strongly on position in the crystal and is certainly associated with inclusions. There is no correlation between the sources of fluorescence and damage. The chemical nature of the fluorescing inclusions has not been determined, but they are probably other organic molecules, such as aldehydes, or perhaps benzene ring compounds and their derivatives, such as tryptophan. The fluorescence spectrum closely matches that of guanidyl, suggesting that energy is transferred from absorbing inclusions to guanidyl ions, which then fluoresce. It may be possible to reduce the fluorescence by purifying the starting materials for crystal growth.

This fluorescence is not necessarily a disadvantage, however. The output of a third harmonic generator will cause fluorescence, which will be monotonic in the 355-nm output energy. The fluorescence can therefore be used to measure the conversion efficiency without calorimeters in the target chamber. This would be of considerable value in fusion experiments.

Temperature Bandwidth. In contrast to uniaxial crystals, in biaxial crystals such as LAP the linear temperature sensitivity $\beta_T = \partial\Delta k/\partial T$ (where Δk is the wave vector mismatch) varies along the phasematching curve. For LAP, β_T actually has zeros for both second and third harmonic generation of 1064 nm and for both Type I and Type II processes. At these zeros, which happen to be close to the optimum phasematching direction, the temperature bandwidth is very high. The threshold powers at these zeros are about an order of magnitude higher than the minimum threshold power quoted earlier, but are nonetheless low enough to provide efficient conversion. For example, the minimum threshold power for Type I second harmonic generation is about 68 MW, whereas it is about 120 MW at the temperature sensitivity zero. Thus LAP and d-LAP are capable of temperature-insensitive operation, but at some cost in threshold power or efficiency.

Average Power. The possibility of high temperature bandwidth makes d-LAP a candidate for medium-average-power devices. However, under thermal load, the crystals will also generate stress, which could lead to fracture along the cleavage plane. We have used estimated elastic constants and thermal expansions to calculate the thermal fracture resistance of d-LAP.⁴² This quantity also varies along the phasematching curve, but at the temperature sensitivity zero it is about twice as high as in KDP. This suggests that d-LAP, whose absorption and thermal fracture properties are similar to those of KDP, has only a small advantage over KDP in average power applications, despite its potentially high temperature bandwidth.

Summary. Deuterated LAP is under development as the leading candidate for use in multimegajoule fusion experiments. In addition to having favorable optical properties, it is amenable to cutting, polishing, diamond-turning, and sol-gel anti-reflection coating. These properties suggest that chiral organic crystals will be generally attractive for high power laser applications, and in particular for laser-driven fusion.

Author: D. Eimerl

New Organic Materials for Harmonic Generation

Introduction. We have examined three groups of crystalline compounds as possible candidates for harmonic generation⁴³⁻⁴⁶: the salts of tartaric acid, amino acids, and N-acetyl amino acids (Fig. 5-26).

All the compounds we examined have at least one chiral carbon (coordinated with four different groups), which ensures that every crystal is noncentrosymmetric and therefore capable of harmonic generation. The second harmonic generation (SHG) signals of powders and the average principal refractive indexes were measured for 60 compounds. This study indicates that the likelihood of finding crystals that are efficient generators of visible or near-UV light among these compounds is very high.

Fig. 5-26. Chemical structures of crystalline compounds studied as possible second harmonic generators.

We refined our materials screening process by developing an empirical method for estimating noncritical wavelengths without making detailed dispersion measurements.⁴⁷ This allowed us to identify, at the powder level, crystals that are most likely to be useful for frequency conversion to wavelengths of interest to the ICF or submarine laser communication (SLC) programs. In addition, this study generated a data base that can be used to test models of structure-property relations in molecular crystals.

Chiral organic salts are produced by reacting the pure enantiomeric (i.e., right- or left-handed) forms of chiral organic acids with a variety of organic or inorganic bases, or by reacting chiral organic bases with a variety of acids. An example of such a material is LAP, which is being evaluated as a potential alternative to KDP in future high power lasers for ICF.

These compounds have a number of attractive properties⁴⁶:

- The use of pure chiral enantiomers guarantees noncentrosymmetric crystals.
- The nonlinear coefficients of some members of this group are as much as 4 times higher than those of KDP.
- These compounds have high optical transparency between 1.06 and 0.355 μm .
- Their mechanical properties are similar to those of ionic crystals such as KDP.

Because of the chirality of the molecules, the crystal structures cannot have a mirror plane as a symmetry element. Most of these compounds therefore belong to point groups 2 (monoclinic) and 222 (orthorhombic); a few belong to the triclinic group 1. The point groups of about two-thirds of our samples are known from published crystal structure determinations.

Preparation of Chiral Organic Salts.

We used two methods to synthesize the

salts of l-tartaric acid. The first method was simply reacting the acid with the hydroxide or carbonate of the desired cation and allowing the aqueous solution to evaporate at room temperature. The second method, gel growth, was used for compounds that were insoluble in water (i.e., they immediately precipitated as a fine powder when the acid and base were mixed). Growth times using the gel method generally ranged from a few days to several weeks. We grew crystals of the magnesium, barium, calcium, strontium, and zinc salts of l-tartaric acid using this method.

In growing crystals of the C₂H₄O₂(COO)₂H₂ di-M tartrates, where M = Li, Na, K, Rb, and Cs, we used a mixture of 1 mole tartaric acid and 5 moles MOH. This avoids the formation of C₂H₄O₂(COO)₂HM (M-hydrogen tartrate), which is much less soluble than the di-M tartrate. For M = Tl, we used an equimolar mixture of tartaric acid and Tl₂CO₃. All the di-M tartrates were prepared by evaporating the mixtures.

All amino acids were purchased and used without further purification. They were dissolved in water and the solutions were evaporated at room temperature. Small (0.5 to 1 mm) crystals of good quality were picked out of the solution before it evaporated to dryness. (This "harvesting" technique is also a way of purifying the compound by recrystallization.)

The exact stoichiometry of the crystals we formed by evaporative growth was not always known. For most of the amino acids, we could make reasonable guesses on the basis of crystallographic data and on published descriptions of synthesis procedures.

Nonlinear Properties of Chiral Organic Salts. The powder-SHG signals of crystalline compounds are a crude measure of their SHG efficiencies. We have

established⁴⁸ that the powder signal is generally within a factor of 2 of the theoretical value for an ideal (i.e., morphologically unbiased) sample. From the powder signal in a good quality sample, it is possible to estimate the effective nonlinearity with an accuracy of about 50%.

We discovered 21 compounds with powder signals stronger than or equal to that of quartz, 9 with powder signals stronger than or equal to that of KDP, and 6 with powder signals stronger than or equal to that of LAP. We therefore estimate that about 10% of the materials in this class of

compounds have nonlinearities greater than that of KDP. Table 5-8 lists the 21 high-powder-signal compounds and their measured white-light refractive indexes.

Nonlinearities greater than or equal to that of quartz (0.5 pm/V) are generally necessary if a material is to have a good figure of merit compared with KDP,⁴⁹ so the high-powder-signal criterion allows us to reject about 70% of the compounds examined in this study. A few useful compounds may be wrongly rejected by this criterion if they belong to point group 222.

The powder signals of group-222 compounds tend to be much weaker than those of the group-2 compounds. Figure 5-27 shows the distribution of powder-SHG intensities from known group-2 and -222 compounds. Only 2 of the 26 group-222 compounds have powder signals stronger than or equal to that of quartz; 9 of the 15 group-2 compounds have powder signals this strong. The mean powder signal from the group-222 compounds is an order of magnitude smaller than that of the group-2 compounds.

Some of these compounds may in fact have reasonably high nonlinear coefficients, but have weak powder signals because of a tendency for cleavage perpendicular to principal directions.^{44,45} But even if a group-222 compound has a large nonlinear coefficient, the effective nonlinear coupling must vanish for all noncritical phasematching configurations.^{50,51} Symmetry considerations lead us to expect that a group-222 compound will have a competitive figure of merit only if it is phase-matchable in a low-birefringence plane⁵¹ and if the propagation direction lies about halfway between the two principal directions in that plane. Such special cases can occur, but are probably rare. Thus we feel justified in neglecting low-powder-signal materials in our initial screening. However, this criterion by itself is not sharp enough to select useful materials for further characterization.

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Table 5-8. Organic crystals with high powder-SHG signals. The powder signal relative to quartz is labeled I/I_q ; the principal indexes of refraction were determined for white light.

Compound	I/I_q	n_a	n_β	n_γ
Sodium bitartrate	1.9	1.508	1.522	1.540
Magnesium tartrate	7.6	1.496	1.535	1.539
Diammonium tartrate	6.9	1.528	1.575	1.582
Dithallium tartrate	1.2	1.740	1.845	1.852
Dirubidium tartrate	8.7	1.529	1.541	1.541
L-arginine phosphate	6.0	1.508	1.575	1.583
L-arginine fluoride	5.6	1.500	1.563	1.587
L-arginine chloride	2.4	1.548	1.640	1.642
L-arginine bromide	1.2	1.568	1.645	1.659
L-arginine acetate	1.2	1.534	1.545	1.581
L-valine	6.2	1.509	1.571	1.581
L-cystine hydrochloride	1.5	1.583	1.583	1.618
N-acetyl valine	2.5	1.518	1.520	1.570
N-acetyl proline	2.7	1.479	1.540	1.557
N-acetyl hydroxyproline	4.2	1.474	1.509	1.535
N-acetyl tyrosine	3.1	1.518	1.599	1.65
N-acetyl asparagine	2.2	1.516	1.534	1.58
N-acetyl histidine	6.5	1.486	1.597	1.635
N-acetyl cystine	1.0	1.535	1.606	1.647
Ammonium n-acetyl valine	3.0	1.486	1.522	1.535
Ammonium formate	20.0	1.397	1.500	1.536

Fig. 5-27. Logarithm of observed powder-SHG signal / relative to that of quartz I_q for chiral organic salts with orthorhombic (222) and monoclinic (2) structures. Approximate powder signal of KDP is indicated.

Estimating Noncritical Wavelengths in Organic Salts

The efficiency of frequency conversion of imperfect (non-transform-limited) laser beams depends critically on angular sensitivity as well as on the nonlinear coupling of the harmonic generating crystal.⁴⁹ A great decrease in the angular sensitivity of phasematching can be obtained under so-called noncritical conditions.⁵⁰⁻⁵² In noncritical phasematching (NCPM), the fundamental and harmonic beams propagate parallel to one of the principal refractive index directions. In uniaxial crystals two of the refractive indexes are of course degenerate, so the NCPM condition simply becomes propagation perpendicular to the unique axis, or "90° phasematching" (Ref. 52). In biaxial crystals NCPM can be achieved theoretically at *some* wavelength for propagation along *any* of the three principal axes.^{50,51} Clearly, those nonlinear crystals that are noncritically phasematchable for doubling or tripling light at around 1.06 μm are most desirable for ICF. For the SLC program we wish to identify frequency doublers that are noncritical near 0.92 μm .

The traditional approach to determining NCPM wavelengths is to obtain accurate dispersion measurements by prism spectrometry (using the minimum deviation method,⁵³ for example) and then to calculate the wavelength. This requires large crystals, however. It has recently become possible to measure dispersion curves using spindle-stage refractometry.⁵⁴ The calculated NCPM wavelength is less accurate than that obtainable by prism measurements, but this procedure requires only 50- to 100- μm samples. However, this procedure is too time-consuming to be useful for primary screening.

The most elegant method of determining NCPM is through direct phase-matching measurements using a tunable laser.⁵⁵ This procedure can be carried out on crystals as small as 300 μm but, like spindle-stage refractometry, it is too time-consuming to apply indiscriminately. In addition, obtaining crystals of the size and quality suitable for this measurement usually requires carefully controlled recrystallization.

Table 5-9. Polarization conditions for NCPM in biaxial crystals.

Propagation direction	Type I			Type II			Type III		
	ω_1	ω_2	ω_3	ω_1	ω_2	ω_3	ω_1	ω_2	ω_3
α	γ	γ	β	γ	β	β	β	γ	β
β	γ	γ	α	γ	α	α	α	γ	α
γ	β	β	α	β	α	α	α	β	α

In principle, approximate determinations of noncritical wavelengths for frequency doubling can also be carried out by powder SHG measurements.⁵⁶ This approach might be useful for characterizing uniaxial crystals, because they have at most two NCPM wavelengths (one each for Type I and Type II doubling). However, interpretation of these measurements is too complex to be useful for biaxial crystals, and it is difficult to extend this kind of measurement to third-harmonic generation.

We have therefore developed a new method for screening crystals for NCPM. This method relies on only the simplest optical data obtainable in microcrystalline samples (the three principal white-light refractive indexes), but it can identify NCPM wavelengths in biaxial crystals with an accuracy of about $\pm 0.05 \mu\text{m}$. On the basis of such an educated guess it is possible to sort materials according to their likelihood of exhibiting NCPM at wavelengths of interest to ICF or SLC.

Detailed discussions of NCPM in biaxial crystals are contained in Refs. 50 and 51. For each type (I, II, or III) of frequency-doubling or -tripling, a biaxial crystal has three noncritical wavelengths, one for propagation along each of the three principal axes for the refractive index. We denote these axes as α , β , and γ , with the convention that $n_\gamma > n_\beta > n_\alpha$. Table 5-9 lists the polarization conditions for noncritical mixing in terms of these axes. Type II and III phasematching are identical for frequency doubling ($1\omega + 1\omega \rightarrow 2\omega$), and the phasematching equations for noncritical configurations can be written as shown in Table 5-10. The equations of Table 5-10 are expressed in terms of refractive index differences because those quantities can be estimated directly from the white-light refractive indexes.

The equations for NCPM frequency tripling ($1\omega + 2\omega \rightarrow 3\omega$) are shown in Table 5-11.

Table 5-10. Noncritical phasematching equations for frequency doubling ($1\omega + 1\omega \rightarrow 2\omega$).^a

Propagation axis	Type I	Type II
α	$\Delta_{\eta\eta}^2 = \gamma_{\gamma}^{2,1}$	$\Delta_{\eta\eta}^2 = \delta_{\gamma}^{2,1} + \delta_{\beta}^{2,1}$
β	$\Delta_{\gamma\alpha}^2 = \gamma_{\gamma}^{2,1}$	$\Delta_{\gamma\alpha}^2 = \delta_{\gamma}^{2,1} + \delta_{\alpha}^{2,1}$
γ	$\Delta_{\beta\alpha}^2 = \gamma_{\beta}^{2,1}$	$\Delta_{\beta\alpha}^2 = \delta_{\beta}^{2,1} + \delta_{\alpha}^{2,1}$

^a Here $\Delta_{\gamma\beta}^2 = n_{\gamma}(2\omega) - n_{\beta}(2\omega)$, $\delta_{\gamma}^{2,1} = n_{\gamma}(2\omega) - n_{\gamma}(1\omega)$, etc.

Table 5-11. Noncritical phasematching equations for frequency tripling ($1\omega + 2\omega \rightarrow 3\omega$).^a

Propagation axis	Type I	Type II	Type III
α	$\Delta_{\eta\eta}^3 = \frac{1}{2} [\delta_{\gamma}^{3,2} + \delta_{\gamma}^{3,1}]$	$\Delta_{\eta\eta}^3 = \delta_{\gamma}^{3,2} + \delta_{\beta}^{3,1}$	$\Delta_{\eta\eta}^3 = \delta_{\beta}^{3,2} + \delta_{\gamma}^{3,1}$
β	$\Delta_{\gamma\alpha}^3 = \frac{1}{2} [\delta_{\gamma}^{3,2} + \delta_{\gamma}^{3,1}]$	$\Delta_{\gamma\alpha}^3 = \delta_{\gamma}^{3,2} + \delta_{\alpha}^{3,1}$	$\Delta_{\gamma\alpha}^3 = \delta_{\gamma}^{3,2} + \delta_{\gamma}^{3,1}$
γ	$\Delta_{\beta\alpha}^3 = \frac{1}{2} [\delta_{\beta}^{3,2} + \delta_{\beta}^{3,1}]$	$\Delta_{\beta\alpha}^3 = \delta_{\beta}^{3,2} + \delta_{\alpha}^{3,1}$	$\Delta_{\beta\alpha}^3 = \delta_{\alpha}^{3,2} + \delta_{\beta}^{3,1}$

^a By analogy with the notation used for frequency doubling, $\Delta_{\eta\eta}^3 = n_{\gamma}(3\omega) - n_{\beta}(3\omega)$, $\delta_{\gamma}^{3,2} = n_{\gamma}(2\omega) - n_{\gamma}(3\omega)$, etc.

To solve these equations exactly, of course, the wavelength dependence of the refractive indexes is needed. Our method of estimating noncritical wavelengths from these equations is based on two approximations: First, we assume that the birefringences Δ_{γ}^2 and Δ_{β}^2 can be replaced by the average birefringence at visible wavelengths. Second, we estimate the dispersions $\delta_{\gamma}^{2,1}$ on the basis of the average principal refractive indexes measured at visible wavelengths.

It is generally recognized that there is a strong correlation between dispersion and average refractive index. To estimate the dispersions we derived interpolating functions using known refractive index data for nine organic salts. For a series of wavelengths we plotted $n_{\gamma}(2\omega) - n_{\gamma}(1\omega)$ vs $n_{\gamma}^2(0.532) - 1$, as shown in Fig. 5-28. The data for each wavelength were fitted to a straight line:

$$\delta^{2,1} = A(\lambda) + B(\lambda)(n_{\gamma}^2 - 1). \quad (32)$$

The parameters $A(\lambda)$ and $B(\lambda)$ were then fitted to appropriate interpolation functions so the dispersion $n(2\omega) - n(1\omega)$ could be calculated for any fundamental (1ω) wavelength. The fitting functions

$$A(\lambda) = P + Q/\lambda^4, \quad (33)$$

$$B(\lambda) = R + S/\lambda^{7/2} \quad (34)$$

fit the data quite well between 1.06 and 0.532 μm , although they do not have the correct limiting behavior for long wavelengths. By contrast, the Sellmeier form

$$A(\lambda) = \frac{T}{U - \lambda}, \quad (35)$$

$$B(\lambda) = \frac{V}{W - \lambda}, \quad (36)$$

does not fit as well, but is more physical at long wavelengths.

For frequency doubling, we simply set the left-hand sides of the equations of Table 5-10 equal to the measured principal white-light birefringence, replaced the right-hand sides by our empirical formulas for $\delta^{2,1}$, and solved for the wavelength using Newton's method. The

Fig. 5-28. Dispersion $\delta^{2,1}$ vs $n^2(0.532) - 1$ for $1\omega = 0.532$, 0.700, and 1.06 μm . Solid lines are best linear fits.

Sellmeier form [Eqs. (35) and (36)] gave more reasonable values in the IR than Eqs. (33) and (34), but sometimes gave meaningless values when the noncritical wavelength was shorter than $0.5 \mu\text{m}$. Equations (33) and (34), on the other hand, gave reasonable values at short wavelengths, but sometimes failed to converge in the IR. Both pairs of equations gave similar values between 0.5 and $1.1 \mu\text{m}$.

For frequency tripling, we plotted $\delta^{3,1}$ and $\delta^{3,2}$ vs $n^2(0.532) - 1$ for our nine known crystals and performed a linear fit. For our unknowns we then simply calculated the left-hand sides of the equations of Table 5-11 for mixing $1.06\text{-}\mu\text{m}$ and $0.532\text{-}\mu\text{m}$ light and compared the resulting value with the white-light birefringence for the appropriate principal plane. If the two values differed by less than 10% that process was considered near NCPM.

To determine the accuracy of our noncritical wavelength estimates, we compared them with calculated results for five materials for which sufficiently accurate refractive index data are available. Table 5-12 shows that our predicted NCPM wavelengths for doubling agree with the calculated wavelengths to within $0.05 \mu\text{m}$. This accuracy is certainly sufficient to allow us to select materials for further characterization.

To test our ability to identify potential noncritical triplers, we calculated the NCPM wavelengths for mixing λ and $\lambda/2$ in the same five compounds. Table 5-13 shows that each time we have identified a principal axis as being noncritical for some type of $1.06\text{-}\mu\text{m}$ tripling, the calculated critical wavelength is indeed close to $1.06 \mu\text{m}$.

Finally, a direct measurement shows that the NCPM wavelength for Type I doubling in the a direction of L-arginine fluoride (LAF) is $0.96 \mu\text{m}$. This compares favorably with our estimates of 0.93 and $0.91 \mu\text{m}$ for that process.

Statistical Distribution of Noncritical Wavelengths in Organic Salts. From our estimates of noncritical wavelengths in the organic salts we can establish the probability of finding materials with NCPM at a given wavelength. Figure 5-29 shows the empirical distribution of NCPM wavelengths for Type I and Type II

Table 5-12. Estimated and calculated noncritical phasematching wavelengths (in μm) for frequency doubling.

Crystal	Axis	Type I		Type II	
		Est.	Calc.	Est.	Calc.
L-arginine phosphate	a	— ^a	—	—	—
	β	0.538	0.478	0.696	0.637
	γ	0.559	0.502	0.729	0.674
L-arginine fluoride	a	0.910	0.926	—	—
	β	0.508	0.443	0.649	0.611
	γ	0.566	0.516	0.738	0.725
Diammonium tartrate	a	—	—	—	—
	β	0.621	0.556	0.822	0.780
	γ	0.655	0.576	0.873	0.845
N-acetyl L-arginine	a	0.676	0.657	0.904	0.944
	β	0.575	0.596	0.752	0.773
	γ	0.953	0.991	—	—
N-acetyl L-valine	a	—	—	—	—
	β	0.632	0.575	0.840	0.823
	γ	0.621	0.543	0.823	0.808

^a Blanks represent configurations for which the noncritical wavelength is longer than $1.2 \mu\text{m}$.

Table 5-13. Estimated and calculated noncritical phasematching wavelengths (in μm) for frequency tripling by mixing.

Crystal	Axis	Type	Est. (μm)	Calc. (μm)
L-arginine phosphate	γ	II	< 1.0	0.831
	β	III	> 1.0	1.13
L-arginine fluoride	γ	II	< 1.0	0.830
	β	III	> 1.0	1.09
Diammonium tartrate	γ	I	< 1.0	0.983
N-acetyl arginine	a	I	> 1.0	0.923
	β	II	< 1.0	0.977
N-acetyl valine	a	II	< 1.0	0.968
	β	II	< 1.0	0.924

frequency doubling. The plot includes all three principal axes. Both histograms are skewed toward short wavelength; this reflects the tendency for these compounds to have high birefringence and low dispersion, and suggests that we might increase the probability of finding NCPM at longer or shorter wavelengths by chemical modifications that increase or decrease the dispersion, respectively. This approach to engineering new materials has not been systematically applied.

New Noncritical Harmonic Generators for ICF and SLC. The aim of this study was to develop a way to identify the most angularly insensitive materials for frequency conversion processes of interest to laser program objectives. From 21 high-powder-signal materials (Table 5-9) we have produced an ordered list of 17 materials with possible advantages over KDP for one or more frequency conversion processes of interest to ICF. In addition, we have selected 10 materials that

are most likely to be useful for efficient doubling of light around 0.9 μm (SLC wavelength).

Table 5-14 lists the materials selected for further characterization for ICF. We indicate the type of frequency conversion process for which each material is of interest. The materials are in a rough order

of priority based on the number of processes for which they are potentially NCPM and on the assumption that triplers were more important than doublers. These priorities are by no means rigid, but serve as a guide for further work.

Table 5-15 lists materials identified as possible noncritical doublers for SLC wavelengths. The order is based on how close the predicted NCPM wavelength was to 0.92 μm .

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Fig. 5-29. Distribution of noncritical wavelengths among the chiral organic salts studied.

Table 5-14. New materials for ICF frequency conversion: column heads give target frequency and conversion type; body entries give axes and NCPM wavelength estimates in μm .

Crystal	2ω (I)	2ω (II)	3ω (I)	3ω (II)	3ω (III)
N-acetyl hydroxyproline	α (1.03)	α (1.06)	γ	β	—
N-acetyl histidine	—	α (1.06)	—	γ	β
Dithallium tartrate	—	—	—	β	—
N-acetyl tyrosine	—	—	α	γ	β
L-arginine bromide	—	—	—	γ	β
L-arginine chloride	—	—	—	γ	β
N-acetyl cystine	—	α (1.04)	α	γ	—
N-acetyl asparagine	γ (0.95)	—	α	β	—
Sodium bitartrate	γ (1.07)	—	—	α	β
N-acetyl proline	α (1.03)	—	—	γ	β
Valine	—	—	—	γ	β
Magnesium tartrate	—	—	—	β, γ	—
Diammonium tartrate	—	—	β, γ	—	—
L-arginine acetate	γ (1.10)	α (0.98)	α, β	—	—
Ammonium formate	—	—	α	—	—
Ammonium n-acetyl valine	—	—	γ	—	—
Cystine hydrochloride	—	α (1.02)	—	—	—

Dispersion Measurements Using the Spindle Stage Refractometer

To assess the frequency conversion characteristics of nonlinear crystals we require accurate values of refractive indexes at the input and output wavelengths. We have therefore developed a spindle stage refractometer to determine linear optical properties of single crystal fragments as small as 50 μm . The technique is a modification of the spindle stage microscopy described in Ref. 57 and permits more accurate measurements over a greater wavelength range.

The new spindle stage refractometer measures refractive indexes at wavelengths from 0.38 to 1.10 μm with an accuracy sufficient to predict phase-matching properties and noncritical wavelengths. The basic procedures for orienting a crystal and for identifying a refractive index match by oil immersion are identical with those for the commercial spindle stage. However, our procedure for determining the refractive index differs from the usual technique.

Spindle Stage Methods. Spindle stage studies utilize a crystal fragment glued to a rotation axis that is perpendicular to the rotation axis of a polarizing microscope. Conoscopic viewing⁵⁸ of the crystal in index matching fluid is used to locate and identify the principal directions of the optical indicatrix. Oil immersions can then be performed on the oriented sample, and several criteria

(Becke line, oblique illumination, and relief contrast⁵⁸) can be used to determine a refractive index match between the oil and the crystal. For precise measurements in monochromatic light, a thermally controlled oil cell may be used to provide continuous variation of the refractive index of the fluid. In routine spindle stage methods, the refractive index of the oil is then calculated from its known optical constants.

The accuracy of the refractive indexes determined by the standard oil immersion method are limited by two main sources of error:

1. The extent to which the actual conditions of match are known, i.e., the precise temperature of the oil/crystal interface and the wavelength of the light.

2. The possibility of contamination of the matching fluid and resulting uncertainty in the optical constants. The refractive index of a fluid is sensitively affected by the loss of volatile components, contamination, and aging.⁵⁹

Thus, the temperature and wavelength derivatives of the refractive index must be accurately known, or the accuracy of the refractive index determined by comparative methods may be severely compromised.

The optical properties of a solid are considerably more stable than those of a liquid, and a number of authors have proposed using a reference crystal to calibrate the oil's refractive index *in situ*.

Our spindle stage refractometer incorporates a well-characterized crystal mounted near the unknown. This allows the true index of the liquid in the oil cell to be determined at the time of match.

The Spindle Stage Refractometer.

Figure 5-30(a) is a schematic drawing of our spindle stage microscope configured for this type of refractometry. Figure 5-30(b) shows how a uniaxial crystal on a spindle axis serves as a microrefractometer to obtain the refractive index of an oil at a particular wavelength. The reference crystal is precisely mounted with its unique (optic) axis perpendicular to the spindle axis and aligned in a plane parallel to the microscope's polarizer. As the reference crystal is rotated on the

Table 5-15. New frequency doubling crystals with NCPM wavelengths near 0.92 μm .

Compound	Type	Axis	Wavelength (μm)
Ammonium n-acetyl valine	II	γ	0.916
L-arginine fluoride	I	α	0.926
N-acetyl hydroxyproline	II	γ	0.909
	I	α	0.907
Magnesium tartrate	II	γ	0.905
	II	β	0.876
N-acetyl tyrosine	II	α	0.896
Ammonium formate	II	α	0.939
L-arginine acetate	II	β	0.893
Diammonium tartrate	II	γ	0.887
	II	β	0.845
N-acetyl asparagine	II	α	0.874
	I	γ	0.954
N-acetyl valine	II	α	0.859
	II	β	0.845

Fig. 5-30. (a) Spindle stage refractometer. (b) Uniaxial reference crystal (represented by its optical indicatrix) illustrates how a continuous change in refractive index from n_e to n_o is observable by rotating the spindle in polarized light.

spindle, the refractive index changes continuously between n_e and n_o , its principal indexes of refraction at the wavelength of the light being used. Once an oil is found that matches the refractive index of the unknown crystal at a wavelength of interest, the reference crystal is rotated by some angle θ (measured between the direction of index match and the optic axis of the reference crystal) until its index $n(\theta)$ matches that of the oil. The common refractive index of the reference crystal, the oil, and the unknown crystal is then determined from the equation

$$n(\theta)^2 = \frac{n_e^2 n_o^2}{n_e^2 \sin^2 \theta + n_o^2 \cos^2 \theta} \tag{37}$$

Fig. 5-31. Dispersion of d-LAP. Curves represent the best fit of spindle stage data to Eq. (38).

Table 5-16. Refractive indexes for d-LAP determined by spindle stage refractometry (ss) and prism spectrometry (ps) for 1.064 and 0.532 μm.

	n_α		n_β		n_γ	
	1.064 μm	0.532 μm	1.064 μm	0.532 μm	1.064 μm	0.532 μm
n_{ss}	1.4945	1.5090	1.5579	1.5763	1.5656	1.5847
n_{ps}	1.4940	1.5092	1.5577	1.5759	1.5655	1.5847

Table 5-17. Refractive indexes at 1.0642 and 0.5321 μm for several organic crystals measured with a spindle stage refractometer.

Compound	n_α		n_β		n_γ	
	1.0642 μm	0.5321 μm	1.0642 μm	0.5321 μm	1.0642 μm	0.5321 μm
L-arginine fluoride	1.4889	1.5054	1.5494	1.5701	1.5744	1.5961
N-acetyl l-arginine	1.5147	1.5332	1.5349	1.5532	1.5777	1.6000
N-acetyl l-valine salt	1.5085	1.5237	1.5101	1.5246	1.5566	1.5753
N-acetyl l-valine	1.4760	1.4943	1.5136	1.5320	1.5222	1.5401
N-acetyl hydroxyproline	1.4636	1.4772	1.4983	1.5129	1.5184	1.5379
N-acetyl tyrosine	1.5039	1.5227	1.5810	1.6058	1.6366	1.6671
N-acetyl asparagine	1.5044	1.5185	1.5227	1.5406	1.5661	1.5908
L-arginine chloride monohydrate	1.5378	1.5556	1.6072	1.6332	1.6308	1.6541

This procedure may be repeated for additional wavelengths to determine the dispersion curve. Our spindle stage refractometer incorporates a CCD camera, coupled to one port of the microscope, to display the reference and unknown crystals on a television monitor. This makes it possible to make visual index matches even in the UV and IR, and enhances the images at all wavelengths. Bandpass filters with 10-nm passbands are used with a white light source to make measurements between 0.38 and 1.10 μm.

Accuracy of Measurement. To assess the accuracy of our apparatus over the full range of wavelengths, we made measurements on a number of materials for which refractive indexes were known. To interpolate our data to other wavelengths, we fitted those data to the Sellmeier equation,

$$n^2 = A + \frac{B}{\lambda^2 + C} + D\lambda^2 \tag{38}$$

Figure 5-31 shows our index data for deuterated l-arginine phosphate (d-LAP), a biaxial second-harmonic generator of current interest, and the dispersion curves obtained from the fits to Eq. (38). Table 5-16 shows our interpolated values at the Nd:YAG 1ω and 2ω wavelengths and values obtained by prism spectrometry⁶⁰; agreement is good.

Using such comparisons, we have determined a standard deviation of 5×10^{-4} for refractive indexes over the full range of wavelengths.

Dispersion Measurements in Organic Crystals. As described in "New Organic Materials for Harmonic Generation," earlier in this section, several organic crystals have been identified as possible candidates for frequency-doubling and -tripling of 1.06- μm light. We characterized eight of these materials on the spin-dle stage refractometer and fitted the data to Eq. (38). Table 5-17 lists the calculated refractive indexes for these compounds at the Nd:YAG 1 ω and 2 ω wavelengths.

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Phasematching Measurements in Biaxial Crystals

We have studied phasematched second harmonic generation in several new biaxial organic crystals using the nonlinear optical goniometer developed in our laboratory in 1986 (Ref. 61). With this technique we can determine the position of phasematching loci, the variation of the nonlinear coupling, and the angular sensitivity of doubling or mixing in very small crystals.

We studied five salts of L-arginine: the phosphate (LAP), fluoride (LAF), chloride (LACl), acetate (LAAC), and diarsenate (LADA). In addition, we studied diammonium tartrate (DAT) because it had a high powder-SHG signal and because we could grow large DAT crystals rapidly from solution.

Figure 5-32 shows the phasematching loci for Type I and Type II doubling of 1.06- μm light (the fundamental wavelength of Nd:YAG), and the positions of maximum and zero nonlinear coupling for these compounds. The angular positions for phasematching are represented by points on a sphere whose reference axes are the directions of the three principal refractive indexes.⁶² The equator of each diagram represents the optic plane of the crystal. The positions of the optic axes are marked by asterisks. The polar axis is the intermediate-index direction, or optic normal.

Table 5-18. Nonlinear coupling and angular sensitivity for selected phase-matching positions in some nonlinear crystals.

Material	Type/position	d_{eff} (pm/V) ^a	Angular sensitivity (cm ⁻¹ /rad) ^a
L-arginine phosphate (LAP)	I/A	1.6	6900
	II/C	1.5	4000
L-arginine fluoride (LAF)	I/A	2.0	3900
	II/B	1.5	4300
L-arginine acetate (LAAC)	I/C	0.35	4300
	II/A	0.30	540
L-arginine diarsenate (LADA)	I/A	0.13	5500
	II/A	0.19	1800
L-arginine chloride (LACl)	I/A	—	3100
	II/A	—	1600
Diammonium tartrate (DAT)	I/C	0.89	6000
	II/C	0.59	2500
Potassium dihydrogen phosphate (KDP)	I	0.41 ^b	4900
	II	0.56	2500

^a $\pm 20\%$.

^b S. Kurtz, J. Jerphagnon, and M. Choy, Landolt-Bornstein New Series, vol. 11 (Springer, New York, N.Y., 1978), p. 671.

Table 5-18 lists the nonlinear couplings and angular sensitivities for selected positions on each locus.

L-Arginine Phosphate (LAP). LAP is being considered as an alternative to KDP for future high power laser systems.⁶³ We studied the variation of nonlinear coupling in LAP along the loci for Type I and Type II frequency doubling of 1.06- μm light. We used this information to deduce the relative signs of the d coefficients whose magnitudes we had measured earlier using the wedge technique.⁶⁴

L-Arginine Fluoride (LAF). The powder-SHG signal of LAF is nearly equal to that of LAP.⁶⁵ This implies that the magnitudes of the nonlinear coupling coefficients are similar. Our phasematching studies confirmed this (see Table 5-18), but also revealed significant differences in the topology of certain phasematching processes.⁶⁶ These differences arise from differences in the linear optical properties of LAF and LAP. For example, in LAF the loci of Type I phasematching positions for doubling 1.06- μm light form two isolated cones around the optic axes [Fig. 5-32(b)]. Figure 5-33 shows the results of a study⁶⁶ of the evolution of these loci with decreasing wavelength. As the wavelength decreases, the loci expand until they merge at the α axis. This is a position of NCPM for that wavelength, and

Fig. 5-32. Phase-matching loci for Type I and Type II frequency doubling of $1.06\text{-}\mu\text{m}$ light in five salts of l-arginine and in diammonium tartrate. The letters A, B, C, and D mark positions where d_{eff} is strong. O's mark the positions where $d_{\text{eff}} = 0$. Asterisks mark the optic axes.

hence is very insensitive to angular deviations.⁶⁷ A further decrease of the wavelength causes the locus to pull away from the α axis in the α - β plane, so that it becomes a closed loop containing both the α and optic axes.

Our lasers could not generate light with wavelengths between 1.06 and $0.92\ \mu\text{m}$, so to determine the wavelength at which the loci just touch at α , we plotted the angle θ at which the loci cut the α - β plane as a function of wavelength, and extrapolated to $\theta = 0$. This procedure, illustrated in Fig. 5-34, gives a value of $0.956\ \mu\text{m}$ for the noncritical wavelength. Therefore, between 1.0 and $0.9\ \mu\text{m}$, LAF has positions of relatively angularly insensitive Type I phasematching that LAP does not have.

Figure 5-35 shows the loci for Type I and Type II mixing of 1.06- and $0.532\text{-}\mu\text{m}$ light (3ω) and the loci for Type I doubling of $0.532\text{-}\mu\text{m}$ light (4ω).

L-Arginine Acetate (LAAC). L-arginine acetate is another salt from the l-arginine family that belongs to point group 2 (Ref. 65). The powder signal of this compound is only slightly stronger than that of quartz, which implies that its effective nonlinear coupling coefficients are somewhat smaller than those of KDP. Since LAAC has a much lower birefringence than LAP or LAF, we investigated LAAC to check for NCPM near $1.06\ \mu\text{m}$. We found that Type II doubling was very nearly noncritical for propagation along the γ axis. Unfortunately, the coupling vanishes in this direction. This is consistent with the crystallographic 2-fold axis being parallel to the γ direction.

L-Arginine Diarsenate (LADA).

L-arginine diarsenate crystallizes in point group 222, and has a very small powder signal (one-tenth that of quartz).⁶⁵ We investigated phasematched harmonic generation in LADA and found the value $d_{14} = 0.4 \pm 0.1\ \text{pm/V}$. This value is only slightly smaller than the value of d_{11} in quartz ($0.5\ \text{pm/V}$), and implies that the weak powder-SHG signal of LADA is at least partly due to crystal morphology.⁶⁴

L-Arginine Chloride (LACl). This crystal also belongs to the monoclinic class 2, and has a powder signal similar to that of LAAC.⁶⁵ We were able to determine the phasematching loci, but the

Fig. 5-33. Positions of Type I doubling loci in LAF for various wavelengths.

Fig. 5-34. Variation with wavelength of the angle at which the Type I doubling loci of LAF intersect the α - β plane. The point of crossing ($0.956\ \mu\text{m}$) represents noncritical phasematching.

sample quality was too poor to allow a satisfactory estimate of the nonlinear coupling. Nonetheless, the low birefringence of this crystal leads to positions of very low angular sensitivity for both Type I and Type II doubling of $1.06\text{-}\mu\text{m}$ light. We will continue our investigation of LaCl when better-quality material is available.

Diammonium Tartrate (DAT). Diammonium tartrate has a strong powder-SHG signal (7 times that of quartz),⁶⁹ and

Fig. 5-35. Loci for Type I and Type II mixing of 1.064- and 0.532- μm light (3ω) and Type I doubling of 0.532- μm light (4ω) in LAF.

can be grown quickly into large, high quality crystals. Previous investigators reported phasematching in principal planes, where the nonlinear coupling was found to be quite weak.⁷⁰ By contrast, we discovered positions on the Type I locus where d_{eff} was nearly twice that in KDP (Table 5-18).⁶² However, neither the nonlinear coupling nor the angular sensitivity is superior to that of KDP for Type II doubling of 1.06- μm light.

Summary. Of the six materials studied, only LAP and DAT are available in sizes large enough to permit the use of traditional wedge and prism measurements⁷¹ to characterize their properties. Using only millimeter-sized samples, our direct phasematching measurements using the nonlinear optical goniometer have allowed us to identify LAF and DAT as potentially useful new materials for second harmonic generation. Another l-arginine salt, LACI, has angularly insensitive positions for frequency doubling of 1.06- μm light. Two crystals, LAAC and LADA, have nonlinear couplings weaker than those of KDP. These results demonstrate that the nonlinear optical goniometer provides a powerful tool for

characterizing single crystals of new nonlinear materials.

Author: S. P. Velsko

Advanced Crystalline Laser Materials

Introduction

The main objective of our research and development program in advanced solid state laser materials is to identify and characterize new materials with the properties required for use as active laser media in a multimegajoule research laser or an ICF power-plant reactor laser. Materials for both applications must satisfy stringent performance- and cost-related requirements not satisfied by any material now available. Our search for suitable new materials is concentrated on crystalline media rather than glasses, because their thermomechanical properties are superior to those of glasses and because they offer a broader range and uniformity of ion-site sizes and coordinations for the incorporation of optically active impurity ions.

The technical requirements for ICF lasers can be met, in principle, by either flashlamp-pumped or laser-pumped gain media. With flashlamp pumping, the required overall laser efficiency of greater than 10% might be achieved by codoping the laser host with two ions—a sensitizer, such as Cr^{3+} , and an activator, such as Nd^{3+} . The first two articles in this subsection ("Oxide Laser Materials" and "Halide Laser Materials") review progress in our search for such media.

It has proven difficult to find a host for flashlamp pumping that satisfies all the criteria for high-peak-power laser applications. The use of a laser-pumped gain medium simplifies the materials problem by dividing it into two. One subset of the requirements must be met by the pump laser medium and the other by the energy-storage amplifier medium.

Semiconductor laser diodes, which have electrical efficiencies greater than

50%, will eventually be a superior technical choice for a pump source, but they are prohibitively expensive for large-scale applications at present. We therefore gave much attention during 1987 to searching for flashlamp-excited pump lasers and corresponding amplifier media for a multimegajoule ICF research laser.

The requirements for the energy-storage medium can be met by rare-earth laser ions such as Nd^{3+} , Tm^{3+} , and Yb^{3+} , which in several easily grown crystalline hosts have long excited-state lifetimes and near-IR optical transitions with optimum emission cross sections. We considered a number of possibilities for pump lasers for these ions, including transition-metal laser ions in crystals, glasses, and liquids, and color centers in crystals. We concentrated most of our effort on tunable, vibronic-sideband Cr^{3+} lasers, which offer several advantages, including the possibility of high efficiency with flashlamp pumping because of the strong, broad absorption bands of Cr^{3+} .

To produce a suitably efficient, tunable Cr^{3+} laser, it is necessary to minimize the excited state absorption (ESA) from the upper ${}^4\text{T}_2$ laser level. Using criteria developed in the Office of Basic Energy Sciences-supported research on ESA, described in "Effect of Excited-State Absorption on the Performance of Cr^{3+} Vibronic Lasers," later in this section, we and our colleagues at Allied-Signal Research Laboratories surveyed many possible oxide and fluoride crystal hosts for Cr^{3+} . This work is reviewed in the next two articles.

The most promising host material we have discovered is LiCaAlF_6 (LiCAF), whose slope efficiency with Kr laser pumping is comparable with that of alexandrite, the best available Cr^{3+} laser. The spectroscopic and laser performance of LiCAF are summarized in "LiCAF ($\text{LiCaAlF}_6:\text{Cr}^{3+}$): A Promising New Solid State Laser Material." Its efficiency and tuning range and its low melting point, inexpensive chemical constituents, and other useful properties suggest that LiCAF has the potential to be a low-cost,

efficient pump laser for an ICF research laser.

Author: L. L. Chase

Oxide Laser Materials

Investigations of oxide materials proceeded along two paths during 1987. One path is the search for laser media that use Cr^{3+} sensitization of Nd^{3+} to raise flashlamp pumping efficiency.^{72,73} This year we concentrated on a particular class of candidates, the metaphosphates. (The other path, a laser-pumped laser, is discussed later in this article.)

In 1986 we reported⁷⁴ on the optical properties of Cr^{3+} in $\text{Y}(\text{PO}_3)_3$, which was then considered the best-growing rodopable metaphosphate. The emission from this material peaks near 885 nm, which does not provide a good overlap to the absorption bands of Nd^{3+} . Such an overlap is required to produce rapid energy transfer from Cr^{3+} to Nd^{3+} . The fluorescence lifetime and emission intensity increase strongly as the temperature is lowered, indicating a low quantum efficiency resulting from multiphonon emission at room temperature. The low quantum efficiency and poor overlap both contribute to the low observed sensitization efficiency.

In 1987 we found similarly low transfer efficiencies (about 30%) for $\text{Lu}(\text{PO}_3)_3:\text{Cr},\text{Nd}$. This low efficiency was due, in part, to a poor overlap of the Cr^{3+} emission with the Nd^{3+} absorption and to a low Cr^{3+} quantum efficiency at room temperature; it was also due to a lower than optimal concentration of Nd^{3+} in our samples. We believe that the thermal quenching of the Cr^{3+} fluorescence results from a large degree of lattice relaxation during emission, caused by the large mismatch of the ionic radii of Cr^{3+} and Y^{3+} . To circumvent this problem, we must find hosts other than the simple metaphosphates.

One suitable type of host would be one with separate cation sites for Cr^{3+} and Nd^{3+} . There are four cation sites in the metaphosphate lattice; we proposed formulations with one (or two) of these sites

Fig. 5-36. Emission spectrum of $\text{Y}_2\text{GdAl}(\text{PO}_3)_{12}:\text{Cr}$.

Table 5-19. Spectroscopic parameters of Cr^{3+} -doped oxides.

Host	Peak emission wavelength (nm)	First e^{-1} time (ms)
CaYAlO_4	744	9
CaLaAlO_4	748	2.4 (1.35 at 800 nm)
SrLaAlO_4	745	0.45 (0.13 at 800 nm)
CaLaGaO_4	830	0.075
LiGa_5O_8	719	1.75 (0.21 at 830 nm)
$\text{La}_3\text{Ga}_5\text{SiO}_{14}$	882	0.006
$\text{LiAlSi}_2\text{O}_6$	755	0.03
ScAlBeO_4	720, 775 ^a	0.03 at 785 nm 12 at 696 nm

^a Depends on concentration.

occupied by Al^{3+} and the other three (or two) sites occupied by lanthanide-type (Gd or Y) ions. If an ordered compound were formed, Cr^{3+} ions would substitute for Al^{3+} ions and Nd^{3+} ions for the lanthanide ions, with each impurity being in a site of well-defined symmetry.

Samples of $\text{GdAl}(\text{PO}_3)_6:\text{Cr}, \text{Nd}$ and $\text{Y}_2\text{GdAl}(\text{PO}_3)_{12}$, both Cr-doped and Cr,Nd-doped, were grown under contract with Allied-Signal's Corporate Technology Center in Morristown, N.J. The crystals were grown in platinum crucibles that were RF-induction heated to the melting point of the compound and then cooled slowly. The $\text{GdAl}(\text{PO}_3)_6$ showed a strong tendency to form a glass. Since room-temperature spectroscopic measurements of both codoped compounds produced very similar results, we discuss here only the measurements on $\text{Y}_2\text{GdAl}(\text{PO}_3)_{12}$.

Figure 5-36 shows the emission of Cr^{3+} in $\text{Y}_2\text{GdAl}(\text{PO}_3)_{12}$. The emission peaks at 900 nm, a much longer wavelength than anticipated for Cr^{3+} in an Al^{3+} site. The fluorescence decay was nonexponential at all temperatures studied (15 to 300 K).

These data suggest that the compound is disordered, with the Cr^{3+} ions residing in a "loose" site. This would account for the low cubic crystal field splitting indicated by the wavelength of the emission.

Such a site will allow considerable lattice relaxation, with correspondingly high thermal quenching, which we confirmed. The fluorescence decay time (e^{-1}) was 24 μs at 15 K and 10 μs at 298 K. A similar decrease in the integrated fluorescence intensity was also found. Thus, this material would be expected to exhibit an energy transfer efficiency no higher than those of the simple metaphosphates. We confirmed this expectation in a codoped sample containing 0.5 at.% Cr^{3+} and 2.5 at.% Nd^{3+} . The Nd^{3+} emission was inhomogeneously broadened, with a nonexponential fluorescence decay (first e^{-1} time = 175 μs), further indicating the disordered nature of this compound. A comparison of the Nd^{3+} excitation spectrum with the absorption spectrum indicated a transfer efficiency of about 20%. While an ordered compound may yet be found among the mixed-cation metaphosphates, more effort at producing crystalline material with some other ratio of Al^{3+} and a lanthanide ion will be needed.

As described in the introduction to this subsection, the second research path taken in 1987 was a new fusion-driver concept in which a free-running laser pumps the driver gain medium. For drivers using Nd^{3+} as the active ion, a pump laser emitting near 730 to 740 nm or 790 to 810 nm would be ideal, because it would effectively pump the Nd absorption band.

Lasers based on the Cr^{3+} ion are known to work in this entire wavelength region. Only alexandrite is known to have a moderate laser efficiency (about 2.5%) in a flashlamp-pumped mode.⁷⁵ A recent investigation of ESA⁷⁶ concluded that efficient Cr^{3+} lasers should result when Cr^{3+} ions are in "tight" sites; that is, when the ionic radius of Cr^{3+} is the same as, or greater than, the ionic radius of the host cation. For oxide hosts, this suggests that we restrict our search to hosts containing six-fold coordinated Al^{3+} or Ga^{3+} ions. The materials selected

for investigation were grown at Allied-Signal as described above; Table 5-19 gives some spectroscopic properties of these materials.

Compounds based on the formula $(\text{AE})(\text{RE})(\text{M})\text{O}_4$, where AE is an alkaline earth ion (Ca, Ba, or Sr), RE is a rare earth ion (Y or La), and M is a trivalent metal ion (Al or Ga), have tetragonal unit cells. The metal ion is six-fold coordinated; the nearest-neighbor cations comprise a statistical distribution of AE and RE ions.⁷⁷ This results in inhomogeneously broadened emission in rare-earth-doped crystals, which occurs (for the same reason) in Cr^{3+} -doped crystals as well. For the first four materials in Table 5-19, the fluorescence lifetime is a strong function of the wavelength of the emission; the shorter lifetimes correspond to Cr^{3+} ions in lower field sites, which emit at longer wavelengths. This lowers the lasing efficiency of these compounds, since much of the absorbed pump light would not be accessible to the lasing ions. Also, in most of the cases studied, the decay times in the desired wavelength region were too long, resulting in low emission cross sections. We are therefore pursuing other classes of host materials.

Neodymium-doped $\text{La}_2\text{Ga}_5\text{SiO}_{14}$ has been used as a laser material.⁷⁸ The Ga^{3+} ions are in both four- and six-fold coordination; the tetrahedral Si^{4+} and Ga^{3+} ions are arranged in planes above and below planes containing the La^{3+} and octahedral Ga^{3+} ions. This is an open structure that permits a considerable degree of lattice relaxation at the Ga^{3+} site. The peak of the Cr^{3+} $^4\text{T}_2$ absorption is at 645 nm, close to the peak absorption wavelength seen in other low-field hosts such as GGG (Ref. 78); the peak of the fluorescence is at 882 nm, however, which represents a much larger shift than usual. There is considerable thermal quenching of the integrated intensity and the fluorescence decay as the temperature is raised to room temperature. Measurements of the room-temperature quantum efficiency (relative to alexandrite and $\text{GSGG}:\text{Cr}$) at Allied-Signal yielded a value of 30%, in excellent agreement with our determination of the

change in integrated intensity with temperature. Even with such a low quantum efficiency, this material was lased at Allied-Signal, using quasi-cw laser pumping. The slope efficiency was relatively low (7.6%, with a 1.1% output coupler), and the tuning range of 885 to 1107 nm was anomalously truncated at wavelengths shorter than the predicted peak of the gain.⁷⁹ This suggests that there are loss mechanisms, such as ESA, in this region. A similar tuning range was obtained by Kaminskii et al. using ruby laser pumping.⁸⁰ We are evaluating this material further to assess its ultimate efficiency and its performance relative to other candidates.

The gallium compound $\text{LiGa}_5\text{O}_8:\text{Cr}^{3+}$ also has octahedral and tetrahedral Ga^{3+} sites. Work on the spectroscopic properties of Cr^{3+} in this host has yielded the crystal field parameters,^{81,82} but only low-temperature emission spectra have been reported.^{81,83} Figure 5-37 shows the room-temperature fluorescence spectrum. The line emission at 719 nm corresponds to the $^2\text{E} \rightarrow ^4\text{A}_2$ transition.^{81,83} The fluorescence lifetime for this emission is long, 1.75 ms. However, it can be seen that there is a sideband that extends over a rather large wavelength range. The fluorescence decay measured at 830 nm was initially nonexponential, with an exponential component having a lifetime of 1.54 ms at longer times. The initial e^{-1} time was 210 μs . The long-wavelength tail arises from disorder in the nearest-neighbor cation site occupancy (Li^+ or Ga^{3+}). In any case only a

Fig. 5-37. Emission spectrum of $\text{LiGa}_5\text{O}_8:\text{Cr}$.

fraction of the excited ions are capable of lasing, making this system unattractive as a pump laser candidate.

The two remaining crystals in Table 5-19, both of which contain Al^{3+} , have peak emissions in the wavelength region of interest. The naturally occurring compound $\text{LiAlSi}_2\text{O}_6:\text{Cr}$ (hiddenite) is a spodumene related to diopside, which was discussed in the 1986 Laser Annual.⁷³ The Al^{3+} site is six-fold coordinated. The hiddenite phase does not melt congruently, but it may be possible to grow this phase with the hydrothermal method. Figure 5-38 shows the emission spectrum of this mineral, given to us by B. H. T. Chai of Allied-Signal. Although the position of the spectrum is characteristic of an intermediate crystal field host, the fluorescence lifetime is quite short, about 35 μs . We believe that the emission is radiative and that the high radiative rate is due to the distortion of the Al^{3+} site. This is the case for diopside, which has

Fig. 5-38. Emission spectrum of $\text{LiAlSi}_2\text{O}_6:\text{Cr}$.

Fig. 5-39. Emission spectrum of $\text{ScAlBeO}_4:\text{Cr}$ for 390-nm excitation. ▼

a Cr^{3+} fluorescence lifetime of 48 μs (Ref. 73). This short lifetime implies a high emission cross section, which should allow flashlamp-pumped, long-pulse laser operation. Growing this material may be difficult.

The compound ScAlBeO_4 is structurally related to alexandrite, with half the Al^{3+} ions replaced with Sc^{3+} ions. It was hoped that the Cr^{3+} dopant would substitute for the Sc^{3+} ions, which, because of their greater size, would be at the mirror site position. The resulting material was pink, and the absorption spectrum unambiguously showed that the Cr^{3+} ions substituted for both the Al^{3+} and Sc^{3+} ions. The emission spectrum, shown in Fig. 5-39 for 390-nm excitation, also indicates a multisite emitter. Selective excitation showed that the Al^{3+} -site Cr^{3+} ions were high-field emitters, with sharp line emissions and vibronic sidebands arising from the ${}^2\text{E} \rightarrow {}^4\text{A}_2$ transition and a long (12 ms) decay time. The Cr^{3+} ion on a Sc^{3+} site is a low-field emitter, with a peak emission at 775 nm and a fluorescence decay time of about 30 μs .

We found some indication of energy transfer from Al^{3+} -site to Sc^{3+} -site Cr ions. The transfer efficiency was low at the Cr^{3+} concentrations studied (0.1 and 1.0 at.% in the melt). Laser action was observed with Kr^+ laser pumping. A slope efficiency of 31% was measured, using an output coupler with a transmission of 4.19%. Using two output mirrors with different transmissions, we found that the maximum slope efficiency that could be attained in the limit of no cavity losses is 39%. Thus, in its present stage of development this material does not appear to be as promising as alexandrite. There are also material issues, such as the cost of Sc_2O_3 and the toxicity of BeO . This mediocre performance may have resulted from the poor optical quality of the samples, so we intend to look at a sample with higher optical quality and with optimum doping.

Thus our investigations on the metaphosphates indicate that the host cation radius necessary to accommodate Nd^{3+} ions is too large to prevent substantial nonradiative relaxation of excited Cr^{3+} ions. This results in low sensitization efficiency. Although it is difficult to satisfy

all the requirements of a Nova-like system with an oxide host, we are continuing to look for suitable candidates. The laser-pumped fusion driver poses entirely new requirements. Our studies show that a suitable Cr^{3+} laser based on an oxide host requires a crystal with a single site that has a well-defined symmetry for the substitution of Cr^{3+} . It also appears that this site will normally be occupied by an Al^{3+} or Ga^{3+} ion. We will use these guidelines to select candidates for study in 1988.

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Halide Laser Materials

Our work on halide laser materials in 1987 was guided by the requirements of the two advanced fusion driver concepts discussed in the previous article. For the laser-pumped fusion driver concept, $\text{LiCaAlF}_6\text{:Cr}$ has emerged as a particularly promising candidate; its spectroscopic and laser properties are described in the next article. For flashlamp-pumped, sensitized media, our experience with the perovskites and a review of the literature suggest that efficient Cr^{3+} sensitization in the halides will be difficult, because these compounds do not allow the simultaneous substitution of trivalent lanthanide and transition-metal ions.^{84,85}

We have proposed the use of host media for which codoping with Cr^{3+} and Nd^{3+} ions results in their being preferentially paired.⁸⁶ This can raise the nonradiative energy transfer efficiency by orders of magnitude over that calculated for a random distribution of ions, thereby allowing the flashlamp pump energy to be used effectively with lower doping.

This type of pairing is known to occur⁸⁷⁻⁸⁹ in materials with the formula AMX_3 , where A is a monovalent cation, M is a divalent cation, and X is a halide. Of these materials, CsCdBr_3 has received the most study. We have studied the spectroscopic properties of CsCdBr_3 singly doped and codoped with Cr and Nd.

Our measurements of the emission spectrum and fluorescence decay of $\text{CsCdBr}_3\text{:Nd}$ agree with those obtained in previous studies.⁸⁹ No fluorescence was detected at room temperature in $\text{CsCdBr}_3\text{:Cr}$, and an excitation spectrum of the Nd^{3+} fluorescence in $\text{CsCdBr}_3\text{:Cr,Nd}$ showed that no energy transfer from Cr^{3+} to Nd^{3+} occurred. Measurements of the fluorescence intensity and decay of Cr^{3+} in CsCdBr_3 as a function of temperature show that thermal quenching occurs above 150 K. The nonradiative rate appears to be much higher than the energy transfer rate. The peak intensity of the low-temperature fluorescence is at 975 nm; the poor overlap with Nd^{3+} absorption contributes to the lack of sensitization.

Pairing is reported to occur⁹⁰ in fluoride perovskites doped with trivalent ions. Our survey of the perovskites⁸⁴ suggested that the best candidate for Cr^{3+} sensitization of Nd^{3+} was KCaF_3 , so we grew singly doped and codoped single crystals of this material. In the perovskites, Cr^{3+} emission peaks near 830 nm, which reduces the overlap with the Nd^{3+} absorption relative to some of the oxide garnets. Nevertheless, the room-temperature excitation spectrum of Nd^{3+} fluorescence indicates that sensitization does occur, although the melt concentration of dopants was low (only 0.5 mol% Nd_2O_3). More measurements must be performed before we can ascertain whether pairing is the unambiguous explanation for the observed energy transfer.

After we found that $\text{LiCaAlF}_6\text{:Cr}$ is an unusually efficient laser medium, we searched for other fluoride compounds (Table 5-20) with analogous structures or compositions. Differential thermal analyses (DTAs) of samples prepared in platinum capsules indicated complete melting of the starting materials. Powders were

Table 5-20. Peak emission wavelengths of several Cr-doped fluoride hosts.

Host	Peak emission wavelength (nm)	Comments
LiSrAlF_6	810	Same structure as LiCaAlF_6
LiBaAlF_6	785	Hexagonal structure
LiMgAlF_6	775	Hexagonal structure
NaCaAlF_6	760	Compound did not form
KCaAlF_6	750	Compound did not form
LiCaAlF_6	760	Laser performance discussed in next article

obtained after cooling, and the resulting crystal structures were confirmed using x-ray diffraction. Table 5-20 lists the fluorescence peak wavelengths. $\text{LiBaAlF}_6\text{:Cr}$ may be another suitable laser candidate, because its free-running laser output would better match the absorption bands of Nd^{3+} or Tm^{3+} than would that of $\text{LiCaAlF}_6\text{:Cr}^{3+}$. However, ESA may be a problem because of the longer emission wavelength.

In addition to investigating candidate pump lasers, we studied a potential driver medium, $\text{Na}_2\text{Y}_3\text{F}_{11}\text{:Nd}^{3+}$. This material has a long lifetime, about 0.9 ms, which allows effective temporal integration of the pump radiation. It also has inhomogeneously broadened absorption bands, which raises its flashlamp pumping efficiency. Its cross section for the $\text{Nd}^{3+} {}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ gain transition is low, however—about $1 \times 10^{-20} \text{ cm}^2$. Other measurements of its spectroscopic properties have been made,⁹¹ but no data on its concentration quenching properties existed.

We grew samples of $\text{Na}_2\text{Y}_3\text{F}_{11}\text{:Nd}$ by the horizontal-melt-zone technique, with starting concentrations of 0.1, 0.5, 2.0, 5.0, and 10.0 mol% NdF_3 . An inductively coupled plasma (ICP) analysis of the 0.5 mol% crystal indicated that the segregation coefficient of Nd^{3+} in this host is 0.9 ± 0.1 . The crystals had very good optical quality for dopings as high as 5 mol%. The fluorescence decay was almost single-exponential at low dopings, with a lifetime of about 0.86 ms. For the 2.0-mol% sample, the decay exhibited classic concentration quenching behavior, with a decay constant at long times of 0.38 ms. The lifetime for a 5.0-mol% sample was 0.32 ms. Thus, it appears that the quenching behavior of this material is not unique, and that the quenching occurs at a concentration too low to make this a good candidate for a flashlamp-pumped fusion driver. On the other hand, the peak cross section of the ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{9/2}$ transition may be too low for the laser-pumped fusion driver concept. More evaluation is required to determine if this is the case.

In the 1986 Laser Annual⁹² we noted research on compounds with the formula LnMF_6 , where $\text{Ln} = \text{La, Gd, or Y}$, and where $\text{M} = \text{Al, Ga, Sc, or In}$. There are

no previously reported investigations on these materials. Compounds of this type could be useful if the M-ion is six-fold coordinated, because they should permit codoping and might permit efficient sensitization of Cr^{3+} and Nd^{3+} in a fluoride host.

Starting materials were loaded in sealed platinum capsules and placed in a DTA apparatus. We attempted to form the compounds by repeated cycling of the reactants in the temperature region where melting and freezing were found to occur. X-ray diffraction analysis of the fused products appears to indicate that, for most of the compositions studied, only mixtures of the starting components formed. The exceptions were in the LnInF_6 system. At the time of our study we believed that the emission of Cr^{3+} in these materials would occur at too long a wavelength to result in useful sensitization. We further explored the $\text{LaF}_3\text{-AlF}_3$ system, with compositions of 2:1 and 1:2. These compositions did not react either.

A recent study of $\text{Na}_3\text{In}_2\text{Li}_3\text{F}_{12}\text{:Cr}^{3+}$ reports⁹³ that Cr^{3+} emits at 770 nm. If the Cr^{3+} emission in LnInF_6 occurs at the same wavelength, efficient sensitization may result. We believe that our investigations on the laser-pumped fusion driver concept have a higher priority, however, and will continue this line of study only as time permits.

We are continuing our work on pairing systems; issues remain on the mechanical strength of the candidates under study and on the prospect for achieving useful dopings and high optical quality. We are also continuing to search for new laser pump sources and driver media.

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LiCAF ($\text{LiCaAlF}_6\text{:Cr}^{3+}$): A Promising New Solid State Laser Material

One concept for a 10-MJ fusion research laser uses a gain medium that is pumped

to a high stored energy density by an efficient, low-cost, long-pulse laser operating near 800 nm. A phonon-terminated, chromium-doped solid state laser is one possible type of pump laser for this application. Computational modeling⁹⁴ indicates that 10% efficiency is a reasonable technical goal for chromium lasers. Accordingly, we are pursuing a directed search for a 10%-efficient chromium laser material that has the potential to meet the cost goal of about \$1/cm³. LiCaAlF₆:Cr (colquiriite, or "LiCAF") is the first candidate laser material to emerge from this search that we judge to have the potential of meeting the stated requirements. (The name of the material comes from the Colquiri tin mine where the mineral was discovered.⁹⁵)

Laser operation activated by the Cr³⁺ ion has been observed in at least 16 host materials, although only BeAl₂O₄:Cr³⁺ (alexandrite)⁹⁶ and Be₃Al₂(SiO₆)Cr³⁺ (emerald)⁹⁷ have demonstrated conversion efficiencies of over 50% in a laser-pumped laser configuration. Neither alexandrite nor emerald has the properties required for an ICF pump laser. Emerald crystals can be grown only in small sizes and with great difficulty and expense. Alexandrite has good thermomechanical properties,^{98,99} but it is moderately difficult to grow, and laser efficiencies of no more than 5% have been achieved, substantially less than the theoretical value of ~10%. In view of this lack of a suitable existing Cr³⁺ laser host, we are screening alternative materials for the single-shot pump laser application.

We selected LiCAF for investigation because it satisfies the criteria for reduced ESA developed from the research discussed in "Effect of Excited-State Absorption on the Performance of Cr³⁺ Vibronic Lasers," later in this section. In this article we report the spectroscopic properties and laser efficiencies of Cr³⁺:LiCAF. We remeasured the efficiencies of the Cr³⁺ laser materials ScBO₃:Cr³⁺ (Ref. 100) and alexandrite (Ref. 96) in order to compare these materials with LiCAF using measurements obtained with the same apparatus and procedures.

LiCAF is a hexagonal crystal with a single Al site in the unit cell that provides a slightly distorted octahedral fluorine coordination for substitutional Cr³⁺. The

material melts congruently at 780°C. We grew single crystal samples with volumes of over 1 cm³ using horizontal zone melting in a platinum boat. The uniform coloration of Cr³⁺-doped crystals suggests that the distribution coefficient of Cr³⁺ is near unity. These characteristics suggest a low ultimate unit cost of crystal growth.

Figure 5-40 shows the absorption and emission spectra of LiCAF:Cr, referred to scales having absolute units of cross section. Since LiCAF is uniaxial, the spectra were recorded with the light polarized parallel and perpendicular to the *c*-axis. The broad absorption bands at 16 000 and 23 500 cm⁻¹ are due to the ⁴A₂ → ⁴T₂ and ⁴A₂ → ⁴T₁a transitions, respectively. The sharp features near 15 400 and 16 400 cm⁻¹ arise from the ²E and ²T₁ excited states. The emission spectra are typical of the ⁴T₂ → ⁴A₂ transition; the peak is near 13 200 cm⁻¹, and the FWHM is about 2000 cm⁻¹.

Emission decays were nearly single-exponential at all temperatures. Lifetimes varied little with temperature—from 200 μs at 20 K to 175 μs at room temperature.

Fig. 5-40. Polarized absorption and emission spectra of LiCaAlF₆:Cr³⁺ at room temperature, plotted in absolute units of cross section, for a sample with 1.9×10^{19} Cr³⁺ ions/cm³ and a path length of 0.85 cm.

Fig. 5-41. Tuning curve and emission cross section of $\text{LiCaAlF}_6:\text{Cr}^{3+}$ ($2.9 \times 10^{19} \text{ Cr}^{3+}$ ions/ cm^3). The short wavelength limit is attributed to the ground state absorption of Cr^{3+} .

It is therefore unlikely that nonradiative decay is important at room temperature, since nonradiative decay of Cr^{3+} excited states is usually accompanied by a precipitous decrease in emission lifetime as temperature is increased.

The procedure used to measure the tuning curves and slope efficiencies is similar to that reported previously,¹⁰¹ using apparatus initially devised by Caird and Staver.¹⁰² The pump source is a Kr^+ laser operating at 647 nm, mechanically chopped at 52 Hz to generate 1-ms square pulses. A lens of 10-cm focal length is used to longitudinally pump the crystal; the pump beam radius is 40 μm . The laser resonator is of a concentric design; the cavity mirrors have a 5-cm radius of curvature. With this resonator geometry, the cavity waist is much larger than the pump beam, so the slope efficiencies are limited only by properties of the gain medium. A birefringent filter was inserted into the cavity to tune the laser output wavelength.

To determine the slope efficiency, one measures the pump power absorbed by the crystal and the output power from

Fig. 5-42. Single-run slope efficiency data (efficiencies in parentheses) for LiCAF , BeAl_2O_4 , $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}$, and ScBO_3 lasers.

the laser. The $\text{Cr}^{3+}:\text{LiCAF}$ laser output is plotted vs wavelength in Fig. 5-41; the emission cross section is shown for comparison. The wavelength of maximum output and the tuning range are determined not only by the emission cross section but also by the cavity mirror transmission and the Cr^{3+} ground state absorption, both shown in Fig. 5-41. The measured maximum of the tuning curve is near 780 nm; this is to be compared with the peak cross section, which occurs near 760 nm. The short-wavelength cutoff is probably due to the onset of the Cr^{3+} ground state absorption. This is reasonable, because the main effect of the ground state absorption is to raise the laser threshold power. When we tried to tune beyond 840 nm, the laser output switched back to shorter wavelengths that correspond to a different order of peak transmission of the tuner plate. Our previous study of the $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ laser¹⁰¹ leads us to expect that the long-wavelength limit will be influenced by the lower emission cross section and by the effects of excited-state absorption.

Figure 5-42 shows typical data plots for four Cr^{3+} solid state laser materials, obtained with output couplings of 4.1 to 5.2%. The $\text{Cr}^{3+}:\text{LiCAF}$ and $\text{BeAl}_2\text{O}_4:\text{Cr}^{3+}$ lasers have considerably higher slope efficiencies than the $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ and $\text{ScBO}_3:\text{Cr}^{3+}$ lasers. It is important to realize, however, that the threshold powers are, in general, sensitively dependent on cavity parameters, so that no useful information can be obtained by considering the relative thresholds appearing in Fig. 5-42.

The dependence of laser output power P_o on pump power P_p is given by¹⁰³

$$P_o = \left[\left(\frac{h\nu_e}{h\nu_p} \right) \eta_p \left(\frac{\sigma_e - \sigma_{\text{ESA}}}{\sigma_e} \right) \right] \left(\frac{T}{T + L} \right) (P_p - P_p^{\text{th}}) \quad (39)$$

for above-threshold operation ($P_p > P_p^{\text{th}}$). Here $h\nu_e$ and $h\nu_p$ are the photon energies of the Cr^{3+} laser output and the pump laser, η_p is the fraction of absorbed pump photons that populate the

Fig. 5-43. Intrinsic slope efficiency determinations for the four crystals of Fig. 5-42. Several runs were averaged to obtain each of the data points shown.

upper laser level, σ_e and σ_{ESA} are the emission and excited-state absorption cross sections at the lasing wavelength, T is the transmission of the output coupler, and L is the passive cavity loss (e.g., sample surface scattering and impurity absorption at the lasing wavelength). Equation (39) can be written as

$$P_o = \eta_o \left(\frac{1}{1 + L/T} \right) (P_p - P_p^{\text{th}}) \\ = \eta(P_p - P_p^{\text{th}}), \quad (40)$$

where η_o embodies the intrinsic losses (i.e., losses intrinsic to the properties of the Cr^{3+} ion) and therefore represents a fundamental limit to laser slope efficiency. A plot of the inverse of the measured slope efficiency η^{-1} vs T^{-1} will yield a line with intercept at $T^{-1} = 0$. This procedure separates out the effects of the passive loss L .

Using about two to four runs of the sort shown in Fig. 5-42 for each of two output couplings, we generated the plots shown in Fig. 5-43, whose intercepts yield intrinsic slope efficiencies. $\text{Cr}^{3+}:\text{LiCAF}$ and $\text{BeAl}_2\text{O}_4:\text{Cr}^{3+}$ have intrinsic efficiencies of 67 and 65%, approaching the quantum-defect-limited value of about 85%.

Both $\text{ScBO}_3:\text{Cr}^{3+}$ and $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ are considerably less efficient. In seeking the reasons for this low efficiency, we have shown¹⁰¹ that the $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ laser is beset by substantial ESA losses at the lasing wavelengths. This was determined by directly measuring the ESA in a separate pump-probe experiment (see "Effect of Excited-State Absorption on the Performance of Cr^{3+} Vibronic Lasers," later in this section). We also demonstrated that the quantum yield of $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ is nearly unity ($\eta_p = 1$), so that poor pumping efficiency cannot account for the low slope efficiency. We believe that non-negligible ESA exists in all Cr^{3+} solid state lasers, although it is clearly much weaker for LiCAF and $\text{BeAl}_2\text{O}_4:\text{Cr}^{3+}$ lasers than for ScBO_3 and $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}$.

In view of the high slope efficiency achieved in our measurements on

$\text{Cr}^{3+}:\text{LiCAF}$, our goal of a 10% overall efficiency may be attainable with this material.

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Advanced Large-Dimension Crystal Growth

Introduction

The goal of this project¹⁰⁴ is to develop technologies for growing crystals with the dimensions, optical quality, and thermomechanical durability required by lasers for fusion reactor drivers and for other applications of national importance. The project is jointly supported by the ICF program and by LLNL Director's Initiative funds.

In 1987 we concentrated on developing a novel process (called "moving melt zone" or MMZ) for growing crystals from high-temperature melts. In this configuration (shown in Fig. 5-44), an ampoule of rectangular cross section is lowered through a temperature-zoned furnace in such a way that a melt zone of fixed volume is kept in contact with the growing crystal. The melt zone is replenished by the progressive depletion of a polycrystalline feed slab. By this technique, crystals may be grown with the slab-like aspect ratios appropriate to laser amplifiers, frequency converters, and electro-optic elements. The process is broadly applicable to materials such as refractory oxides and fluorides grown from congruently melting compositions (in which the crystal and melt have identical compositions).

Large-aperture optical crystals are at present grown mostly by the Czochralski (CZ) process.¹⁰⁵ (See "Large Dimension Czochralski Crystal Growth," in Sec. 6.) In the CZ process, a seed crystal is

Fig. 5-44. In the moving melt zone process, an ampoule of rectangular cross section is lowered through a zonal furnace, in which the temperature distribution is such that a melt zone of constant dimensions separates the growing crystal from a progressively depleted polycrystalline feed slab.

touched to the surface of a melt in a crucible and is then rotated and slowly withdrawn to form a cylindrical boule of larger diameter than the seed. Czochralski is a batch process, during which the melt volume and the composition and temperature distributions change continuously. This complicates process control and results in a crystal of nonuniform dopant concentration. A major limitation of the CZ process is periodic fluid flow in its large-volume melts, which results in temperature fluctuations and consequent growth striations at the interface.¹⁰⁶ Other limitations include difficulty in controlling interface shape,¹⁰⁷ loss of melt components by evaporation, and incompatibility of cylindrical boules with the production of large plates. Some of the problems inherent in the CZ technique may be lessened with the use of a shallow inner crucible to

stabilize fluid flow and dopant concentration.¹⁰⁸ We were motivated to search for a more favorable process by difficulties we encountered with CZ growth¹⁰⁹ and by the desire to avoid or minimize the problems listed above.

The MMZ process is configured so as to maintain a small and constant-volume melt adjacent to the growing crystal. Because of the constancy of melt volume and continuous feed, the melt and crystal composition and the temperature distribution do not change with time, and optimized growth conditions may be maintained throughout growth.

A tendency towards periodic fluid flow increases with melt dimensions. Stable flow, which is required if temperature fluctuations and striae are to be avoided, is achieved by minimizing the melt dimensions. The process uses a closed

rectangular ampoule to reduce material losses and to achieve the desired plate geometry directly.

The optical quality of crystals, no matter how they are grown, depends on melt and atmosphere composition, on the presence of trace contaminants, and on the physical and chemical compatibility of melt, crucible, and contacting furnace parts. Thus the development of MMZ involves materials issues and requires the engineering of a furnace with favorable temperature distribution and stability.

Testing this concept requires a furnace of specialized design. Successful application of this process requires the development of advanced thermal diagnostics to allow accurate determination of temperature distribution in the furnace and to detect small-amplitude temperature fluctuations. To aid in the prediction and interpretation of temperature distribution and stability during crystal growth, we have developed and applied physical and computational models of

Fig. 5-45. To test the MMZ concept, Crystallox, Ltd., is developing a furnace with an inductively heated stack of graphite disks with a rectangular axial cavity to accommodate the ampoule.

magnetic-induction heating of the furnace and of convective heat and mass transport in high-temperature melts.

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Advanced Crystal Growth Technology Development

The primary objective of our research during 1987 was to begin developing a specialized, inductively heated furnace for testing the MMZ concept described in the preceding article. In parallel with the furnace development, a gradient-freeze furnace (which has certain similarities with MMZ) was developed and used to grow YAG:Nd crystals to permit testing diagnostics and control schemes and to resolve materials problems we expect to encounter with MMZ. To measure temperature gradients and to detect small temperature fluctuations, we installed a scanning optical-fiber thermometer in the furnace. Temperature distribution in the MMZ process was modeled using a commercial finite-element fluid dynamics code, FIDAP.¹¹⁰ We also modeled inductive heating and the resulting temperature distribution in the furnace.

Development of a Furnace for Testing the MMZ Concept. A contract was awarded to Crystallox, Ltd. (Wantage, U.K.) for development of a furnace for testing the MMZ concept. The Crystallox design (Fig. 5-45) is based on an inductively heated graphite core, or susceptor. The core, made up of a stack of graphite disks, is heated by two 40-kW, 25-kHz generators. The core contains an axial slot of rectangular cross section to accommodate the ampoule, which is held in a graphite cradle that is lowered into the core slot by a precision mechanical actuator. The susceptor is surrounded by low-density graphite felt insulation. The interface between the growing crystal and the melt is maintained within a central zone, which is insulated to promote flat or slightly convex isotherms and a steep temperature gradient. The modular design allows segments of the susceptor to be removed and modified as required to fine-tune the temperature distribution.

Such modifications will be necessary to achieve azimuthal temperature uniformity on the surface of the ampoule in the vicinity of the growth interface.

The furnace will be delivered in late 1988 as a Bridgman furnace (i.e., a two-zone furnace for growth of a crystal from a progressively depleted melt). In Bridgman growth, the dopant concentration increases along the length of the crystal because of the selective rejection of the dopant species. Most of the engineering issues associated with MMZ will be addressed in this simpler furnace configuration. After initial testing, the furnace will be modified for MMZ growth by the addition of a third heater and a second insulating zone, as shown in Fig. 5-44.

Modeling Convective Heat Transport in MMZ. We continued modeling work begun in 1986 on coupled heat transport and fluid flow in the MMZ process, using the FIDAP code to perform the simulations. The calculations were primarily steady-state, and were designed to evaluate the sensitivity of model predictions to as-yet unknown material properties and to process imperfections. A limited number of time-dependent calculations were carried out to determine whether the flows are stable in the presence of various perturbations.¹¹⁾

We examined the effects of melt viscosity, solid and melt thermal conductivities, heat loss through the insulated

("adiabatic") zone, and crucible wall thickness on melt flow structure and melt/crystal interface shape. We found that convection always had a negligible effect on the lower interface because of the influences of the adiabatic zone and the stabilizing temperature gradient, both of which damp fluid flow in the vicinity of the interface. But we found that the melt viscosity has a strong effect on the flow structure (Fig. 5-46). As viscosity increases from 4 to 40 centipoise the lower flow cell weakens and eventually vanishes. The viscosity of YAG:Nd is not known within this range, so any of the flows shown in the figure are possible. While the shape of the interface remains relatively constant, the different flow structures may have a significant effect on mass transfer and hence on the dopant distribution within the crystal. This issue will be examined further in 1988.

Lateral heat loss through the adiabatic zone also affects the melt flow structure. As lateral heat flow increases, the lower flow cell becomes stronger because of the greater driving force for fluid flow. This phenomenon may affect mass transfer and crystal dopant distribution.

The ratio of the thermal conductivities of the solid and the melt directly influences the interface shape; more convex interfaces result from higher ratios. For a 3:1 solid-to-melt conductivity ratio, the

Fig. 5-46. Flow structure, as visualized by streamlines, depends strongly on melt viscosity μ . (Prandtl and Rayleigh numbers are shown.)

interface is still flat enough to prevent undesired facet formation.¹¹¹

The time-dependent studies demonstrated the stability of a steady-state solution to perturbations in the flow and temperature structure. In all cases, the perturbed initial state returned to the original steady-state solution. This implies that the hydrodynamically induced striations that result from periodic or chaotic flows in large-scale Bridgman and Czochralski systems will not arise in the MMZ. To verify this conclusion, which is based on two-dimensional simulations, we will carry out fully three-dimensional simulations in 1988.

Modeling Induction Heating of a Graphite Susceptor. In conjunction with procurement of the Crystalox Bridgman furnace, we began modeling studies to aid in the design of the susceptor, insulation, and coils. The modeling is necessary because of the complicated shape of the susceptor (a cylinder with a rectangular channel through it). Azimuthal temperature uniformity, to some precision, is required to maintain a flat crystal

interface; this requirement can be met with simple modifications of the susceptor design.

We model the induction heating of the susceptor with a modified version of FIDAP that solves the time-dependent Maxwell equations with a harmonic input. This approach, developed by Gresho and Derby¹¹² for modeling the induction heating of a Czochralski crucible, was extended to the MMZ configuration. Using the induced heating in the susceptor as a source term, FIDAP solves the three-dimensional heat conduction equation to determine the temperature distribution in the susceptor and the insulation. These models allow us to examine the effects of coil spacing, susceptor geometry, and alternative materials on the susceptor temperature distribution.

Preliminary results show only a 15°C azimuthal temperature variation in the vicinity of the growth interface in a 40°C/cm vertical temperature gradient (Fig. 5-47). The interface can be represented as the melting point isotherm (not shown in the figure). The 15°C variation would therefore produce only a 4-mm deflection in the interface (for a 40°C/cm temperature gradient), which is well within tolerances for the prevention of facet formation.

A remaining difficulty is that this susceptor configuration produces temperatures that are highest near the center of the growth zone, which would tend to produce a concave interface. We are examining configurations that will reverse the heat flow near the growth zone by providing active heating in this vicinity. Modeling work in 1988 will be complemented by induction-heating experiments with representative materials in a nearly full-scale furnace to verify model predictions and to examine susceptor configurations that are not readily modeled, such as a radiation hohlraum to aid in reducing azimuthal temperature variations.

Growth of YAG:Nd and Diagnostics Development in a Gradient-Freeze Furnace. In a gradient-freeze furnace, a vertical temperature gradient is maintained in the vicinity of the ampoule by means

Fig. 5-47. We use induction heating and 3-D heat transfer calculations to determine temperature distributions in candidate MMZ susceptor designs. (a) Finite-element mesh; (b) in-phase component of the magnetic field; (c) section view of susceptor, insulation, and ampoule (isotherms at 100°C intervals).

of specially designed shielding. As power is reduced, the crystal is allowed to grow in accordance with the upwards displacement of the melting-point isotherm (Fig. 5-48). Experiments in this configuration yield information valuable to the design of the MMZ, because the two techniques share the features of crucible-contained growth, vertical stratification of melt density, and analogous profiles of temperature and chemical activity. The growth of high-quality, flat-interface YAG:Nd (Ref. 113) and titanium sapphire¹¹⁴ by this method provides data that validate the MMZ concept, and establishes a benchmark for evaluating crystals grown in our gradient-freeze furnace.

A vertical temperature gradient of $10^\circ\text{C}/\text{cm}$ (the minimum gradient capable of preventing supercooling of the melt) was achieved by surrounding the tungsten crucible with shields whose number decreased with distance from the cold pedestal. The gradient was initially measured with W/W-26%-Re thermocouple junctions formed by wrapping W-26%-Re wires about the crucible at various points along its vertical axis. The gradient was also measured with an optical-fiber pyrometer.^{115,116}

We determined the critical temperature gradient for YAG:Nd growth as follows. During growth, neodymium ions are rejected at the growth interface; this rejection is reflected by a low segregation coefficient, $k = 0.16$. (The segregation coefficient is the ratio of the solid and melt dopant concentrations at the interface). The buildup of Nd^{+3} at the interface lowers the melting point. If the thermal gradient in the melt adjacent to the interface is not high enough, the melt may become supercooled and unstable. The critical gradient required to prevent supercooling is given by¹¹⁷

$$\nabla T = \frac{vmC_{\text{Nd}}k - 1}{Dk}, \quad (41)$$

where v is the growth velocity, m is the slope of the liquidus curve of the YAG: Nd_2O_3 phase diagram, C_{Nd} is the concentration in the crystal, which is identical with x in the formula $(\text{Y}_{1-x}\text{Nd}_x)_3\text{Al}_5\text{O}_{12}$, and D is

Fig. 5-48. In a gradient-freeze furnace, a vertical temperature gradient is maintained in the vicinity of the ampoule by shielding. As power is reduced, the crystal grows as the melting-point isotherm is displaced upwards.

the diffusion coefficient of Nd in the melt. With the estimated values $D = 2 \times 10^{-9} \text{ m}^2/\text{s}$ and $m = -34^\circ\text{C}$ (atomic substitution basis), Eq (41) indicates that a gradient of $10^\circ\text{C}/\text{cm}$ is required for a growth of 1%-doped YAG:Nd at 10^{-7} m/s (0.36 mm/h).

Single crystals such as that shown in Fig. 5-49 were grown under these conditions in an argon atmosphere in closed ampoules. The crystals are free of the multiphase morphologies associated with constitutional supercooling. The strong curvature of the crystal at the crystal/melt/ampoule contact is the result of heat conduction through the metal and associated distortion of the thermal gradient.¹¹⁸ A convex curvature, which results from the greater thermal conductivity of the crystal than of the melt, has the favorable effect of directing crystallographic dislocations that originate near the ampoule wall away from the central portions of the crystal. With larger crystal diameters, the interface shape will flatten; we will control the flattening by appropriate choices of wall thickness and overall furnace thermal profiles.

Fig. 5-49. Crystals 3 to 5 cm long are grown under well-defined temperature gradients. Heat transfer and birefringence models predict correlations between power fluctuations and striations in the crystal.

YAG:Nd grown by gradient-freeze technique from 0.01-Nd melt and seed at 0.63 mm/h

Crystallographic Striations and Temperature Control. Striations in crystals are periodic variations of optical properties along the direction of growth. In YAG:Nd, a periodic strain-induced birefringence results from changes in local dopant concentration. Hayakawa¹⁰⁶ developed a model relating stress-birefringence to a lattice mismatch resulting from abrupt changes in dopant concentration. Using thermodynamic data, we extended the model to allow us to predict the dependence of birefringence on temperature fluctuations; we obtained the linear dependence

$$\frac{d\Delta n}{dT} = 3 \times 10^{-6} \text{ } ^\circ\text{C}^{-1} \quad (42)$$

We used Eq. (42) to estimate the temperature and power stability requirements for the MMZ process. For a birefringence of less than 3 nm/cm, the interfacial temperature must be stable to

within 0.1°C, which requires a power stability of 0.0% in an idealized, unmuffled furnace.

In a real furnace, the crystal itself and any extrinsic mass will attenuate temperature fluctuations by the factor

$$\frac{\Delta T_i}{\Delta T} = \exp\left(-x\sqrt{\frac{\omega}{2\alpha}}\right), \quad (43)$$

where ΔT_i is the temperature fluctuation of the crystal growth interface, ΔT is the amplitude of the unmuffled temperature fluctuation of frequency ω , and x and α are the thickness and thermal diffusivity of the muffler. For YAG, $\alpha \approx 0.0025 \text{ cm}^2/\text{s}$; for $x = 1 \text{ cm}$ and for oscillations of period 1000 s, the attenuation factor given by this relation is 3. Thus the 0.1°C interface temperature stability requirement is conservative: 0.3°C fluctuations of periods less than 1000 s will produce birefringence of less than 3 nm/cm.

The commercial power controller supplied with the furnace was unable to give

Fig. 5-50. Temperature control has been characterized and improved for crystal growth applications. (a) Commercial controller supplied with furnace; (b) micro-computer control with LLNL software. (Both are closed-loop controls.)

the control necessary to achieve crystal growth without striations, so we wrote process-control software for use with a dedicated microcomputer (Hewlett-Packard 9816) and data acquisition system (Hewlett-Packard 3497A). Figure 5-50 shows the improvement obtained with our software for a programmed linear power rampdown. The commercial controller permitted 10°C fluctuations over periods of 100 s; these would lead to pronounced striae and probably to constitutional supercooling. The improved control achieved with the LLNL process control system (in the closed-loop mode) arose primarily from increasing sampling frequency to 5 Hz and from experimental tuning of the proportional integral derivative coefficients. The fluctuations at around 1000, 2500, and 3500 s are still large enough to produce discontinuities in dopant concentration and associated strain-birefringence. Further improvements in power stability are being sought through combinations of muffling and process control strategies involving response functions, which we have measured.

In addition to the increased control capability, our control program also incorporates temperature-dependent control

coefficients that allow us to reduce furnace power gradually to zero, following a linear or programmed function of time, after growth is complete. Thermal forces, which produce cracking or disintegration of the crystal, are thereby minimized. The program also collects data from our thermal and power diagnostics for use in process diagnosis and design.

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Basic Material Research

Introduction

The Optical Materials Research Program is supported by the Office of Basic Energy Sciences (OBES) of the Department of Energy through its Division of Materials Science. This program involves basic research on topics and materials of importance to the Laser Program. Three research areas of intermediate or

long-range impact on laser development provide the framework for our research effort:

- Spectroscopy and theory of optically active ions in crystals and glasses.

- Measurement and modeling of the magnitude and dispersion of the nonlinear refractive indexes of optical materials.

- Fundamental mechanisms of optical surface damage in optical materials. In addition to the research done in the OBES program, work on composite crystalline-glass laser materials, funded by an LLNL Institutional Research and Development award, is also discussed in this subsection.

Spectroscopic and Theoretical Studies of Optically Active Ions in Crystals.

In 1987 our work emphasized ESA of laser ions in crystals. This topic is of great importance to the development of efficient solid-state lasers; research on Cr^{3+} and V^{2+} in particular has had an immediate impact on the search for an efficient Cr^{3+} pump laser for a multimegajoule ICF laser [see "LiCAF ($\text{LiCaAlF}_6:\text{Cr}^{3+}$): A Promising New Solid State Laser Material," earlier in this section]. The ESA spectra of Cr^{3+} and V^{2+} are strongly influenced by asymmetric relaxation of neighboring ions. We verified this conclusion by *ab initio* calculations of the adiabatic potential energy curves for the V^{2+} ion and its fluorine neighbors in KMgF_3 and MgF_2 . This knowledge enabled us to develop a few simple criteria that can be used for selecting crystalline hosts in which Cr^{3+} and V^{2+} will have low ESA cross sections at the laser wavelength, and that will therefore exhibit good lasing efficiency.

ESA measurements on the Sm^{2+} ion in fluoride-structure hosts indicate that ESA is likely to be a problem for laser media based on this ion. Our ESA spectra permitted us to make the first determination of the intraionic coupling between the 4f and 5d electrons in the excited states of the $4f^65d$ configuration, which had long been a topic of controversy.

A comparison between measured spectroscopic properties of Cr^{3+} in scandium sites in ScF_3 and Sc_2O_3 and published spectra of Cr^{3+} in numerous related oxide and fluoride crystals gave us

insight into the relative influence of the oxygen and fluorine neighbors on the crystal field splitting of Cr^{3+} , and showed that next-nearest-neighbor cations also have a significant influence on the crystal field. A qualitative understanding of this sort is useful in searching for hosts for tunable Cr^{3+} lasers with wavelengths and efficiencies needed for various applications.

We surveyed a number of ceramic-crystalline composites, which consist of very small crystalline particles embedded in a glass matrix. It had been hoped that these materials would lead to low-cost laser media based on ions such as Cr^{3+} that normally have poor spectroscopic properties in glasses. Although the spectroscopic properties of the Cr^{3+} ion are markedly improved in composites, we found that the scattering caused by the optical heterogeneity cannot be reduced enough to permit effective laser application of these materials.

Nonlinear Refractive Index of Optical Crystals. The nonlinear refractive index n_2 of optical media causes self-focusing and beam breakup in laser media and other optical components of high-power laser systems. A knowledge of the dependence of n_2 on composition, structure, and wavelength is useful for developing new optical materials and for using existing materials in new intensity and wavelength regimes. In 1986 we reported¹¹⁹ a rapid and reliable method for measuring n_2 in small samples and presented measurements for a number of glasses. In 1987 we completed measurements of n_2 at $1.06\ \mu\text{m}$ for a large number of single crystals and compared those values with values obtained from empirical models and from recent first-principles calculations of third-order susceptibilities in condensed media. We also deduced values for the third-order polarizabilities (hyperpolarizabilities) of the fluoride, oxide, and sulfide anions coordinated by various cation partners, and found that they are strongly dependent on the size and outer electron configuration of the neighboring cations. In 1988 we will perform n_2 measurements at 0.53 and $0.35\ \mu\text{m}$ to investigate the dispersion of the third-order susceptibility.

Fundamental Mechanisms of Optical Surface Damage. We are investigating the interaction of laser light with surfaces of optical materials by measuring the properties of the charged and neutral particles ejected in this interaction. We have found evidence in several optical materials for desorption of surface atoms and molecules by excitation of defect and surface states. The particle temperatures characteristic of this emission increase strongly with increasing laser fluence, suggesting an electric breakdown in the region near the surface. In a number of materials, high-energy neutral atoms and molecules are observed for laser fluences below the optical damage threshold; a likely origin for these neutrals is "fractoemission" caused by cracking or cleaving, which can result from quite modest heating of the surface region. There is also evidence for changes in surface composition caused by these laser-induced emission processes. These changes may affect both single-pulse and multiple-pulse surface damage thresholds. In collaboration with Argonne National Laboratory, we are using laser-induced fluorescence and laser-ionization spectroscopy to detect emitted neutrals. These experiments¹²⁰ offer improved sensitivity over the mass spectrometer techniques we use in our research at LLNL.

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Nonlinear Refractive Indexes of Optical Crystals

The nonlinear refractive index n_2 strongly influences the propagation of intense laser beams through optical materials. It causes self-focusing and amplification of spatial intensity fluctuations of an incident beam. (The nonlinear index is defined by

$$n = n_0 + n_2 \langle E^2 \rangle, \quad (44)$$

where n is the refractive index in the presence of an optical electric field E and n_0 is the linear refractive index. The brackets imply a time average over the optical period.) Our research is aimed at

gaining an understanding of the magnitude and wavelength dependence (dispersion) of n_2 for optical materials.

We have developed a rapid and reliable method,¹²¹ based on three-wave mixing, for measuring nonlinear refractive indexes of small samples of modest optical quality. The technique is sensitive only to the electronic and Raman-active vibrational contributions to the nonlinear index; the effects of electrostriction, thermally induced refractive effects, and optically excited free carriers are too slow to contribute to the measured signal. This feature of our technique has enabled us to determine n_2 values for nanosecond-pulse propagation at $1 \mu\text{m}$ for a large number of optical crystals and glasses. Although the ultimate goal of our research in this area is to extend these measurements to shorter wavelengths, the large $1\text{-}\mu\text{m}$ data base provides a unique opportunity to examine the influence of structure and composition on the nonlinear refractive behavior of materials.

Table 5-21 summarizes our single-crystal measurements. A complete description of the experimental method, its application to a number of optical glasses, and its relation to other measurements is given elsewhere.¹²¹⁻¹²³ Table 5-21 includes halides, oxides, and sulfides with a range of physical properties, encompassing ionic insulators, wide-bandgap semiconductors, and ferroelectric oxides.

It is interesting to compare these measurements with the predictions of a widely used, but relatively untested, empirical expression for n_2 developed by Boling, Glass, and Owyong (BGO) on the basis of a phenomenological model.¹²⁴ This formula, which relates n_2 to the linear index and its dispersion [through the Abbe number $v_D = (n_D - 1)/(n_F - n_C)$], is

$$n_2 \text{ (} 10^{-13} \text{ esu)} = \frac{K(n_D - 1)(n_D^2 + 2)^2}{v_D \left[1.517 + \frac{(n_D^2 + 2)(n_D + 1)}{6n_D} \right]^{1/2} v_D}, \quad (45)$$

Table 5-21. Nonlinear and linear refractive index data for some optical crystals.

Sample	n (1.06 μm) ^a	Abbe number, ν_D	n_2 (10^{-13} esu)	
			Measured	Calculated
LiF [100]	1.3866	98.0	0.26	0.40
NaF [100]	1.3213	85.2	0.34	0.38
KF [100]	1.3583	97.9	0.75	0.36
NaCl [100]	1.5312	42.9	1.59	2.28
KCl [100]	1.4792	44.1	2.01	1.84
NaBr [100]	1.6228	31.7	3.26	4.74
KBr [100]	1.5435	33.7	2.93	3.41
MgF ₂ (o)	1.3735	104.9	0.25	0.34
CaF ₂ [100]	1.4285	95.1	0.43	0.49
SrF ₂ [100]	1.4328	93.9	0.50	0.50
CdF ₂ [100]	1.56	61.0	3.95	1.37
BaF ₂ [100]	1.4682	81.8	0.67	0.70
LaF ₃ (o)	1.60	57.0	1.4	1.78
CeF ₃ (o)	~1.6	—	1.3	—
AgCl (polycrystalline)	2.020	21.2	23.3	23.0
MgO [100]	1.72	53.4	1.61	2.8
CaO [100]	1.83	—	5.20	—
SrO [110]	1.81	—	5.07	—
ZnO (c)	1.96	11.6	23.0	57.6
(a)	1.99	12.3	25.0	45.0
Al ₂ O ₃ (o)	1.75	71.8	1.23	1.9
(e)	1.75	75.2	1.30	1.8
Ga ₂ O ₃	1.96	—	5.80	—
Y ₂ O ₃	1.92	37.5	5.33	7.2
Er ₂ O ₃	1.96	—	4.53	—
SiO ₂ (fused)	1.4496	67.8	0.85	0.83
SiO ₂ (quartz) (o)	1.5342	71.6	1.12	1.06
(e)	1.5429	70.1	1.16	1.13
TiO ₂	2.48	9.8	55.8	189
ZrO ₂	2.12	35.8	5.8	12.6
BeAl ₂ O ₄	1.73	72.5	1.46	1.74
MgAl ₂ O ₄	1.72	60.6	1.50	2.3
CaMgSi ₂ O ₆	1.67	—	1.73	—
YAlO ₃ (γ)	1.933	51.2	3.37	4.88
Y ₃ Al ₅ O ₁₂	1.822	52.4	2.7	3.6
Gd ₃ Sc ₂ Al ₃ O ₁₂	1.891	48.0	4.0	5.6
Gd ₃ Sc ₂ Ga ₃ O ₁₂	1.943	37.3	5.5	8.0
Gd ₃ Ga ₅ O ₁₂	1.945	37.6	5.8	8.0
Y ₃ Ga ₅ O ₁₂	1.912	40.0	5.2	5.8
La ₃ Lu ₂ Ga ₃ O ₁₂	1.930	36.4	5.8	8.2
SrTiO ₃	2.31	13.6	26.7	83.0
CaCO ₃ (o)	1.6425	47.6	1.11	2.7
(e)	1.4795	76.8	0.83	0.79
KH ₂ PO ₄ (o)	1.4598	70.9	0.72	0.85
(e)	1.4938	56.6	0.78	1.35
K (TiO)PO ₄ ($x + y$)	1.74	23.5	5.73	10.0
L-arginine phosphate (y)	1.559	47.5	1.87	2.14
L-arginine phosphate ($y + z$)	1.51	50.0	3.04	2.0
KTaO ₃	2.25	—	29.0	—
CaWO ₄ (o)	1.89	30.0	4.2	10.4
(e)	1.91	28.0	5.6	12.0
ZnS (c)	2.29	15.5	49.3	64.0
ZnS (a)	2.29	15.5	47.3	64.0
CdS (c)	2.34	3.8	283.0	643.0
CdS (a)	2.33	4.4	304.0	494.0

^a Reference 123 gives source references to refractive index data.

where n_F , n_D , and n_C are the refractive indexes at the standard wavelengths 4861.3, 5875.6, and 6562.7 Å, respectively.

Figure 5-51 shows measured values of n_2 plotted vs those calculated from Eq. (45). The solid line corresponds to the constant $K = 68$ in the original BGO formula. The dotted line, which corresponds to $K = 48$, gives the best fit to the data; with this fit, the average absolute deviation from the predictions of the BGO formula is 30%. This represents great success for an empirical expression, given that the values of n_2 for these crystals vary over about three orders of magnitude.

The prevailing concept of polarizability in ionic materials involves the response of individual ions or molecular ions to the electric field of the light wave. The polarization p of these ions is expanded in a power series in the local field E_l acting

on them. Only terms up to the third order are needed here, so that we have

$$p = \alpha E_l + \frac{\gamma}{6} E_l^3, \quad (46)$$

where α is the linear polarizability and γ is the hyperpolarizability. The nonlinear refractive index of a material is given in terms of the hyperpolarizabilities γ_i and number densities N_i of the constituent ions by

$$n_2 = \frac{\pi f^4}{2n_0} \sum_i N_i \gamma_i, \quad (47)$$

where f is the local field factor, which gives the ratio of the local electric field to the macroscopic field. Equation (47) can be used either to obtain values of γ_i for various cations and anions or to

Fig. 5-51. Measured nonlinear refractive indexes of optical crystals vs values of n_2 calculated from the BGO formula [Eq. (45)].

calculate n_2 for materials containing ions of known γ_r .

The value of γ_i for a particular ion is dependent on its environment. This effect is more pronounced for the anions, which have much larger hyperpolarizabilities than the cations. This results from the fact that the outer electrons are more tightly bound in cations than in anions because of the opposite signs of the ionic charges. The cations have a strong influence on the magnitudes of the hyperpolarizabilities of the anions, however.¹²⁵ This is due to the compressive effect of the cation charge cloud on the weakly bound anion valence electrons. We must therefore treat the values of γ_i in Eq. (47) as effective hyperpolarizabilities of the anion-cation pair. For simple oxides, fluorides, and sulfides of a

single cation, Eq. (47) can be used to obtain the values of the effective hyperpolarizabilities per anion γ^- for coordination with that particular cation.

Tables 5-22 and 5-23 give the values of γ^- for fluorine, oxygen, and sulfur obtained from our measured values of n_2 for binary compounds. It is striking that these values differ by about a factor of 7 for the fluorine compounds and by a factor of 20 for the oxides. We have compared these values of γ^- with the predictions of a phenomenological model for the interaction of the anion and cation charge clouds.¹²⁶ The model predicts that the linear polarizability α^- of the anions is related to the nearest-neighbor distance R , which is, of course, determined by the cation radius; the relation is

$$\alpha^- = \alpha_0 \exp(-C/R^2), \tag{48}$$

where α_0 is a constant for a particular anion. Wang¹²⁷ has shown that the atomic hyperpolarizability γ is approximately proportional to the square of the linear polarizability for atoms having their lowest excited states at about the same energy. Thus, γ^- can be related to R by

$$\gamma^- = \gamma_0 \exp(-D/R^2), \tag{49}$$

where γ_0 is another constant depending on the anion.

Figures 5-52 and 5-53 show plots of $\ln \gamma^-$ vs R^{-2} for fluorine and oxygen ions in various crystal structures. We consider first the ionic materials with cations that are not transition metals. In Fig. 5-52, the coordination of the fluorine ions differs considerably for these crystals. Each F^- is coordinated by six cations in the alkali halides (LiF, NaF, and KF), by four cations in the fluorites (CaF_2 , SrF_2 , BaF_2 , and CdF_2), and by seven cations in LaF_3 and CeF_3 . Nevertheless, the most significant variable seems to be the nearest-neighbor distance. Similarly, in Fig. 5-53, the values of γ^- for ionic oxides of cations with stable, rare-gas cores (Al, Mg, Ca, and Sr) seem to fall on a line.

The numerous exceptions to this behavior in Fig. 5-53 and the point for CdF_2 in Fig. 5-52 result from three factors. BeO , SiO_2 , and ZnO are tetrahedrally

Table 5-22. Experimental neutral formula unit hyperpolarizabilities for fluorides.

Neutral formula unit, M_XF	Anion-cation separation (Å)	Hyperpolarizability, $\gamma(M_XF)$ (10^{-39} esu)
LiF	2.01	141
NaF	2.32	320
KF	2.67	889
$Mg_{1/2}F$	1.99	128
$Ca_{1/2}F$	2.37	282
$Sr_{1/2}F$	2.51	412
$Cd_{1/2}F$	2.33	1520
$Ba_{1/2}F$	2.69	626
$La_{1/3}F$	2.50	487
$Ce_{1/3}F$	2.48	440

Table 5-23. Experimental neutral formula unit hyperpolarizabilities for oxides and sulfides.

Neutral formula unit M_XO , M_XS	Anion-cation separation (Å)	Hyperpolarizability, $\gamma(M_XO, M_XS)$ (10^{-39} esu)
BeO	1.65	436 ^a
MgO	2.11	485
CaO	2.41	1500
ZnO	1.98	4850
SrO	2.58	2290
$Al_{2/3}O$	1.91	248
$Sc_{2/3}O$	2.11	920 ^b
$Ga_{2/3}O$	2.00	887
$Y_{2/3}O$	2.27	945
$Er_{2/3}O$	2.26	1040
$Si_{1/2}O$ (fused)	1.61	510
$Ti_{1/2}O$	1.96	2520
$Zr_{1/2}O$	2.26	650
ZnS	2.34	8420
CdS	2.53	57000

^a Obtained from $BeAl_2O_4$ and Al_2O_3 assuming additivity.

^b Obtained from the measured n_2 of $Gd_3Sc_2Al_3O_{12}$ by subtraction of the contributions from $Gd_{2/3}O$ and $Al_{2/3}O$.

bonded materials in which the bonding is largely covalent, and they would be not be expected to follow the predictions of Eq. (49), which is based on an ionic model. Cations such as Ti^{4+} and Sc^{3+} have low-lying unfilled d orbitals, which greatly increase the number and accessibility of conduction-band states that can be occupied by the valence electrons of the anion in response to the optical field, thereby greatly increasing the effective α^- and γ^- . On the other hand, cations such as Zn, Cd, and Ga have filled outer d shells that lie close in energy to the valence orbitals of the anions in an ionic solid; these cations therefore have a large hyperpolarizability of their own. The cations with filled outer d shells are, apparently, exceptions to the usual situation of negligibly small cation hyperpolarizabilities.

We have also compared the values of γ^- for alkali halide crystals with recent self-consistent-field (SCF) calculations using the local density approximation (LDA). Table 5-24 gives calculated values of α^+ , α^- , γ^+ , and γ^- , and calculated and measured values of $\gamma^+ + \gamma^-$. Although the linear polarizabilities of the cations are generally somewhat smaller than those of the anions of similar atomic number, the hyperpolarizabilities of the cations are negligible compared with those of the anions. This behavior can be explained using expressions for

Fig. 5-52. Measured hyperpolarizability per fluorine ion vs R^{-2} for some binary fluorides with various crystal structures. The $Cd_{1/2}F$ was not included in the fit to Eq. (49).

Fig. 5-53. Measured hyperpolarizability per oxygen ion vs R^{-2} for some binary oxides with various crystal structures. The points shown as square symbols were not included in the fit to Eq. (49).

Table 5-24. Measured and theoretical hyperpolarizabilities of alkali halides.

Sample	Polarizability ($\text{\AA}^3$)		Hyperpolarizability (10^{-39} esu)		Comparison with experiment at $1.06 \mu\text{m}^a$	
	α^+	α^-	γ^+	γ^-	γ_{theor}	γ_{expt}
LiF	0.032	0.848	0.17	240	257	141
NaF	0.158	1.13	3.61	518	563	320
KF	0.839	1.28	62.4	780	906	889
RbF	1.39	1.38	174.0	1014	—	—
LiCl	0.032	2.81	0.17	1210	—	—
NaCl	0.158	3.26	3.55	2030	2380	1630
KCl	0.838	3.50	62.4	2750	3280	3050
RbCl	1.39	3.68	174.0	3444	—	—
LiBr	0.032	3.86	0.17	2030	—	—
NaBr	0.158	4.40	3.55	2750	4000	3370
KBr	0.838	4.66	61.8	4190	5240	4930
RbBr	1.39	4.89	172.0	5220	—	—
LiI	0.032	5.67	0.17	3780	—	—
NaI	0.159	6.37	3.55	5790	—	—
KI	0.838	6.68	61.2	7260	—	—
RbI	1.38	6.95	170.0	8820	—	—

^a Theoretical values obtained from Ref. 125 and extrapolated to $\lambda = 1.06 \mu\text{m}$ using the formula of Ref. 125.

Table 5-25. Nonlinear indexes calculated assuming additivity of hyperpolarizabilities (Table 5-23).

Sample	n (1.06 μm)	n_2 (10^{-13} esu)	
		Measured	Calculated ^a
BeAl ₂ O ₄	1.73	1.46	—
MgAl ₂ O ₄	1.72	1.50	1.3
CaMgSi ₂ O ₆	1.67	1.73	2.2
YAlO ₃	1.93	3.37	4.8
Y ₃ Al ₅ O ₁₂	1.82	2.7	3.0
Gd ₃ Sc ₂ Al ₃ O ₁₂	1.89	4.0	—
Gd ₃ Sc ₂ Ga ₃ O ₁₂	1.94	5.5	5.7
Gd ₃ Ga ₅ O ₁₂	1.94	5.8	5.2
Y ₃ Ga ₅ O ₁₂	1.91	5.2	5.5
La ₃ Lu ₂ Ga ₃ O ₁₂	1.93	5.8	5.4
SrTiO ₃	2.31	26.7	30.0

^a Calculated from simple oxides.

α and γ obtained from perturbation theory.¹²³ It is found that α depends on the second moment of the ionic radial charge density distribution, whereas γ also depends on the fourth moment, which is much more sensitive to the larger radial extent of the anions compared with the cations.

The effects of increasing cation size on anion hyperpolarizabilities are clearly evident in Table 5-24; for example, the calculated γ^- for fluorine increases by a factor of 4.22 when the coordinating cation changes from Li to Rb. The measured changes in $\gamma^+ + \gamma^-$ are even larger; the values for LiF and KF differ by a factor of 7. Nevertheless, the calculated values of γ^- are in quite reasonable agreement with the measured values, considering the difficulty of the calculations and the lack of absolute calibration of the experimental values. The greatest differences between theory and experiment occur for the lighter cations, Li and Na, which are most difficult to treat in SCF/LDA calculations.

We have investigated whether the values of γ^- obtained from measurements on simple binary compounds can be used to calculate n_2 for different crystal structures or for more complex crystals containing several cation species. We used the values of γ^- in Table 5-23 to calculate n_2 for several complex oxides using Eq. (47), assuming that f is given by the Lorentz local field correction $f = (n^2 + 2)/3$. Note that the γ^- for Gd_{2/3}O and Lu_{2/3}O were assumed to be the same as for Er_{2/3}O.

The measured and calculated values of n_2 in Table 5-25 agree to within an average absolute error of 13%. This agreement is significant, because the coordination of oxygen in many of the compounds in Table 5-25 is very complex. For example, in the garnets each O²⁻ ion has as nearest neighbors two rare-earth ions in dodecahedral coordination, one transition-metal ion in sixfold coordination, and another transition-metal ion in tetrahedral coordination. This suggests that the pair hyperpolarizabilities in Tables 5-22 and 5-23 are simply additive, although a larger number of oxides and halides must be studied to thoroughly investigate this property.

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Effect of Excited-State Absorption on the Performance of Cr³⁺ Vibronic Lasers

Most vibronic lasers do not operate at their full theoretical efficiency. There are many possible sources of loss in a laser material, including passive absorption, low emission quantum yield, and pump-induced transient absorbers. Despite the general difficulty of identifying the loss mechanism responsible for low efficiency in any particular system, we have determined that the ESA of Cr³⁺ probably represents a non-negligible loss mechanism in all Cr³⁺ lasers.

We find that the measured efficiency η_{exp} is lower than the predicted value by the factor $1 - \sigma_{\text{ESA}}/\sigma_{\text{em}}$, where σ_{ESA} and σ_{em} are the ESA and emission cross sections, so it is important to determine the ESA spectrum of Cr³⁺ in several crystal hosts near the lasing wavelengths. It is also useful to obtain results in other spectral regions to gain a fundamental understanding of the nature of the ESA bands. Our goal is to establish criteria with which potential Cr³⁺ hosts can be selected so as to minimize the ESA cross section at the laser wavelengths.

We measured excited-state absorption spectra with the classic pump-probe arrangement, in which the transmission of

Fig. 5-54. (a) ESA and ground-state bleaching spectra of $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$. (b) Conventional absorption and emission data.

a probe beam is measured in the presence and absence of a pump pulse. A flashlamp-pumped dye laser served as the pump source, and a cw tungsten-halogen lamp served as the probe source. The resulting difference spectra display positive features for pump-induced ESA bands and negative dips representing ground-state bleaching effects.

Figure 5-54 shows pumped ground- and excited-state spectra for $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ at 295 K, and conventional (unpumped) absorption and emission spectra. The absorption spectra have long been understood on the basis of the Tanabe-Sugano (TS) diagram¹²⁸ for the octahedrally coordinated Cr^{3+} ion (Fig. 5-55). The three broad features in the absorption spectrum, from lower to higher energy, correspond

Fig. 5-55. Tanabe-Sugano diagram for octahedral Cr^{3+} ($C/B = 4.50$). The values $Dq = 1640 \text{ cm}^{-1}$ and $B = 745 \text{ cm}^{-1}$ were used to fit the positions of the absorption peaks in Fig. 5-54 (solid vertical line). Under the assumption that the Stokes shift of the emission band is entirely due to the a_{1g} displacement mode, $Dq/B = 1.8$ fits the energy of the relaxed excited state (dashed vertical line).

to the transitions ${}^4A_2 \rightarrow {}^4T_2$, 4T_1a , and 4T_1b , respectively. The value $Dq/B = 2.2$ corresponding to the solid vertical line of Fig. 5-55 fits the absorption spectra well. Here Dq is the crystal field splitting parameter, which is inversely related to the Cr-F separation; the Racah parameter B is used to normalize the value of Dq . The 2E and 2T_1 states are observed, because of interference effects, as sharp dips superimposed on the broad bands.

Following excitation into any of the absorption bands, the energy accumulates in the lowest excited level, the 4T_2 state, shown as a filled circle in Fig. 5-55. The relaxation of the neighboring anions with respect to their positions in the ground state is described by the difference in the Dq/B values for the ground and excited states. The experimental manifestation of this lattice relaxation is the Stokes shift between the absorption and emission bands shown in Fig. 5-54. Once the system is in the 4T_2 state, one of three processes

may occur at the Cr^{3+} ion: radiative decay to the ground state, nonradiative decay, or excited-state absorption. We have verified in separate experiments that nonradiative decay is not important.¹²⁹

Figure 5-54(a) shows the change in absorption induced by optically pumping the $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ sample. The negative response results from bleaching of the ground-state absorption feature. The ESA band at 510 nm ($19\,600 \text{ cm}^{-1}$) is due to the ${}^4T_2 \rightarrow {}^4T_1b$ transition. The energy of the ESA band is predicted to be near $18\,000 \text{ cm}^{-1}$ on the basis of fitting the positions of the conventional absorption and emission spectra to the TS diagram; this value is in reasonable agreement with the observed position.

We now discuss the ESA band observed in the IR. The filled circles in Fig. 5-54(a) are the results of a pump-probe experiment. The scan was truncated at 1000 nm because the emission output from the sample became brighter than the probe source below this wavelength. The square points are the inferred values of the ESA cross section obtained by Caird et al.¹²⁹ In those experiments, the gain of the $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ crystal was compared with that expected in the absence of ESA. Caird et al. attributed the difference between the measured and calculated gains to the influence of ESA. In Fig. 5-54(a) these two sets of data have been reconciled by drawing a single Gaussian curve through them, with a peak energy of $11\,800 \text{ cm}^{-1}$ and a FWHM of 4000 cm^{-1} .

The observed IR ESA band is in striking contrast to the expected character determined from the TS diagram of Fig. 5-55, which predicts that the ${}^4T_2 \rightarrow {}^4T_1a$ band is a sharp line at about 7000 cm^{-1} . (This prediction arises because the slopes of the 4T_2 and 4T_1a curves in Fig. 5-55 are nearly identical.) The striking disagreement between the observed and predicted spectra indicates that a crucial aspect of the physics has been neglected in the TS diagram. Before considering the physical nature of this transition in detail, we first provide proof that this broad IR ESA band exists in all low-field d^3 transition-metal systems (Cr^{3+} and V^{2+}).

Figure 5-56 shows the ground- and excited-state spectra for Cr^{3+} in BeAl_2O_4 (alexandrite). The spectral properties of

Fig. 5-56. (a) ESA and ground-state bleaching spectra of $\text{BeAl}_2\text{O}_4:\text{Cr}^{3+}$. (b) Conventional absorption and emission data.

alexandrite differ somewhat from those of $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ because the ^2E level is the lowest excited state for alexandrite. The ^2E population is responsible for the sharp line emission and for the intense ESA feature between 1300 and 1700 nm. The broad $^4\text{T}_2 \rightarrow ^4\text{T}_1\text{a}$ transition is nevertheless observed at about 1000 nm. This band was recognized by extrapolating from the inferred ESA values of Shand

et al.¹³⁰ to our pump-probe data. The important point here is that the broad IR ESA band occurs at about the same energy as in the $\text{Na}_3\text{Ga}_2\text{Li}_3\text{F}_{12}:\text{Cr}^{3+}$ system. It is also worth noting that Andrews et al.¹³¹ found an analogous ESA band for Cr^{3+} in $\text{Gd}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$, and that we have shown¹³² that ESA is the important loss mechanism in the $\text{KMgF}_3:\text{V}^{2+}$ and $\text{MgF}_2:\text{V}^{2+}$ lasers.

The key to understanding the ESA is to determine the source of the broadening and shift of the ${}^4T_2 \rightarrow {}^4T_1a$ transition relative to the TS-diagram predictions. It is important to note that only the totally symmetric a_{1g} distortion is taken into account in the TS diagram. We have found, however, that the distortion of e_g symmetry is the main source of the broadening for the ${}^4T_2 \rightarrow {}^4T_1a$ transition. Because of a Jahn-Teller vibronic interaction, the most stable configuration for the 4T_2 state has an $e_g\theta$ distortion, which means that four of the fluorine ions (in a single plane) move outward, and the other two fluorines move inward, as shown in Fig. 5-57(a). Using group-theoretical arguments and the strong-field wave functions of the 4T_2 and 4T_1a states, it can be shown that the 4T_1a state will in fact suffer this same type of distortion, although of opposite sign; two fluorines move outward and four inward. The important point here is that the 4T_2 and 4T_1a states will have different equilibrium

configurations, and that transitions between these two states will therefore be observed as a broad band, rather than as a sharp feature as predicted by the TS diagram. Figure 5-57(b) shows the effect of the Jahn-Teller stabilization energy E_{JT} in the e_g mode Q_θ . The transition labeled 2 is expected to give rise to a broad absorption band.

We turn now to developing criteria for selecting host crystals for Cr^{3+} in which the ESA at the laser wavelengths can be minimized. The distortion in the 4T states is stabilized by the Jahn-Teller energy¹³³

$$E_{JT} = \frac{A^2}{2k}, \quad (50)$$

where A is the linear electron-phonon coupling constant and k is the harmonic force constant of the host. Little is known about the systematic variation of A with crystal host, so it is difficult to assess its dependence on the properties of the host. It is clear, however, that if k is maximized the host lattice will, in effect, resist the tendency of the CrF_6 cluster to undergo Jahn-Teller distortion. In this way, the ESA band should be shifted to lower energy, where it will interfere less with the emission process. Small sites in stiff crystal lattices will be best suited for this purpose.

The TS diagram of Fig. 5-55 indicates that the IR ESA band is not sensitive to increasing values of Dq/B , while the emission peak energy varies linearly with Dq/B . It is therefore best to select a host in which the ${}^4T_2 \rightarrow {}^4A_2$ emission is at the highest possible energy consistent with a predominance of 4T_2 (rather than 2E) population in the excited state. We therefore desire a host with a small site to shift the emission to shorter wavelength, and a rigid lattice structure so as to shift the ESA band to longer wavelength.

References 129 and 131 give a more complete discussion of the effects of ESA in Cr^{3+} lasers.

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Fig. 5-57. (a) Nearest neighbor displacements in the $e_g\theta$ distortion. (b) Cross section of the adiabatic potential energy surface in the Q_θ coordinate. Labels ξ , η , ζ , etc., refer to orbital components of the states.

Theoretical Studies of Optical Properties of $3d^3$ Ions in Fluoride Crystals

Tunable lasers based on divalent vanadium substituted into MgF_2 and $KMgF_3$ have been considered for use as high-average-power laser drivers for fusion applications.¹³⁴ In an extensive experimental study of the laser performance of $MgF_2:V^{2+}$ and $KMgF_3:V^{2+}$, Moulton, Fahey, and Krupke^{135,136} attributed the low gain to ESA. Recently Payne, Chase, and Wilke¹³⁷ measured the ESA in both systems and observed a broad band, which they attributed to the ${}^4T_{2g} - {}^4T_{1ga}$ transition and which significantly overlapped the emission spectrum in both cases. They explained the positions and widths of the ESA transitions on the basis of the non-totally symmetric (NTS) distortions of the lattice ions in the excited states. To quantify their model and to gain new insight into impurity-lattice interactions and their role in ESA, we have carried out *ab initio* electronic structure calculations of the potential energy curves for the NTS distortions in $MgF_2:V^{2+}$ and $KMgF_3:V^{2+}$.

We determined the potential energy curves for the a_g , e_g , and t_{2g} distortions of the near-neighbor F^- ions for each ion/host system. For cubic $KMgF_3:V^{2+}$, the e_g and t_{2g} modes lead to Jahn-Teller electron-phonon coupling in the excited states. This results in tetragonal distortions in the excited states, which must be included to estimate the ESA accurately. The MgF_2 host has rutile symmetry and therefore no Jahn-Teller effect, but the lower site symmetry of the V^{2+} leads to tetragonal splitting in the excited states and therefore to similar ESA bands. We found that the t_{2g} distortion was not important in $MgF_2:V^{2+}$, but that it made a significant contribution to the ESA wavelength for $KMgF_3:V^{2+}$.

Figure 5-58 shows the symmetric-stretch potential energy curves for the quartet states of $KMgF_3:V^{2+}$ and the vertical excitation energies relative to the minimum of the ${}^4A_{2g}$ ground state. The calculated excitation energies are in good agreement with low-temperature absorption spectra measured by Sturge.¹³⁸ Figure 5-59 shows the potential energy curves for the e_g

Fig. 5-58. Symmetric stretch curves for $KMgF_3:V^{2+}$.

Fig. 5-59. Potential energy curves for the e_g distortion.

Fig. 5-60. Potential energy curves for the t_{2g} distortion.

distortion of the ${}^4T_{2g}$ and ${}^4T_{1ga}$ excited states. As indicated, the ${}^4T_{2g} \rightarrow {}^4T_{1ga}$ ESA energy for cubic symmetry ($Q = 0$) is 7022 cm^{-1} . When the ESA energy is calculated from the minimum of the ${}^4T_{2g}$ potential energy curve, however, it increases by 593 cm^{-1} to 7615 cm^{-1} . Figure 5-60 gives the potential energy curves for the t_{2g} distortion of the ${}^4T_{2g}$ and ${}^4T_{1ga}$ states. The t_{2g} NTS distortion increases the ESA by 300 cm^{-1} over the value obtained assuming cubic symmetry.

Including the contributions from both the e_g and t_{2g} NTS distortions increases the ESA energy to 7915 cm^{-1} (1260 nm). Corrections estimated from the errors in the calculated zero-phonon line of the ${}^4T_{2g}$ state ($\sim 150 \text{ cm}^{-1}$) and the vertical ground state excitation energy of the ${}^4T_{1ga}$ state ($\sim 50 \text{ cm}^{-1}$) lead to our best value of 8015 cm^{-1} (1250 nm). This is in good agreement with the measurements of Payne et al.¹³⁷ on cubic $\text{KMgF}_3:\text{V}^{2+}$.

Using our calculated potential energy curves^{139,140} for the three independent local modes associated with the near neighbors of a V^{2+} impurity in MgF_2 , we calculated that the vertical absorption energies for the ESA transitions from the ${}^4T_{2g}(\text{B}_{2g})$ state to the crystal-field-split components of the ${}^4T_{1ga}$ state are 6682, 6984, and 8535 cm^{-1} . After correcting these excitation energies to account for the errors found by comparing the

calculated and observed ground state absorption, we obtain frequencies for transitions to the ${}^4T_{1ga}(\text{B}_{3g}, \text{B}_{1g}, \text{B}_{2g})$ components of 5952, 6250, and 8000 cm^{-1} . The predicted vertical ${}^4T_{2g}(\text{B}_{2g}) \rightarrow {}^4T_{1ga}(\text{B}_{2g})$ ESA transition at 1250 nm is identical to the last data point of Payne et al.,¹³⁷ which they estimate to be near the band peak. The calculated ${}^4T_{2g}(\text{B}_{2g}) \rightarrow {}^4T_{1ga}(\text{B}_{1g}, \text{B}_{3g})$ ESA wavelengths at 1600 and 1680 nm may be contributing to the absorption seen¹³⁷ in the long-wavelength region when the light is polarized parallel to the c axis. To obtain a more accurate comparison with the measured ESA bands, we are carrying out a Franck-Condon analysis of the vibronic energy levels of $\text{MgF}_2:\text{V}^{2+}$ and $\text{KMgF}_3:\text{V}^{2+}$, similar to that carried out for the ground state absorption.^{139,140}

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Excited-State Absorption of the Sm^{2+} Impurity

$\text{SrF}_2:\text{Sm}^{2+}$ was first demonstrated as a laser material by Sorokin and coworkers in 1962.¹⁴¹ It is still not clear, however, whether the Sm^{2+} laser can operate efficiently, because there has been little work in this area. Nevertheless, $\text{SrF}_2:\text{Sm}^{2+}$ has been considered as a possible fusion laser candidate at LLNL.¹⁴² To shed light on the processes that may fundamentally limit the efficiency of this laser, we measured the ESA spectra of $\text{SrF}_2:\text{Sm}^{2+}$.

After optical pumping in the allowed $4f \rightarrow 5d$ absorption bands of $\text{SrF}_2:\text{Sm}^{2+}$, the Sm^{2+} ion relaxes to the lowest excited state, ${}^5D_0(4f^6)$. While the 7F_0 ground state and the 5D_0 excited state both have the $4f^6$ electronic configuration and $J = 0$, they are of different spin, having $S = 3$ and $S = 2$, respectively. The spin selection rule dictates that the highest oscillator strength will be for transitions from the 5D_0 and 7F_0 states to the $4f^55d$ states of the same spin. The spin of the $5d$ electron can be oriented either parallel or antiparallel to the total spin of the $4f^5$ electron core, resulting in $4f^55d$ states

with total spins of $S = 3$ and $S = 2$, respectively. The energies of the ground- and excited-state absorption bands can therefore provide a direct measure of the difference in energy between the $S = 2$ and $S = 3$ states of the $4f^55d$ configuration, which is determined by the $4f$ - $5d$ exchange interaction.

Figure 5-61 shows the ground- and excited-state absorption spectra of $\text{SrF}_2:\text{Sm}^{2+}$. The ESA spectra are shifted by the energy of the ${}^7F_0 \rightarrow {}^5D_0$ transition ($14\,616\text{ cm}^{-1}$) to facilitate comparison with the ground-state spectrum (both spectra are plotted relative to the energy of the 7F_0 ground state). The $S = 2$ components (indicated by offset arrows) appear in the ESA spectrum and are shifted by about 3000 cm^{-1} from the $S = 3$ states, as observed in the ground-state spectrum.

Yanase¹⁴³ has developed an approximate, analytical description of the energy levels for the $4f^55d$ configuration of the Sm^{2+} impurity. Following Yanase, we write the Hamiltonian as

$$H = H_{ff} + H_{df} + V_f + V_d, \quad (51)$$

where H_{ff} involves the Coulomb and exchange interactions between the $4f$ electrons and H_{df} involves the interactions between the $5d$ and $4f$ electrons. The terms V_f and V_d are responsible for the crystal-field splitting of the $4f$ and $5d$ electrons, respectively. The spin-orbit terms are omitted from Eq. (51) for simplicity.

The $4f^55d$ energy levels of Sm^{2+} can be analyzed by considering the terms of Eq. (51) in order of increasing importance. The H_{ff} term will produce the $2S+1L$ states of the $4f^5$ core; the two lowest of these are 6H and 6F . The crystal field terms in Eq. (51) are known to be much larger for the $5d$ electron than for the $4f$ electron (that is, $V_d \gg V_f$); we therefore take V_f to be zero. The effect of V_d is to split the $5d$ orbital into e_g - and t_{2g} -type components, with the e_g components lying lower. Four states result from combining the $5d$ electron with the 6H or 6F core states. In order of increasing energy, they are $|{}^6H, e_g\rangle$, $|{}^6F, e_g\rangle$, $|{}^6H, t_{2g}\rangle$, and $|{}^6F, t_{2g}\rangle$.

The exchange terms in H_{df} split each of these states into an $S = 3$ and an $S = 2$ component. As noted earlier (and as shown in Fig. 5-61), the $S = 3$ and $S = 2$ states are observed in the ground and excited-state absorption spectra, respectively. These energy levels are represented in Fig. 5-62 as well. Reference 144 describes the spectroscopy and energy levels of Sm^{2+} in detail.

It is known¹⁴¹ that $\text{SrF}_2:\text{Sm}^{2+}$ lases on the ${}^5D_0 \rightarrow {}^7F_1$ transition (shown in Fig. 5-62) at $14\,350\text{ cm}^{-1}$. The absolute efficiency of this laser is determined, in part, by the relative values of the emission and ESA cross sections σ_{em} and σ_{ESA} , since the net effective cross section is given by

$$\sigma_{eff} = \sigma_{em} - \sigma_{ESA}. \quad (52)$$

Fig. 5-61. Excited- and ground-state absorption spectra of $\text{SrF}_2:\text{Sm}^{2+}$ at 20 K. The ESA spectra are shifted to facilitate comparison with the ground-state spectrum.

Fig. 5-62. Ground- and excited-state energy levels and transitions of Sm^{2+} in SrF_2 . The $5D_0 - 7F_1$ laser transition is shown.

Assuming that the absorption cross section of $\text{SrF}_2:\text{Sm}^{2+}$ is the same as for CaF_2 (Ref. 145), we use the data of Fig. 5-61 to estimate the ESA cross section at the laser wavelength to be $7 \times 10^{-20} \text{ cm}^2$; we calculate the emission cross section to be $(8 \pm 4) \times 10^{-20} \text{ cm}^2$. Although these values are clearly estimates, the fact that they are comparable indicates that ESA is likely to represent a severe detriment for the $\text{SrF}_2:\text{Sm}^{2+}$ laser.¹⁴⁴

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Optical Properties of Cr^{3+} in Scandium Sites of Crystals

A great deal of research is being carried out to develop new solid state lasers based on the Cr^{3+} impurity. The spectral positions of the Cr^{3+} absorption and emission bands are of central importance to this work, because these features

determine the pumping and lasing wavelengths of prospective new materials. While the overall performance of a Cr^{3+} solid state laser will depend on a multitude of material properties, the peak wavelengths of the absorption and emission bands are the most salient characteristics that must be initially considered. Despite their obvious importance, no clear predictive understanding of these Cr^{3+} properties exists at this time. In this article we show that the effects of lattice relaxation at the Cr^{3+} site are crucial to understanding these properties.

The simplest understanding of the optical properties of transition-metal ions is based on the crystal field (CF) or point-charge model. As an illustration, we apply this model to the K_2NaCrF_6 system, in which Cr^{3+} is surrounded by six fluoride ions in a perfectly octahedral arrangement. According to CF theory, the lowest absorption band occurs at the energy $10 Dq$. We concentrate on understanding this band, because the positions of all the other absorption and emission bands are correlated with it.

The CF theory of octahedral transition-metal ion complexes gives for Dq the expression

$$Dq = \frac{kZe^2 \langle r^4 \rangle}{6R^5}, \quad (53)$$

where $k = 9 \times 10^9 \text{ N m}^2 \text{ C}^{-2}$, Ze is the anion charge, r is the radial electronic coordinate, and R is the Cr-anion separation. For K_2NaCrF_6 , $R = 1.93 \text{ \AA}$, and we take $Z = 1$ for the fluoride ions. The free-ion wave functions¹⁴⁶ of Cr^{3+} give $\langle r^4 \rangle = 1.29 \text{ \AA}^4$. Equation (53) then gives $Dq_{\text{calc}} = 930 \text{ cm}^{-1}$; the lowest absorption band of K_2NaCrF_6 occurs at 16100 cm^{-1} , so $Dq_{\text{obs}} = 1610 \text{ cm}^{-1}$. The difference between these two values of Dq is substantial, but given the great simplicity of the point charge model it is clear that the crucial physical mechanisms are accounted for. Perhaps the most important aspect of Eq. (53) is the strong (R^{-5}) dependence of Dq on the Cr-F separation.

Table 5-26 gives Dq_{obs} for the elpasolites K_2NaCrF_6 and $\text{K}_2\text{NaScF}_6:\text{Cr}^{3+}$. The Cr-F distance is known to be $1.93 \text{ \AA}$ for

K_2NaCrF_6 . If it is assumed that, when Cr^{3+} substitutes for the Sc^{3+} ion, the Cr-F distance remains fixed at the Sc-F distance of 1.99 Å, the predicted Dq will be $1610\text{ cm}^{-1} \times (1.93/1.99)^5 = 1380\text{ cm}^{-1}$. [This result follows from the observation, from Eq. (53), that these materials differ only in the magnitude of R .] The $K_2NaScF_6:Cr^{3+}$ value $Dq_{obs} = 1560\text{ cm}^{-1}$ differs substantially from this prediction.

The only reasonable way to explain this discrepancy is to assume that there is substantial lattice relaxation at the Cr^{3+} site. That is, we assume that the six fluoride ions surrounding the Cr^{3+} ion in K_2NaScF_6 move inward from the original Sc-F distance in such a way that the Cr-F distances of the CrF_6 cluster in K_2NaCrF_6 and in K_2NaScF_6 turn out to be very similar.

The same situation arises for CrF_3 and $ScF_3:Cr^{3+}$. From Table 5-26 we note that CrF_3 and $ScF_3:Cr^{3+}$ have similar values of Dq, and we therefore suggest that the Cr-F distance in $ScF_3:Cr^{3+}$ is probably closer to the CrF_3 value of 1.90 Å than to the Sc-F value of 2.01 Å. The lattice relaxation seems to be a crucial factor that must be considered.

It is interesting to observe in Table 5-26 that CrF_3 has a lower Dq than K_2NaCrF_6 , despite the expectation (based on the similar Cr-F distances) that the value should be the same or slightly higher. An analogous situation exists with respect to the $K_2NaScF_6:Cr^{3+}$ and $ScF_3:Cr^{3+}$ crystals. At least for fluoride hosts, it appears that the crystal structure (here, elpasolite vs VF_3 -type) is more important in determining Dq than the size of the substitutional site in which Cr^{3+} is placed.

Table 5-27 lists Dq_{obs} for a number of Cr^{3+} -doped oxides that have substitutional Sc sites. Although each of these systems involves a Sc site, the Dq values vary substantially. The purpose of this listing is to show that the Dq values of oxides are very sensitive to the second nearest neighbors of chromium. This is true because the charge distribution and bonding of O^{2-} are more strongly influenced by the neighboring cations and by

Table 5-26. Observed values of Dq and R for Sc-containing crystals and pure Cr^{3+} compounds at 300 K.

Crystal	Dq (cm^{-1})	Metal-anion distance, R (Å)
K_2NaCrF_6	1610	1.93
$K_2NaScF_6:Cr^{3+}$	1560	1.99
CrF_3	1460	1.90
$ScF_3:Cr^{3+}$	1410	2.01

Table 5-27. Observed values of Dq for Sc-containing oxide hosts doped with Cr^{3+} at 300 K.

Crystal	Dq (cm^{-1})
Sc_2O_3	1520
$Gd_3Sc_2Ga_3O_{12}$	1563
$Gd_3Sc_2Al_3O_{12}$	1587
$Y_3Sc_2Ga_3O_{12}$	1613

the lattice structure than is the case for fluorides.

Reference 147 gives our spectroscopic data for $ScF_3:Cr^{3+}$ and $Sc_2O_3:Cr^{3+}$ and a fuller discussion of our results.

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Off-Center Distortion of Cu^{+} in Alkali Halides

When an impurity metal ion is incorporated into a host crystal, it often substitutionally replaces a metal ion of the host lattice. In this situation, the positions of the nearest-neighbor anions surrounding the impurity are often assumed to be similar to those expected for the pure, unperturbed host material. This approximation is often sufficient to permit the derivation of an energy level diagram from which an adequate understanding of the spectral and dynamical properties can be developed. The case of Cu^{+} in the alkali halides¹⁴⁸ serves to demonstrate the importance of taking into account the modified nearest-neighbor positions in interpreting the absorption spectra.

The transitions of interest are indicated in Fig. 5-63, which shows the $d^{10}1S$ and d^9s1D levels of free Cu^{+} . We take the $NaCl:Cu^{+}$ system as an example; we

Fig. 5-63. Energy levels of Cu^+ as a free ion and as an impurity. The ${}^1A_{1g} \rightarrow {}^1E_g$ and ${}^1A_{1g} \rightarrow {}^1T_{2g}$ transition rates are designated R_E and R_T , respectively.

Fig. 5-64. Cu^+ off-center distortion in an alkali halide host. Three halide ions move ΔR outwards with respect to the Cu^+ ion; the three others move ΔR inward.

expect that Cu^+ will substitute for Na^+ in this crystal and will therefore be surrounded by six Cl^- ions. The $d^{10} 1S$ ground state remains nondegenerate and becomes designated ${}^1A_{1g}$ in the O_h point group. The $d^9 s 1D$ excited state is split by the interaction of the octahedral crystal field with the d hole of the electronic configuration. As a result, the $d^{10} 1A_{1g} \rightarrow d^9 s 1E_g$ and $d^{10} 1A_{1g} \rightarrow d^9 s 1T_{2g}$ transitions, shown in Fig. 5-63, should be observed in the crystal. Their respective transition rates are designated R_E and R_T .

Numerous types of experiments make it clear that Cu^+ has a propensity to move off-center with respect to the octahedron of halide ions. The distortion occurs in such a way that Cu^+ can be considered to be displaced along the $[111]$ crystal axis, which corresponds to

the c_3 axis of the octahedron, as shown in Fig. 5-64. For mathematical simplicity, we treat the Cu^+ motion as the displacement of three halides toward the Cu^+ by an amount ΔR , and the displacement of the other three halides a distance ΔR away from the Cu^+ .

While the major influence of the environment on the in-crystal energy levels of Cu^+ arises from the sixfold coordination at the impurity site, the $d \rightarrow s$ transition strengths depend sensitively on the detailed positions of the ions. This is so because the $d \rightarrow s$ transition is parity forbidden, and can become allowed only through motions that result in the loss of the center of inversion of the CuX_6 octahedron. For small excursions of the Cu^+ from the center of the octahedron, the ${}^1A_{1g} \rightarrow {}^1E_g$ and ${}^1A_{1g} \rightarrow {}^1T_{2g}$ transitions are affected equivalently, and the relative transition rates are expected to be degeneracy weighted ($R_E/R_T = 2/3$). For large distortions, in which the Cu^+ is far off-center, the ${}^1A_{1g} \rightarrow {}^1E_g$ transition is expected to become much stronger. This is essentially because the e_g hole of the 1E_g state is directed along the Cu-X axes, and is strongly mixed with the p orbitals of the halide ions, while the t_{2g} hole of the ${}^1T_{2g}$ state points between the halides and does not mix nearly as much.

Using the well-known techniques of crystal field theory, it can be shown that, for moderate off-center displacements,¹⁴⁸

$$\frac{R_E}{R_T} = \frac{|\bar{2} + \sqrt{3}\beta(1 + 3\Delta^2)|^2}{|\bar{3} - \sqrt{2}\beta(1 + 3\Delta^2)|^2}, \quad (54a)$$

where $\Delta = \Delta R/R$ is the fractional displacement of the halide ions (see Fig. 5-64), and

$$\beta = \frac{6}{7} \left(\frac{3}{2} \right)^{1/2} \frac{\langle 3d r^4 4s \rangle}{R^2 \langle 3d r^2 4s \rangle}; \quad (54b)$$

here r is the electronic radial coordinate. The important result here is that, since we expect β to be positive, the relative strengths of the ${}^1A_{1g} \rightarrow {}^1E_g$ and ${}^1A_{1g} \rightarrow {}^1T_{2g}$ transitions will reverse as the off-center displacement of Cu^+ becomes substantial ($\Delta, \beta > 0$). As is often the case, however, crystal field theory underscores

the important physical mechanisms, but does not provide a truly quantitative result.

Figure 5-65 shows the near-UV spectra of Cu^{2+} in NaCl and KCl. The relative transition strengths are reversed in the two systems; in agreement with this observation, it has long been recognized that Cu^{2+} is on-center in NaCl and off-center in KCl. Thus it appears that the absorption (or, equivalently, the excitation) spectra provide a direct measure of the ground state position of the Cu^{2+} ion.

The value of R_0/R_1 can be determined from the absorption spectrum for many Cu^{2+} -doped alkali halides. In Fig. 5-66 the logarithm of observed R_0/R_1 values is plotted vs nearest-neighbor distance for a variety of alkali halides. A greater nearest neighbor distance implies a higher R_0/R_1 , which in turn indicates greater off-center displacement of the Cu^{2+} ion. The nearest-neighbor distance provides an overall measure of several contributions to the magnitude of the displacement of the Cu^{2+} ion, including the radius of the cation and the polarizability of the halide ion. A larger cation radius implies more room for off-center displacement of the substitutional Cu^{2+} ion, while a greater polarizability of the halide ion is known to stabilize the off-center position because of the short range of the monopole-induced dipole interaction.

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Laser-Induced Emission of Neutrals from Optical Materials and its Relation to Optical Surface Damage

Introduction. The fundamental problems involved in understanding and controlling optical surface damage are to identify the mechanism for deposition of laser energy at the surface and the resulting physical processes that precipitate catastrophic failure.

Several interaction mechanisms could account for the absorption of laser energy at the surface of a nominally transparent optical material. Direct absorption

of light by the intrinsic material can result only from multiphoton excitation across the band gap, which generates electrons and holes, or by multiphonon absorption, which directly heats the material. Both of these processes are very weak, and the energy density deposited in the near-surface region is certainly too low to cause direct thermal failure of the material. It is possible, however, that free

Fig. 5-65. 1-K excitation spectra of typical on-center ($\text{NaCl}:\text{Cu}^{2+}$) and off-center ($\text{KCl}:\text{Cu}^{2+}$) systems.¹¹⁹ Relative strengths of the ${}^1\text{A}_{1g} \rightarrow {}^1\text{E}_g$ and ${}^1\text{A}_{1g} \rightarrow {}^1\text{T}_{2g}$ transitions are reversed for the two systems.

Fig. 5-66. Logarithm of measured relative transition rates R_0/R_1 of the ${}^1\text{A}_{1g} \rightarrow {}^1\text{E}_g$ and ${}^1\text{A}_{1g} \rightarrow {}^1\text{T}_{2g}$ transitions of Cu^{2+} vs nearest-neighbor cation-anion separation in the alkali halide host. R_0/R_1 is a measure of the off-center displacement.

carriers produced by the multiphoton excitation could initiate avalanche ionization.

There are also *extrinsic* absorption processes that produce free carriers or direct heating near the surface of an optical material. For example, surface states and defects can supply "cheap" (low-excitation-energy) electrons or holes by direct absorption of the laser light, and highly absorbing particles embedded in or near the surface can cause local heating.

Detecting these surface absorption processes is a formidable problem, because they may be very localized and may cause unobservably small changes in the transmitted or reflected laser intensities and the average surface temperatures. We have investigated the possibility of observing the interaction of the laser beam with the surface by detecting neutral atoms or molecules emitted as a result of the excitation.

Several characteristics of the emitted neutrals could provide evidence for the nature of the interaction of the laser beam with the surface; these include emission yields, the identity of the emitted species, and the temporal and spatial dependence of the emission. If neutral emission is to be useful for surface-damage research, it must be established that it is caused by the same laser-surface interaction that leads to observable damage events. In addition, the emission must be detectable at fluences considerably below the threshold for significant optical damage, so that it probes the *initial* interaction of the laser beam with the surface, rather than a catastrophic breakdown of the surface, which will always result in copious neutral, ion, and electron emission.

We have investigated these issues for representative optical materials, including fused silica and single crystals of ZnS, NaF, Al_2O_3 , and BaF_2 .

Experimental Procedure. The experiments were carried out in two ultrahigh-vacuum systems with base pressures of 5×10^{-10} and 2×10^{-9} Torr, respectively. Before laser excitation, a heater on the sample holder was used to desorb contaminants from the surfaces at temperatures up to 500°C in ultrahigh vacuum. Excitation was provided by a

Q-switched YAG:Nd laser, which delivered pulses of 5 to 8 ns duration at 1.06, 0.532, 0.355, and 0.266 μm ; beam diameters were about 500 μm . To obtain fluence estimates we measured the beam diameter with a traveling knife-edge and the total pulse energy with a calorimeter. Optical damage was identified visually by increased scattering of a probe laser beam. The optical damage thresholds quoted here are therefore to be regarded only as estimates of relative thresholds for the various samples and laser wavelengths.

A quadrupole mass spectrometer was mounted 11.3 cm from the sample to detect the emitted neutrals. The response time of the detection system was limited to about 10 μs because of the transit-time dispersion in the quadrupole system. Data were obtained with increasing fluence on each new spot on the sample, starting from about one-tenth of the observed damage threshold. About ten to twenty shots were taken at each spot before damage occurred or the spot was abandoned.

The time of flight from the sample to the ionizer on the mass spectrometer is inversely proportional to the particle velocity. For a Maxwellian particle velocity distribution characterized by a temperature T , the detected signal S will thus have the time dependence¹⁵⁰

$$S = Kt^{-4} \exp\left[-(t_0/t)^2\right], \quad (55)$$

where $t_0 = (M/2kT)^{1/2}d$, M is the particle mass, d is the sample-to-detector distance, and K is a normalizing constant. This distribution has a maximum at a time delay $t_p = t_0/\sqrt{2} = d(MkT/4)^{1/2}$. For temperatures up to a few thousand kelvin, the time delay is a sensitive measure of the effective particle temperature, particularly for more massive particles. Of course, Eq. (55) is valid only if the emission occurs within a time interval smaller than t_p after the laser pulse.

Table 5-28 summarizes the samples studied, the laser wavelengths, and the general characteristics of the observed emission. Unambiguous evidence of emission of *surface constituents* at fluences below the optical damage threshold was obtained for all samples except Al_2O_3 . In all cases, of course,

desorption of surface contaminants was found, usually at a very low level, after thermal cleaning of the surface. For fused silica, ZnS, and NaF, one or more Maxwellian-shaped components dominated at low fluences. In all of these materials we observed high-energy (HE) neutrals as the fluence approached the optical damage threshold. Much of the data for fused silica were presented in the 1986 Laser Annual,¹⁵¹ so the remainder of this discussion concerns ZnS and NaF, for which the most significant new results have been obtained.

Zinc Sulfide. ZnS was selected for these studies because it is a wide-band-gap semiconductor and because it is used for optical coating; it was also selected because it has a relatively small optical band gap ($E_g = 3.9$ eV), so that measurements can be made with photon energies both above and below E_g using Nd:YAG laser wavelengths between 1.064 and 0.266 μm . In addition, we had already investigated ion and electron emission from ZnS, and had conjectured that the primary emission component must be neutrals^{151,152}; the present work confirms that hypothesis.

Figure 5-67 shows time-of-flight data for the emission of Zn from a ZnS single crystal under excitation with an 8-ns, 1.064- μm pulse. Near the threshold for observation of neutral Zn emission (about 4 J/cm^2), a gradual onset of the signal suggests that the emission occurs for several hundred microseconds, and that the signal then decreases because of pumping and adsorption on the chamber walls. Above about 8 J/cm^2 , however, a peak develops at a time delay of about 250 μs ; we attribute this peak to promptly emitted Zn particles with a kinetic temperature < 1000 K. This behavior (also observed for the emission of S and S_2) shows that very little surface heating is required for the emission to occur. As the damage threshold is approached, however, this peak shifts very rapidly to shorter time delays, indicating characteristic temperatures of several thousand kelvin. Simultaneously, a spike appears at short time delays. Even after the laser shots in the two upper traces in Fig. 5-67, there was no visible evidence of optical damage, although the apparent particle

Table 5-28. Optical materials and neutral emission characteristics under excitation at 1.064, 0.532, 0.355, and 0.266 μm .

Sample	Species emitted	Characteristics
Fused silica ^a	Si, SiO, O, O ₂ , OH	Maxwellian; HE neutrals
ZnS crystal	Zn, S, S ₂	Complex; HE neutrals
NaF crystal	Na, F	Maxwellian; cleaving, cracking; HE neutrals
Al ₂ O ₃ crystal	No predamage emission observed	—
BaF ₂ crystal	Ba, F	HE neutrals only

^a 1.064- μm excitation only.

temperature of $\sim 10^4$ K suggests that either the sample surface or the emitted gas of Zn atoms was hot enough to cause local surface modification. This spot on the sample did, in fact, suffer catastrophic damage on the next shot at 11 J/cm^2 .

Similar data were obtained at 0.532, 0.355, and 0.266 μm . As the wavelength decreases, the general characteristics of the time-of-flight distributions are unchanged, suggesting that the same processes are involved in the emission at all

Fig. 5-67. Time-of-flight data for Zn emission from ZnS following a series of 1.06- μm , 8-ns laser pulses of increasing fluence. Temperatures are estimates based on the time delay t_d of the peak of the Maxwellian emission component.

four wavelengths. On the other hand, the threshold fluences at which emission is observed (and at which optical damage occurs) gradually decrease.

We have compared the thresholds for observable neutral Zn emission with the thresholds for observed optical damage, obtained by taking shots at successively higher fluences on a single site until optical damage occurred. In Fig. 5-68, the two thresholds for each wavelength are plotted against one another. There is a rough correlation between the thresholds for wavelengths that span the region of optical transparency of ZnS. At $0.266\text{ }\mu\text{m}$, however, the emission threshold is very low (below 0.01 J/cm^2), while there is still a reasonably well-defined optical damage threshold of about 0.5 J/cm^2 .

Fig. 5-68. Threshold for observable neutral Zn emission from ZnS vs optical damage threshold for four Nd:YAG laser wavelengths (pulse durations 5 to 8 ns).

Fig. 5-69. Total Zn emission from ZnS surface for a series of $1.064\text{-}\mu\text{m}$ laser pulses of increasing fluence. ▽

A potentially important characteristic of the neutral emission is its effects on the surface composition. In general, any difference in the emission yields of the surface constituents can alter the composition. It is difficult to measure these yields precisely, however. The data for ZnS at fluences far below the damage threshold provide a more unambiguous indication of composition change. We have found¹⁵³ that the Zn emission at such low fluences is delayed relative to that of S or S_2 . This suggests that the surface composition is altered by the laser ablation, at least during the laser pulse. Figure 5-69 shows a possibly related effect: the emission observed on successive shots on a single site decreases ("bleaches") by an amount that varies with the fluence. As the fluence approaches the damage threshold the bleaching is greatly reduced, if not eliminated. This shows that active absorption sites on the surface are deactivated at various fluences. We also observed this bleaching behavior for fused silica and, to a lesser extent, for NaF.

Sodium Fluoride. The experiments were done on single crystals of NaF that were cleaved and polished. Neutral emission characteristics were similar at all four Nd laser wavelengths, except for a gradual decrease in the threshold fluence with decreasing wavelength, which was similar to the behavior observed for ZnS. The same two characteristic features, a time-of-flight peak and a background that persists for several milliseconds, found for fused silica and ZnS were also present in NaF.

Figure 5-70 shows a time-of-flight distribution of the neutral Na emission following a $0.266\text{-}\mu\text{m}$ pulse at 3.8 J/cm^2 , about half of the observed optical damage threshold. An approximately Maxwellian distribution with a maximum at $t_p = 200\text{ }\mu\text{s}$ indicates a particle temperature of about 300 K , very close to the actual sample temperature. This "cold" emission is accompanied by a peak with a very short time delay; this peak was recorded by a second transient recorder with a time resolution of $10\text{ }\mu\text{s}$. This peak is shown in the inset of Fig. 5-69, in which the $9.6\text{-}\mu\text{s}$ transit time in the mass

spectrometer has been subtracted from the time axis. Various properties of this emission show that it is not caused by detection of scattered or emitted light, or by the electron-multiplier detector of the mass spectrometer, and that only a small fraction of it is due to emitted ions.¹⁵² The very short time delay indicates neutrals of surprisingly high energy—roughly 10 to 100 eV.

The time-of-flight data for NaF shown in Fig. 5-70 suggest a likely explanation for the emission of the HE neutrals. The lower trace corresponds to a 0.266- μm laser fluence just below the optical damage threshold. The Maxwellian emission and the short-time-delay HE emission were both observed on this shot.

The upper trace represents the next shot on the same spot, at a slightly higher fluence; in this shot the surface suffered optical damage, and a large emission of Na was observed at short time delays. In addition, a series of aperiodic spikes persisted for at least tens of milliseconds after the laser pulse. These spikes are often followed by Maxwellian-like tails. Many large and small cleavage cracks were observed in the damaged region after this shot. The similarity of these sharp spikes to the HE neutral components observed immediately after the laser pulse at lower fluences suggests that microscopic cleaving or cracking of the surface is the origin for the HE neutrals, even below the threshold for observable damage.

Discussion. The properties of the neutral emission suggest a close relation to optical damage mechanisms. The ubiquitous correlation between the thresholds for observable neutral emission and observable optical damage is the strongest evidence that these phenomena are related. For all materials studied except Al_2O_3 , the emission-threshold fluence is about 10 to 40% of the optical damage threshold fluence for all laser wavelengths at which the materials are transparent. In fused silica and ZnS the rapid increase in the effective particle temperature, to values typical of an optical breakdown plasma just below the optical damage threshold, also suggests a connection with damage events. In NaF and

Fig. 5-70. Time-of-flight data for Na emission from NaF following a 0.266- μm , 3.8-J/ cm^2 laser pulse. Inset: initial 100 μs portion of the data.

Fig. 5-71. Time-of-flight data for Na emission from NaF following two laser shots. Sample suffered catastrophic damage on the second shot (7.46 J/ cm^2).

BaF_2 , neutral emission has always been observed to include the HE neutrals that probably result from cracking or cleaving, indicating that some degree of damage occurs on each laser shot.

Although these investigations are still far from complete because of the small number of materials studied and the lack of information concerning the effects of surface treatment and other variables, we can already draw some conclusions regarding the cause of the emission.

Consider first multiphoton interband excitation. The dependence of the Zn emission threshold on laser wavelength in Fig. 5-68 is far too weak to be consistent with multiphoton absorption, since the absorption at $1.06\text{ }\mu\text{m}$ would correspond to a five-photon process, and that at $0.355\text{ }\mu\text{m}$ to a two-photon process. At the peak intensities (10^8 W/cm^2) used in these experiments, these multiphoton transition rates would differ by many orders of magnitude.¹⁵⁴ The same behavior is found for NaF, BaF₂, and fused silica.

Desorption caused by thermal excitation (evaporation or sublimation) is also unlikely to be the cause of the neutral emission. The characteristic temperatures of the Maxwellian emission components of fused silica, ZnS, and NaF are in the range 300 to 800 K at the lowest fluences for which they are detectable. Not only are these temperatures far below the melting (or sublimation, for ZnS) temperatures, but they are comparable with the temperatures at which the surfaces are cleaned, and no emission is observed during cleaning. In addition, modeling of the surface temperature rise of ZnS resulting from excitation at $0.266\text{ }\mu\text{m}$, for which the absorption coefficient is known, suggests that the temperature rises less than 50 K at the threshold fluence for observable emission.¹⁵³

It has been suggested¹⁵⁵ that microscopic cleaving or cracking can result from modest increases in surface temperature because of the large surface stresses accompanying differential thermal expansion. This may be the cause of the HE neutrals observed just below the damage threshold in most of the materials we studied. Indeed, copious charged and neutral emission ("fractoemission"), as well as visible discharge, has been observed to accompany the fracture of numerous materials (see Ref. 156, for example). In addition to the direct damage caused by the crack propagation, the high gas densities and ionization levels produced by this localized phenomenon can also precipitate local optical breakdown, which could grow catastrophically because of the resulting surface bombardment.

The Maxwellian-like emission from fused silica and ZnS is observed even without the HE neutrals at low fluences,

so it cannot be attributed to any mechanism of thermal origin, including cracking. It is most likely that this emission results from electronic excitation of defects or surface states. The carriers released by this process can cause bond breaking, followed by desorption of atoms in analogy with the well-documented desorption processes observed in the electron, ion, and UV photon excitation of a variety of materials.^{157,158} The F center, which is a singly charged sulfur vacancy, is an example of a defect in ZnS that absorbs throughout the visible; photoconductivity measurements show that free carriers are produced by this absorption.¹⁵⁹

Despite its rather innocuous appearance, the electronic desorption discussed in the preceding paragraph could play a central role in optical damage because of the energy transferred to the surface or through the initiation of avalanche ionization. Furthermore, if the emission alters the surface composition, a catastrophic, positive feedback into the density of absorbing defects could result. In ZnS, for example, a predominance of sulfur emission would produce S vacancies, and possibly F centers, at the surface. Obviously, this surface ablation could also reduce the damage threshold for multiple-pulse excitation.

Conclusion. Laser-induced neutral emission appears to be a common precursor to observable optical surface damage. Not only is it a signature of the interaction of the laser light with the surface, but it may be a cause of optical damage.

Authors: L. L. Chase and L. K. Smith

Transparent Glass/Crystal Composites for Laser Hosts

Under a Lawrence Livermore National Laboratory Institutional Research and Development award, we have investigated the feasibility of producing a transparent glass/crystal composite for use as a laser host by combining crystallites into a glass tailored for a precise refractive index match. Although we produced suitably matched composites, the feasibility of this concept is severely limited

by glass dissolution of the crystallites, boundary layers of different refractive indexes, and devitrification at the glass/crystal interface.

Both glasses and single crystals are used as laser hosts. We investigated the possibility of producing an optical-quality glass/crystal composite that combined the ease of formation, low cost, and large sizes of glass components¹⁶⁰ with the good laser and thermomechanical properties of single crystals.¹⁶⁰ Our approach was to add crystals to a glass matrix of the same refractive index.

Our initial goal was to produce a high-optical-quality glass/crystal composite with a scattering loss of less than $0.1\% \text{ cm}^{-1}$. Historically, the use of glass/ceramics as laser materials has been limited by scattering losses, which were minimized by homogeneously crystallizing very small particles from the melt. The crystallized particles have a different chemical composition and a different refractive index than those of the bulk material. The most successful of these efforts¹⁶¹ resulted in scattering losses of $4\% \text{ cm}^{-1}$. We sought to decrease the refractive index mismatch between the crystallite and glass, which would make it possible to use larger crystals in the composite.

In our composites, scattering losses can arise from a variety of sources, particularly from a mismatch between the refractive indexes or thermal expansion coefficients of the glass and crystal phases. Temperature gradients, which are often present in laser components, produce refractive index variations (and hence scattering) if $\partial n / \partial T$ is different for the glass and crystal.

Surface-related effects, including contamination, interdiffusion between the glass and crystal, and compositional differences between the bulk and surface of either material, can also produce scattering. Scattering losses are also associated with bubbles, striations, or index inhomogeneities resulting from uneven cooling of the glass.

In producing these composites, one critical question was the dependence of scattering coefficient on refractive index mismatch and crystallite size; Fig. 5-72 shows the calculated relation between

Fig. 5-72. Scattering coefficient vs index mismatch for crystal particles of various diameter [50 vol% crystal; spherical shape factor ($k^2 = 1$)].

these quantities. To reach our scattering goal of $0.1\% \text{ cm}^{-1}$, the refractive index mismatch must be less than 10^{-4} for $1\text{-}\mu\text{m}$ -diam crystals.

For our experiments, we first selected candidate crystals. The crystals had to be cubic, so that they were not naturally birefringent, and had to have refractive indexes and coefficients of thermal expansion compatible with those of known glass systems. We looked for crystals with high melting temperature, to minimize their dissolution in molten glass.

We then selected the glass system. The glass had to have a refractive index range that included the crystal index, a thermal expansion coefficient similar to that of the crystal (to minimize stress), a large glass-forming region to inhibit devitrification, and a low melting temperature to minimize crystal dissolution in the glass.

To match the refractive indexes we first carried out enough glass melts to permit the creation of reasonably accurate isofrac diagrams (glass phase diagrams on which are superimposed contours of constant refractive index). We could then select a glass with a refractive index tailored to match that of the crystal. We used annealing to homogenize the refractive index of the glass and (by varying the annealing time) to fine-tune the index to more closely match that of the crystal. We examined only oxide glasses in these preliminary studies, because of the convenience of forming the composites in air and because oxide glass powders could be produced by sol-gel methods, which permitted the

formation of small, homogeneous glass billets in a single melting.

Formation of the Composites. The compatibility of the best glass/crystal systems was determined through solubility studies and optical characterization (see Table 5-29). The most common problems we encountered were excessive thermal expansion mismatch between the glass and crystal, rapid dissolution of the crystallites (especially in high-index glasses that contained heavy metal oxides) and spontaneous devitrification. Devitrification occurred when we tried to increase the refractive indexes of several glasses to match those of the crystals by adding high-index constituents, which acted as nucleating agents in the glass.

The best simultaneous match of refractive index and thermal expansion coefficient was obtained with a lanthanum borate glass and MgAl_2O_4 (spinel; shown near the top of Table 5-29). The composites formed by simple mixing with this

system suffered excessive crystal dissolution, so we investigated a number of new formation methods. These methods progressed from room-temperature formation to formation at the glass transition temperature and finally to formation above the glass melting temperature.

Low-temperature forming was unsatisfactory: the refractive index of the glass was not homogeneous, there were seeds in the glass, and a seam was visible between the glass and the crystallites. The high-temperature composite formation methods were also unsatisfactory, because of extensive crystal dissolution (and resulting striations) in the glass near the crystal/glass interfaces. We then worked on decreasing the time and temperature required for the composite formation, so as to inhibit dissolution. We sintered highly reactive sol-gel glass powders in the presence of crystals, but the glass devitrified at the glass/crystal interface. Further investigation of the

Table 5-29. Physical properties of the best crystal/glass composites made in this study.

Glass system and number of compositions melted	Primary compatibility problem	Refractive index mismatch ^a	Calculated density (g/cm ³)	Calculated thermal expansion mismatch ^b (10 ⁻⁶ °C)
MgAl₂O₄ crystal				
Na ₂ O · SiO ₂ · TiO ₂ (1)	Thermal expansion mismatch	0.017	2.75	5
Na ₂ O · SiO ₂ · TiO ₂ (2)	Devitrified glass	— ^c	2.94	8
Na ₂ O · SiO ₂ · TiO ₂ · Al ₂ O ₃ (1)	Thermal expansion mismatch	0.003	2.73	6
PbO · P ₂ O ₅ (1)	Thermal expansion mismatch	0.012	5.56	6
SiO ₂ · PbO · Al ₂ O ₃ (7)	High crystal dissolution rate	0.003	4.90	1
La ₂ O ₃ · SiO ₂ · B ₂ O ₃ · ZrO ₂ · CaO (1)	Best match	0.002	3.55	0.2
Y₃Al₅O₁₂ crystal				
SiO ₂ · PbO · Al ₂ O ₃ (3)	High crystal dissolution rate	0.003	5.06	0.2
SiO ₂ · PbO · Al ₂ O ₃ (4)	High crystal dissolution rate	0.023	5.49	0.2
B ₂ O ₃ · Sb ₂ O ₃ (1)	High crystal dissolution rate	0.023	3.65	3
B ₂ O ₃ · Sb ₂ O ₃ (2)	High crystal dissolution rate	0.023	3.76	2
B ₂ O ₃ · PbO (1)	High crystal dissolution rate	0.007	4.14	0.4
B ₂ O ₃ · SiO ₂ · PbO (1)	Devitrified at interface	0.007	4.26	0.9
PbO · P ₂ O ₅ (2)	Devitrified; low index	— ^c	6.02	5
WO ₃ · PbO · CaO · TiO ₂ · ZrO ₂ · BaO ₅ (1)	Absorbed visible light	—	5.06	6
P ₂ O ₅ · TiO ₂ (2)	Low index; devitrified when raised	— ^c	5.77	6
GeO ₂ · K ₂ O · BaO · Sb ₂ O ₃ (1)	Devitrified at interface	0.117	5.51	0.7
GeO ₂ · K ₂ O · BaO · Sb ₂ O ₃ (2)	Devitrified at interface	0.037	4.94	1
CaO · SiO ₂ · TiO ₂ (1)	Low index; devitrified when raised	— ^c	2.99	4
CaO · SiO ₂ · TiO ₂ (2)	Low index; devitrified when raised	— ^c	2.90	3
MgO crystal				
La ₂ O ₃ · SiO ₂ · B ₂ O ₃ · ZrO ₂ · CaO (2)	Thermal expansion mismatch	0.018	3.40	0.7

^a Refractive indexes n_D were measured using immersion oils and an optical microscope.

^b Reference 162.

^c Devitrified.

sol-gel-derived powders sintered in the presence of moderate-sized crystals revealed that devitrification always occurred (apparently because of the nucleating effect of the crystal) and that the resulting material had an interfacial refractive index that differed from that in the bulk of the glass.

Three principal problems thus limit the usefulness of our concept. The primary problem is that molten glass is an excellent solvent for crystals. While it might be possible to minimize this behavior by using more sophisticated forming methods (such as hot isostatic pressing) to reduce the reaction temperature, we believe that dissolution fundamentally limits the feasibility of this concept.

A second problem is the relatively small overlap in refractive indexes between oxide glasses and oxide crystals. In special cases we have produced adequate matches ($\Delta n < 10^{-4}$), but fortuitous matches cannot form the basis of a general method for producing transparent composites. It may be possible to reduce this problem by using other glass systems, such as fluorides; this would increase the number of glass systems whose refractive indexes could be matched. However, fluorides do not form glasses as readily as oxides, and devitrification at the glass/crystal interface would probably be exacerbated.

A third problem is that the refractive index of the boundary layer differs from that of the glass or crystal. It may be possible to correct this problem in isolated systems (at least in two dimensions) by engineering an interface with a refractive index gradient that does not produce significant scattering, but this approach would not be widely applicable.

Thus, while it may be possible to produce optical-quality glass/crystal composites for isolated systems, the problems of crystal dissolution, a lack of refractive index overlap between glasses and crystals, and boundary layer effects will in general severely restrict the feasibility of this approach to making low-cost transparent materials.

Authors: J. B. Davis, J. E. Marion,
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Theoretical Atomic and Molecular Physics

Introduction

The Theoretical Atomic and Molecular Physics Group has continued the development of state-of-the-art numerical techniques for the study of problems of interest to many aspects of the Livermore Laser Program. We have pursued accurate descriptions of electron scattering from atoms and molecules, the interactions of atoms, molecules, and ions with radiation, and the phenomena of charge transfer and similar atomic scattering processes. An effort to understand in detail the deposition of laser energy in and radiation from laser-produced plasmas was another significant part of our research in 1987.

Author: K. C. Kulander

Potential Storage Laser Based on the SO Molecule

Introduction. Over the past few years the advanced laser program has supported experimental work by D. Setser and his group at Kansas State University aimed at finding a gaseous medium capable of storing significant amounts of excitation energy to be used in a fusion laser system.

The major focus of this work in 1987 was the sulfur monoxide radical, which is easily generated in a number of ways. The SO system has a long-lived excited $^3\Pi_{0,1,2}$ state whose excitation energy is approximately 4 eV. Accurate determinations of the radiative lifetime and of the electronic quenching rates of this state by various gases have been made, and the spin-orbit relaxation rate constants for the upper state components have been measured. Data obtained in this work and in earlier spectroscopic studies permitted us to predict the gain cross sections for the $A\ ^3\Pi_0\ (v' = 0) \rightarrow X\ ^3\Sigma\ (v' = 4)$ proposed laser transitions for

for various buffer gas densities. These studies allow us to characterize the value of this system as a fusion laser medium with reasonable completeness.

The investigation of SO is part of a broad study of possible laser applications of the class of isovalent molecules that also includes O_2 , PF and NF. The SO ($A \rightarrow X$) transition has a number of promising attributes for a UV laser: a long radiative lifetime (tens of microseconds), modest electronic quenching cross sections, a high degree of chemical inertness [which allows the preparation of high SO (X) concentrations], and an appreciable change in the equilibrium internuclear distances between the two electronic states, which permits transitions from the lowest vibrational level of the excited state to high, unpopulated vibrational levels of the ground state. These attributes are expected to be common to the other isovalent species mentioned above. The SO studies should therefore illustrate the behavior of a class of possible fusion lasers.

Results. The experiments¹⁶³ were carried out using a flow reactor with a three-axis arrangement in which the directions of the gas flow, the pump laser beam, and the laser-induced fluorescence (LIF) observations were mutually perpendicular. Two gas input ports were available: one for the $SOCl_2/Ar$ mixture (0.1% $SOCl_2$, total flow rate of 4 mmole/s), which passed through a microwave

discharge, and one for the carrier gas (and added reagent). The excitation laser beam was generated by a Questek XeCl-excimer laser pumping a Lambda Physik FL2002 tunable dye laser; the output was frequency doubled with an Inrad Autorakker 2 system (based on a KDP-A crystal). The SO ($A \rightarrow X$) fluorescence was observed with a 0.5-m Minuteman monochromator (17 Å/nm dispersion) fitted with a 1200 nm⁻¹ grating blazed at 250 nm and a Hamamatsu R212UH photomultiplier tube.

Rotationally resolved excitation spectra of the SO ($X \rightarrow A$) transitions were obtained by observing the $A \rightarrow X$ emission. Wavelengths selected for excitation of the $^3\Pi_0$, $^3\Pi_1$, and $^3\Pi_2$ states (from X , $v = 0$) were 263.7, 262.47, and 261.4 nm, respectively.

The radiative lifetime for SO ($A\ ^3\Pi_2$, $v' = 0$) was determined by extrapolation of a Stern-Volmer plot of the fluorescence decay constants obtained in argon carrier gas over the range 0.1 to 2 Torr with $[SOCl_2] = 1 \times 10^{13}$ molecules cm⁻³. The measured quenching rate constant for argon is $(8.5 \pm 1.0) \times 10^{-13}$ cm³ molecule⁻¹ s⁻¹, and the intercept gives a radiative lifetime of 35 ± 3 μs. The relative emission intensities of the bands from the $^3\Pi_0$, $^3\Pi_1$, and $^3\Pi_2$ states in 1 Torr of argon are in the ratio of the Boltzmann populations, so we conclude that the lifetimes of the individual spin-orbit states are not substantially different from one another.

The quenching rate constants for the other reagents were measured by fixing the argon pressure at 0.5 Torr and adding the reagent to the reactor while recording the SO decay times following excitation. Table 5-30 gives our results for a number of reagents. It is interesting that the rate constants for CO_2 , CF_4 , CH_4 , and N_2 are small, whereas those for CO , NO , and HCl are large.

To study the collisional relaxation between the $\Omega = 2$, 1, and 0 states, we monitored the fluorescence from each state with a monochromator. The relaxation obeys the selection rule $\Delta\Omega = \pm 1$. Two-body rate constants, obtained with a kinetic model of the observed data, are of the order of 2×10^{-11} cm³ molecule⁻¹ s⁻¹.

Table 5-30. Rate constants and cross sections for quenching of SO (A , $v' = 0$).

Reagent, Q	k_Q (10^{12} cm ³ molecule ⁻¹ s ⁻¹)	σ_Q^a (Å ²)
He	0.53 ± 0.09	0.04
Ar	0.85 ± 10	0.16
H ₂	75 ± 11	4.1
D ₂	100 ± 10	—
N ₂	38 ± 9	6.3
O ₂	220 ± 40	38
NO	310 ± 70	53
CO	200 ± 40	33
HCl	260 ± 30	47
CO ₂	21 ± 3	4.0
CF ₄	1.5 ± 0.2	0.33
CH ₄	4.4 ± 0.3	0.61
$SOCl_2$	220 ± 30	51

^a $\sigma_Q = k_Q/v$, where $v = (8kT/\pi\mu)^{1/2}$, and μ is the reduced mass of SO and Q. $T = 300$ K.

Gain cross sections were determined with the newly determined excited-state lifetime and the spectroscopic analysis^{164,165} of these two electronic states. The upper state $A^3\Pi_0$ ($v' = 0$) has a favorable branching ratio to the $X^3\Sigma^-$ ($v'' = 4$) lower level. The gain cross section for the transition between vibrational-rotational levels $v'J'$ and $v''J''$ is given by

$$\sigma_{v'J',v''J''}(V) = \frac{c^2}{8\pi\nu^2} g(\nu - \nu_0) A_{v',J',v''} \frac{S_{J',J''}}{(2J' + 1)L'} N_{v',J'} \quad (56)$$

To evaluate this expression we require the inverse $A_{v',J',v''}$ of the spontaneous lifetime of the initial vibrational level; the vibrational branching factor $I_{v',v''}$; the rotational line strength factor $S_{J',J''}$; the relative population of the initial state $N_{v',J'}$; and the line shape factor $g(\nu - \nu_0)$. Here L' is the lambda-doubling degeneracy factor, which is 2 for the initial $^3\Pi_\Omega$ electronic state.

We assume that the line shape factor is a Gaussian of width $\Delta\nu$ given by

$$g(\nu - \nu_0) = \left(\frac{4 \ln 2}{\pi}\right)^{1/2} \frac{1}{\Delta\nu} \exp\left[-4 \ln 2 \left(\frac{\nu - \nu_0}{\Delta\nu}\right)^2\right] \quad (57)$$

where the transition frequency is

$$\nu_0 = \frac{1}{h}(E_{v',J'} - E_{v'',J''}) \quad (58)$$

We assume a pressure broadening of 10 MHz/Torr. The Doppler width dominates for total pressures below 200 Torr. The Boltzmann factor defining the initial state population is given by

$$N_{v',J'} = \left[\frac{\exp(-E_{elec}/kT)}{Q_{elec}} \right] \left[\frac{\exp(-E_{vib}/kT)}{Q_{vib}} \right] \times \left[\frac{(2J' + 1)\exp(-E_{rot}/kT)}{Q_{rot}} \right] \quad (59)$$

where the Q 's are the respective partition functions.

The total gain cross section at frequency ν from all transitions is therefore given by

$$\sigma_{v',v''}(V) = \sum_{J',J''} \sigma_{v',J',v'',J''}(V) \quad (60)$$

(Contributions from different spin components of the upper state and from different vibrational transitions do not overlap significantly.) Multiplying this cross section by the total population in the A state gives the gain per unit length at this frequency.

Figure 5-73 shows the calculated gain cross section for collisionally broadened lines with a width of 0.25 cm^{-1} , corresponding to 1 atm of buffer gas. These calculations show that the gain cross section is expected to be $3 \times 10^{-19} \text{ cm}^2$. This means that if 1 Torr of excited states can be generated, the predicted gain will be a few percent per centimeter.

Our calculations show that the gain cross section is acceptable for a fusion laser medium, but the moderately high quenching rates (even for the lighter inert gases) may limit the storage capability of this system. A reasonable amount of buffer gas must be present to maintain thermal stability, to vibrationally relax the excited state, and to broaden the

Fig. 5-73. SO A ($v' = 0$) $\rightarrow$ X ($v'' = 4$) gain cross section in 1 atm of buffer gas.

emission lines. It is unclear whether any of the other isovalent species mentioned above will be better suited to this purpose. Electronic structure calculations are required to clarify the electronic quenching mechanism, particularly the role of the nearby $^3\Delta$ state, to complete our understanding of this system.

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Analysis of Dot-Target Plasma

A understanding of the basic processes occurring in highly stripped, dense plasmas is important for understanding the microphysics of laser-driven fusion implosions. Characterization of these plasmas requires experimental diagnostics capable of resolving details of plasmas that are typically not stationary, optically thin, nor described by steady-state atomic kinetics. One very powerful and steadily evolving technique is the spectroscopy of laser-produced plasmas. Recent advances in the use of "microdot" or "spot" targets,^{166,167} which produce relatively homogeneous, confined, optically thin plasmas, make first-principles treatments possible. The production of temporally and spatially resolved experimental spectra^{168,169} offers the prospect of refining both the experimental and theoretical approaches to more complicated systems.

A complete simulation of these spectra provides a direct test of the major components of a theoretical treatment that couples hydrodynamic motion with radiation transport, using detailed atomic rates for the many kinetic processes occurring. A deficiency or inaccuracy in any of these components will be clearly manifest in the predicted spectra. The work reported here describes an attempt to synthetically produce the spectra observed in a spatially resolved, time-averaged microdot experiment that has been the subject of several other analyses.^{168,170,171} The analysis given here differs from these others in using no

phenomenological adjustment of the synthetic spectra and in providing a detailed picture of the evolving conditions within the plasma.

The strategy pursued here is similar in spirit to that of several other theoretical calculations.¹⁷² First, a coupled hydrodynamic and radiative transport computation is performed to describe the laser-material interaction that produces the plasma. Second, zonal information describing this evolving plasma is extracted at a succession of times and used to provide the conditions for an atomic kinetics calculation. Finally, the predicted emissivities at different times and spatial locations in a specified spectral range are selected and plotted.

In addition to the deficiencies inherent in the calculations at any one of these stages, there is clearly the possibility that this scheme is not self-consistent, because the hydrodynamic-radiative calculation is separated from the detailed atomic treatment. If the atomic processes severely modify the material's macroscopic properties (for example, if they lead to heating or cooling), one would not expect this procedure to yield agreement with experiments. While it is desirable to carry out the first and second steps self-consistently, the large number of atomic rates would probably render this approach overwhelming.

The microdot technique used in the experiment considered here is amply discussed by Bailey et al.¹⁶⁸ The target geometry and laser characteristics chosen for calculation correspond to a shot with relatively low laser fluence. The target was a 10% NaBr/90% NaCl dot (10 μm radius, 5000 Å thick) deposited uniformly on a Mylar substrate. The Nd laser was operated with green light (523 nm) and delivered 5.8 J in a Gaussian temporal pulse with a 0.7-ns FWHM. The focal spot radius was 91 μm , which reproduces the measured illuminated area of $2.58 \times 10^{-4} \text{ cm}^2$. The corresponding fluence was $3.2 \times 10^{13} \text{ W/cm}^2$.

These values were used in the Lagrangian radiation-hydrodynamic code LASNEX, which then described the plasma evolution from cold material. This simulation was performed in two dimensions (cylindrical symmetry

Fig. 5-74. Time averaged, spatially resolved experimental x-ray spectrum for spatial bin $15\ \mu\text{m}$ from target surface. Labeled are neon-like $2s\text{-}3p$ (3A, 3B), $2p\text{-}3d$ (3C, 3D), and $2p\text{-}3s$ transitions (3F, 3G). Other features are sodium-like or blends of neon-like and sodium-like emissions.

enforced) with an average-atom non-LTE model (XSN) for the atomic radiative rates and a hydrogenic model including some dielectronic recombination (LASNEX-DCA). Radiative transport was handled with an escape-probability treatment.

The second step in this procedure uses the information generated by the hydrodynamic simulation in the time-dependent atomic-rate-equation code XRASER. A one-dimensional slab of material zones was followed at 50-ps intervals with the emissivity and opacity for the slab calculated by a detailed atomic model. The atomic model used is similar to one used in a previous analysis of this experiment.¹⁷¹ The rates

included in this data base treated three ion stages of the bromine in great detail (fluorine-like, neon-like, and sodium-like); hydrogenic level descriptions were used for several neighboring charge states. These values were consistently incorporated using the model-generating code ADAM.

The 113 fluorine-like levels treated included all one-electron excitations from $n = 2$ to $n = 3$; the 89 neon-like levels included all such excitations from $n = 2$ to $n = 3$ and $n = 4$; and the 237 sodium-like levels included the five low-lying $2p^6 3l$ levels and the $2p^5 3l 3l'$ and $2s 2p^6 3l 3l'$ levels. Surrounding ion stages

(carbon-like through silicon-like) were incorporated in the hydrogenic approximation for completeness. A detailed oxygen-like stage was also used in preliminary calculations but was found to be unnecessary in the interpretation of the spectral information.

Radiative transition and electron collisional excitation rates between all levels were evaluated. Electric quadrupole and magnetic dipole forbidden rates were included for completeness. Because the observed spectrum resolved neon-like to sodium-like recombination features, it was necessary to include these rates specifically. Fluorine-like recombination to the neon-like stage was not seen in the experimental spectrum, but we found that its exclusion from the calculations led to incorrect results. Since information on individual spectral lines is not needed, the fluorine-like recombination was handled by summing over the intermediate doubly excited states and weighting by recombination rates and fluorescence yield. The same procedure was used for sodium-like recombination to the magnesium-like ion stage.

Synthetic spectra are readily produced from this accumulated information. Figure 5-74 shows the temporally averaged, spatially resolved information taken during the experiment for a bin centered at

Fig. 5-75. Time averaged, spatially resolved synthetic x-ray spectrum corresponding to experimental data of Fig. 5-74.

15 μm from the target surface; several of the more important neon-like features are identified. Figure 5-75 shows the corresponding calculated spectrum, generated with the approximate experimental resolution $\Delta\lambda/\lambda \simeq 10^{-3}$ as an instrumental convolution width.

The agreement between the observed and calculated results is generally good. Most of the neon-like and sodium-like recombination features are well represented. No significant fluorine-like emission is predicted, in accord with the experimental results. The primary features in disagreement appear to be the weakness in the predicted 2p-3s lines (3F and 3G), which are strong in the experiment, and the strength of the predicted 2p-3d lines (3C and 3D), which are not observed. This difficulty could be removed if the relative strength of the 2p-3d lines were reduced somewhat. These lines have the greatest optical depths, so they are most susceptible to inaccuracies in treating the plasma column thickness; we speculate that uncertainties in the reduction of the two-dimensional problem to a one-dimensional slab may be the primary cause of this error.

It is clear, then, that resolved x-ray diagnostics can provide a stringent test of theoretical treatments. In particular, microdot spectroscopy seems to offer an excellent technique for the calibration of analytic methods, since the experiment can control several of the complicating features of laser-produced plasmas. Time- and space-resolved spectra will offer an even greater opportunity for the confrontation of experiment and theory.

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Variational Methods for Calculating Electron Impact Excitation of Molecules

Introduction. The development of reliable and efficient theoretical methods for computing cross sections for low-energy electron-molecule scattering has been difficult. These cross sections are needed in such diverse fields as advanced laser development, x-ray diagnostics, and atmospheric physics.

Traditional algebraic variational methods such as the Kohn method,¹⁷³ which have been used extensively in electron-atom scattering problems, have been virtually ignored in molecular applications despite the considerable flexibility they allow in the choice of the trial wave function. Now interest in the Kohn method has been rekindled. Our recent work¹⁷⁴ shows that the spurious singularities that can plague the traditional method are eliminated when the Kohn method is applied to the T matrix rather than the K matrix (which simply involves a change to complex, outgoing-wave boundary conditions).

During 1987 we removed another stumbling block to application of the Kohn method to molecules.¹⁷⁵ The problem involved practical difficulties associated with the computation of certain continuum matrix elements. We showed that

the method could be formulated within a separable-potential approach, so as to eliminate the need for computing bound-free and free-free exchange matrix elements. Correlation and polarization effects could also be systematically included in the treatment through the use of optical potential techniques.

Discussion. We consider the scattering of low-energy electrons by an N -electron target molecule. To describe the scattering of an electron, incident in a channel denoted by the label Γ' , we use an antisymmetrized trial wave function of the form

$$\psi_{\Gamma'} = \sum_{\Gamma} \mathcal{A}(\chi_{\Gamma} F_{\Gamma\Gamma'}) + \sum_{\mu} d_{\mu}^{\Gamma'} \Theta_{\mu}, \quad (61)$$

where the first sum runs over the energetically open N -electron target states, denoted by the normalized functions χ_{Γ} , and the operator $\mathcal{A}$ antisymmetrizes the orbital functions $F_{\Gamma\Gamma'}$ into the functions χ_{Γ} . The Θ_{μ} are an orthonormal set of antisymmetric, square-integrable $(N+1)$ -electron functions used to represent polarization and correlation effects not included in the first summation. The symbol Γ labels all the quantum numbers needed to represent a physical state of the composite system, i.e., the internal state of the target molecule and the energy and orbital angular momentum of the scattered electron.

To calculate physical differential cross sections, it is convenient to project the T matrix onto a set of angular functions. To this end, the channel continuum functions $F_{\Gamma\Gamma'}$ are further expanded as

$$r F_{\Gamma\Gamma'}(\mathbf{r}) = \sum_l [f_l^{\Gamma'}(r) \delta_{ll'} \delta_{\Gamma\Gamma'} + T_{lm' \Gamma' m}^{\Gamma} g_l^{\Gamma'}(r)] Y_{lm'}(\hat{\mathbf{r}}) + \sum_k c_k^{\Gamma\Gamma'} \phi_k^{\Gamma}(\mathbf{r}), \quad (62)$$

where Y_{lm} is a normalized spherical harmonic, the ϕ_k^{Γ} are a set of square-integrable functions, and $f_l^{\Gamma'}$ and $g_l^{\Gamma'}$ are linearly

independent continuum orbitals that are regular at the origin and that behave asymptotically as regular and outgoing Riccati-Bessel functions, respectively:

$$f_l^{\Gamma'}(r) = \frac{j_l(k_{\Gamma} r)}{\sqrt{k_{\Gamma}}} \underset{r \rightarrow \infty}{\sim} \frac{\sin(k_{\Gamma} r - l\pi/2)}{\sqrt{k_{\Gamma}}},$$

$$g_l^{\Gamma'}(r) = \frac{i[j_l(k_{\Gamma} r) + i n_l(k_{\Gamma} r) c(r)]}{\sqrt{k_{\Gamma}}}$$

$$\underset{r \rightarrow \infty}{\sim} \frac{\exp[i(k_{\Gamma} r - l\pi/2)]}{\sqrt{k_{\Gamma}}}. \quad (63)$$

The function $c(r)$, which makes $g_l^{\Gamma'}$ regular at the origin, is chosen to be $[1 - \exp(-\alpha r)]^{2l+1}$. The channel momenta k_{Γ} (atomic units) are determined by energy conservation:

$$\frac{k_{\Gamma}^2}{2} = E - E_{\Gamma}, \quad (64)$$

where E is the total energy of the composite system and E_{Γ} is the energy of the target molecule corresponding to χ_{Γ} . The coefficients $T_{lm' \Gamma' m}^{\Gamma}$ are elements of the T matrix and are the fundamental dynamical quantities that determine scattering amplitudes and cross sections.

It is important to note that we are not attempting a complete single-center expansion of the channel wave function. The sum over l that appears in Eq. (62) can be restricted to those "asymptotic" l values needed to represent the channel wave function at large distances and to provide converged cross sections at a particular energy. The burden of representing the short-range, high- l components of the wave function near the nuclei is carried by the ϕ_k^{Γ} .

The closed-channel correlation functions Θ_{μ} can be formally incorporated into an effective optical potential by using projection operator techniques. If Q is the operator that projects onto the subspace defined by the Θ_{μ} , and P is its orthogonal

complement, then we can manipulate the Schrödinger equation in a standard way to produce a modified Hamiltonian,

$$\begin{aligned} \mathbf{H}_{\text{eff}} &= \mathbf{H}_{PP} \\ &+ (\mathbf{H} - \mathbf{E})_{PQ}(\mathbf{E} - \mathbf{H})_{QQ}^{-1}(\mathbf{H} - \mathbf{E})_{QP} \\ &\equiv \mathbf{H}_{PP} + \mathbf{V}_{\text{opt}}. \end{aligned} \quad (65)$$

The coefficients $T_{lm'l'm'}^{\Gamma\Gamma'}$ and $c_k^{\Gamma\Gamma'}$ in Eq. (62) can be treated as variational parameters whose values are to be determined from a stationary principle. The Kohn principle can be used to characterize the T matrix as the stationary value of the functional¹⁷³

$$[\mathbf{T}^{\Gamma\Gamma'}] = \mathbf{T}^{\Gamma\Gamma'} - 2 \int \psi_{\Gamma}(\mathbf{H}_{\text{eff}} - \mathbf{E})\psi_{\Gamma'}, \quad (66)$$

where the terms Θ_{μ} no longer appear in ψ_{Γ} . In a condensed matrix notation, in which open-channel indices are suppressed, the result can be written as

$$[\mathbf{T}] = -2(\mathbf{M}_{00} - \mathbf{M}_{0q}\mathbf{M}_{qq}^{-1}\mathbf{M}_{0q}), \quad (67)$$

where the elements of $\mathbf{M}_{00}$ are defined as

$$\begin{aligned} (\mathbf{M}_{00})_{lm'l'm'}^{\Gamma\Gamma'} &= \int \mathcal{N}(\chi_{\Gamma} f_l^{\Gamma} Y_{lm}) (\mathbf{H}_{\text{eff}} - \mathbf{E}) \mathcal{N}(\chi_{\Gamma'} f_{l'}^{\Gamma'} Y_{l'm'}), \\ & \quad (68) \end{aligned}$$

and $\mathbf{M}_{0q}$ and $\mathbf{M}_{qq}$ are similarly defined as matrix elements of $(\mathbf{H}_{\text{eff}} - \mathbf{E})$, with q referring to the subspace spanned by $\mathcal{N}(\chi_{\Gamma} g_l^{\Gamma} Y_{lm})$ and $\mathcal{N}(\chi_{\Gamma} \phi_k^{\Gamma})$. Because of the outgoing-wave behavior of the g_l^{Γ} , $\mathbf{M}_{qq}$ will be complex-symmetric and its inverse will be nonsingular for real energies. Thus we obtain an anomaly-free expression for the T matrix.¹⁷⁶

For practical calculations on many-electron systems, it is essential to work with an orthogonal basis. The transfer invariance of the total wave function guarantees that the T matrix is unchanged by any unitary transformation between the bound functions ϕ_k^{Γ} and the continuum orbitals f_l^{Γ} and g_l^{Γ} , and that

they can be taken to be mutually orthogonal. In addition, we construct the ϕ_k^{Γ} so that they are orthogonal to the set of molecular orbitals used to form the target states χ_{Γ} and the correlation terms Θ_{μ} . This prescription imposes a strong orthogonality condition between the continuum channel functions and the target wave functions.

All the matrix elements needed to construct $\mathbf{M}_{qq}$, $\mathbf{M}_{00}$, and $\mathbf{M}_{0q}$ can be formed from one- and two-electron integrals. The integrals involving purely bound functions may be extracted from standard electronic structure programs. Of the two-electron integrals involving either one (bound-free) or two (free-free) continuum functions, it is useful to distinguish between direct integrals and exchange integrals, which arise from interchange of "bound" and "free" electron coordinates. The direct integrals may be expressed in terms of local, one-particle transition potentials $\mathbf{V}_{\Gamma\Gamma'}$ and can be easily evaluated by numerical quadrature. The exchange integrals are considerably more difficult to evaluate.

The formulation of the Kohn method is considerably simplified by using a separable representation of the exchange components of $\mathbf{H}_{PP}$; because of the short-range nature of the exchange interaction, this representation converges quite rapidly.¹⁷⁵ In making up the $(N+1)$ -electron configurations with which to form the separable representation of $\mathbf{H}_{PP}^{\text{exch}}$, we use the same set of square-integrable functions to which the free functions f_l^{Γ} and g_l^{Γ} have been orthogonalized, then all free-free and bound-free exchange integrals will simply vanish as a consequence of the strict orthogonality condition we have imposed on the trial wave function. The entire effect of exchange is thus carried by the bound-bound components of $\mathbf{H}_{\text{eff}}$. This same separable expansion is used in evaluating matrix elements of the optical potential, so that all bound-free and free-free matrix elements of $\mathbf{V}_{\text{opt}}$ are eliminated from consideration as well.

The final working expression for $[\mathbf{T}]$ is

$$[\mathbf{T}] \simeq 2(\hat{\mathbf{M}}_{00} - \hat{\mathbf{M}}_{0q}\hat{\mathbf{M}}_{qq}^{-1}\hat{\mathbf{M}}_{0q}), \quad (69)$$

where

$$\begin{aligned}
 & (\tilde{M}_{00})_{lm'l'm'}^{\Gamma\Gamma'} \\
 &= \delta_{\Gamma\Gamma'} \delta_{ll'} \delta_{mm'} \left\langle f_l^{\Gamma} \left| -\frac{1}{2} \frac{d^2}{dr^2} + E_{\Gamma} - E \right| f_{l'}^{\Gamma'} \right\rangle \\
 &+ \langle f_l^{\Gamma} Y_{lm} | V_{\Gamma\Gamma'} | f_{l'}^{\Gamma'} Y_{l'm'} \rangle. \quad (70)
 \end{aligned}$$

The matrix $\tilde{M}_{0q}$ is similarly defined, with the subscript q again referring to the sub-space spanned by $\mathcal{V}(\chi_{\Gamma} g_l^{\Gamma} Y_{lm})$ and $\mathcal{V}(\chi_{\Gamma} \varphi_k^{\Gamma})$. The matrices $\tilde{M}_{00}$ and $\tilde{M}_{0q}$ contain neither exchange nor optical potential terms; these only appear in the bound-bound components of $\tilde{M}_{qq}$, which are defined as

$$\begin{aligned}
 & (\tilde{M}_{qq})_{kk'}^{\Gamma\Gamma'} \\
 &= \int \mathcal{V}(\chi_{\Gamma} \varphi_k^{\Gamma}) (\mathbf{H}_{pp} + \mathbf{V}_{\text{opt}} - \mathbf{E}) \mathcal{V}(\chi_{\Gamma'} \varphi_{k'}^{\Gamma'}). \quad (71)
 \end{aligned}$$

These matrix elements can be readily extracted from electronic structure codes. All other matrix elements are evaluated by numerical quadrature.

The techniques outlined here have been found to be accurate and efficient in e^- -H and e^- -He scattering calculations,¹⁷⁵ and we have successfully performed calculations on e^- -H₂ in both the static-exchange and two-state approximations.¹⁷⁷ Detailed studies of the electron-impact dissociation of H₂ are in progress.

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The Cu + e⁻ System: Electron Scattering and Photodetachment

Introduction. The excited states of atomic copper relevant to the copper-vapor laser¹⁷⁸ (CVL) are populated primarily by collisions with electrons. The ground 3d¹⁰4s ²S^e state is preferentially excited by collisions to the 3d¹⁰4p ²P^o states, where a population inversion is

created with respect to the lower, meta-stable 3d⁹4s² ²D^e states. The ²P^o-²D^e transitions are the sources of the observed laser lines.

Recent interest in more powerful CVLs has prompted detailed kinetic calculations¹⁷⁹ to optimize laser geometries, input voltages, and electron temperatures and densities. These studies require as input the various electron-collisional cross sections in the energy range typical of the CVL environment, in which the electron temperature¹⁷⁹ is usually less than 5 eV.

Electron Collisions with Atomic Copper. We have demonstrated¹⁸⁰ that resonances involving the Cu⁻ ion dominate many of the excitation cross sections. In particular, the 3d¹⁰4s ²S^e-3d⁹4s² ²D^e cross section was found to be higher by an order of magnitude near threshold because of a resonance. This transition is responsible for the depopulation of the lower laser states, and is therefore important to achieving the maximum obtainable repetition rate.

Another important process in the CVL is the transfer of momentum from the electrons to the copper atoms and the buffer gas, thereby cooling the electrons and heating the gas. Our calculated ground state momentum-transfer cross section is three orders of magnitude higher than that formerly used in the kinetic models, again because of previously unknown negative-ion resonances.

The modeling codes that include these new cross sections now predict electron temperatures somewhat lower than in previous calculations, but still higher than measured values. We therefore calculated the electron-collisional excitation cross sections at a much higher level of detail. We developed a ten-state description of copper that involves, in addition to the four states used earlier, the lowest six states of the 3d⁹4s4p manifold. Using the new basis we have determined the cross sections for excitation to and between these six states. They are substantial, and are in fact higher than that of the ²S^e-²P^o laser-pumping transition. These inelastic processes, which have not been included in the kinetic models

Fig. 5-76. Total photodetachment cross section of Cu^- vs relative electron energy, calculated with two basis sets, both using the length and velocity forms of the dipole operator.¹⁸¹

until now, provide an important additional electron cooling mechanism. The amount of cooling per inelastic collision is large compared with the amount of cooling due to momentum transfer.

Photodetachment Cross Section of Cu^- . The population inversion necessary to the CVL is created by collisions between atomic copper and low-energy electrons produced in a discharge. The laser operates at two wavelengths, both in the visible, with corresponding transition energies of 2.14 and 2.43 eV. If a significant amount of Cu^- is produced in the CVL (through dissociative attachment of the dimer, radiative capture, etc.), then photodetachment of the Cu^- (electron affinity 1.23 eV) by the laser photons could result in an important loss mechanism that would degrade the laser efficiency.

Given the possible importance of photoabsorption by negative ions in the CVL and the possible resonant enhancement of the corresponding cross section, we undertook a theoretical study of this problem. The main results of our calculation are that (1) the photodetachment cross section of Cu^- exhibits a large, broad peak ($\sigma_{\text{max}} = 7.3 \times 10^{-17} \text{ cm}^2$ at $h\nu = 1.95 \text{ eV}$) due to a $3d^{10}4s4p^1\text{P}^o$ negative-ion shape resonance, and (2) that the cross sections at the two CVL wavelengths are substantial ($\sigma = 7.1 \times 10^{-17} \text{ cm}^2$ at $h\nu = 2.14 \text{ eV}$ and $\sigma = 6.8 \times 10^{-17} \text{ cm}^2$ at $h\nu = 2.34 \text{ eV}$). Figure 5-76 shows the total photodetachment

cross section, as calculated with the two basis sets mentioned above (in both the length and velocity forms¹⁸¹).

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Theory of the Multiphoton Ionization of Xenon by Short, Intense Laser Pulses

Introduction. Multiphoton ionization of atoms by intense laser fields has been the subject of active theoretical and experimental interest in recent years. Short-pulse lasers are capable of delivering single pulses with peak powers that produce rapid, multiple ionization of isolated atoms. Theoretical treatments of these phenomena require nonstandard approaches because the electromagnetic field strengths associated with the laser light can exceed the Coulomb fields in the atom, making traditional perturbation theoretical techniques inappropriate. Furthermore, the excitation and ionization processes take place on a time scale such that the temporal shape of the laser pulse can play a significant role in determining the final states produced, so that one must treat the time evolution of the field explicitly in any model of these phenomena. In many instances, for example, the atom will be ionized before the laser pulse has reached its maximum intensity.

Theoretical treatments of multiphoton ionization of atoms using perturbation theoretical techniques have explained most experimental observations. Only recently have pulse lengths become short enough and peak intensities high enough that the development of new theoretical methods has become necessary. We have found it convenient to use an explicit time-dependent picture¹⁸² to investigate the preionization dynamics of the atom, i.e., to understand how the laser energy is deposited in the atom. We have performed calculations to examine the multiphoton ionization of electrons from the outer shell of xenon.

We use a frozen, single-electron model of xenon¹⁸³ based on the time-dependent Hartree-Fock (TDHF) wave function for the system. We constructed an effective potential for the core electrons with a Hartree-Slater (local) approximation for the exchange terms. From this potential we calculate the ground state orbitals for the outer 5s and 5p electrons, then freeze all but one of these electrons. The free electron is allowed to interact with the laser field, and its time-dependent ionization probability (and therefore the ionization rate) is determined by explicitly solving the time-dependent Schrödinger equation for the electron wave function. The final state is determined by projecting the time-dependent wave function onto field-free states. We have examined the excitation and ionization dynamics of xenon for a number of laser wavelengths and intensities to obtain cross sections and rates for single- to eleven-photon ionization of the outer p electrons.

Our results illustrate multiphoton excitation and ionization for pulses of arbitrary length and intensity for a realistic, three-dimensional system without the complications of many electrons. These results will assist in the interpretation of the multielectron calculations that will follow.

Method. The frozen, single-electron model used in these calculations is an approximation to the TDHF model. We freeze the orbitals of all but one of the electrons. These remain fixed throughout the calculation. To derive the equations to be solved, we begin with the time-dependent Schrödinger equation for the atom, initially in its field-free ground electronic state and subjected to a time-varying laser pulse:

$$i \frac{\partial \Psi(r, t)}{\partial t} = (H_0 + V_1) \Psi(r, t), \quad (72)$$

where H_0 is the atomic Hamiltonian. The interaction term

$$V_1 = - \sum_i e E(t) z_i \sin \omega t \quad (73)$$

is obtained using the assumption that the laser field, linearly polarized in the z direction, can be treated classically and that the dipole approximation is valid. Both these assumptions are quite accurate for the laser intensities and wavelengths considered. We approximate the electronic wave function by an antisymmetrized product of single-particle orbitals. Putting this determinantal wave function into Eq. (72), we obtain the TDHF equations, a set of coupled partial differential equations for the time-dependent orbitals.

The TDHF equations are in fact too complex for numerical solution, because of the presence of nonlocal exchange terms. Because we must solve these equations thousands of times for a single calculation, we use the local free-electron-gas exchange potential of Slater.

We used the valence shell (5s and 5p) orbitals thus obtained to construct an effective core potential after making the orbitals nodeless. We used a single effective core potential, independent of angular momentum, in our calculations. Thus we obtain for the time evolution of the valence orbitals the equation

$$i \frac{\partial \phi_i(r, t)}{\partial t} = \left[-\frac{1}{2} \nabla^2 + V_{\text{rep}}(r) + V_{\text{Coul}}(r) + V_{\text{xc}}(r) + V_1(r) \right] \phi_i(r, t), \quad (74)$$

where $i = 5s$ and $5p$, and where the four potential-energy terms on the right-hand side correspond, respectively, to the effective core, Coulomb, exchange correlation, and matter-field interactions.

We use a finite-difference numerical technique to integrate these coupled partial differential equations. We chose a laser pulse whose electric field increases linearly over a preselected number of optical cycles and is then constant for several more cycles, so that an ionization rate for this orbital can be determined. If the chosen wavelength is far from resonance, we generally observe an exponential decay of the initial state component in the time-dependent orbital during the

constant-intensity period, and identify this with the rate for multiphoton ionization.

Results. To illustrate the method we present results for a range of peak laser intensities and for three laser wavelengths (1064, 586, and 293 nm). These wavelengths represent, in lowest-order perturbation theory, 11-, 6-, and 3-photon ionization, respectively. Figure 5-77 shows the calculated ionization rates for these wavelengths for laser intensities between 1×10^{13} and 2×10^{14} W/cm². Only for three-photon ionization at 293 nm is the ionization rate proportional to a power of the intensity, at least for the lower part of this range. All rates are saturated before intensities reach 1×10^{14} W/cm², indicating the onset of the

Fig. 5-77. Multiphoton ionization rates for the valence p shell of xenon vs intensity for 293, 586, and 1064 nm. Open symbols denote experimental results.

Fig. 5-78. Excited resonance state for 5p₀ initial state (586 nm, 3.5×10^{13} W/cm²). ▼

tunneling ionization mechanism and the breakdown of perturbation theory.

The open symbols in Fig. 5-77 indicate experimental ionization rates derived from reported saturation intensities: measurements by Perry et al.^{184,185} (293 nm, 1×10^{13} W/cm², and 586 nm, 4.2×10^{13} W/cm²); by l'Huillier et al.¹⁸⁶ (1064 nm, 1×10^{13} W/cm²); and by Freeman et al.¹⁸⁷ (1064 nm, 2.8×10^{13} W/cm²).

We obtain excellent agreement with all the measurements except that reported by l'Huillier et al.¹⁸⁶ The uncertainty in the absolute intensities obtained by these workers may be as high as 50%, which would lead to better agreement with our calculations and the other experimental result.

We found evidence in our calculations for the excitation of resonances. Figure 5-77 shows that the ionization rate for 586-nm light shows an interesting jump just above 3×10^{13} W/cm². Density plots of the time-dependent wave function indicate that a strong excitation of a temporarily bound, but highly excited state occurs at 3.5×10^{13} W/cm². Figure 5-78 shows a contour plot of the excited-state wave function for a particular time. Each successive contour corresponds to a change of one order of magnitude in the probability density. The nodal structure of the wave function indicates that it is dominated by a large $l = 3$ component; this results from a 6-photon excitation of an f orbital whose energy has been shifted into resonance by the dynamic Stark effect. As the intensity is increased further, this bound state tunes out of resonance again. We also find a resonant state playing an important role in the excitation process at intensities above 1×10^{14} W/cm² for 293-nm photons. In this case there is evidence of a slight inhibition of the ionization process arising from the presence of this resonant state. These calculations are the first to show these strong resonance effects at these high intensities.

Conclusions. We have calculated the multiphoton ionization of the outer valence shell of xenon by a linearly polarized laser beam in the frozen, single-particle approximation. Our results for 1- to 11-photon ionization agree quite

well with most measured rates. We also obtained excellent agreement with other theoretical results for the lower-order processes.

One important result from these calculations is that, for all wavelengths studied, the ionization out of the p-orbital aligned along the direction of polarization was much more rapid than for the other orbitals. At 1064 nm, the ionization rates per electron differed by more than a factor of 20. This means that the residual ion is predicted to be left in an oriented final state.

These calculation show that we now have a reliable tool for studying multi-photon ionization for wavelengths and intensities being used in the laboratory. Ionization rates and the effects of Stark-shifted intermediate resonant states show good agreement with experimental data.

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Charge-Transfer Cross Sections Important in Muon-Catalyzed Fusion

One novel direction in fusion research involves a fusion process using muons that takes place at modest temperatures. The process does not destroy the muon, so that it can catalyze hundreds of fusion events during its 2.2- μ s mean lifetime. This so-called muon-catalyzed fusion (MCF) is also called "cold fusion" because it occurs efficiently at the low temperatures at which atoms and molecules normally exist, and might thereby eliminate the need for high-temperature plasmas or powerful lasers for fusion power generation.

Muon-catalyzed fusion can be described by the following series of reactions:

The free muon leaving these reactions, in slowing down in the deuterium-tritium mixture, replaces the electron in a deuterium

atom to form a $d\mu$ mesic atom. In a subsequent collision the muon is transferred from the deuteron to a triton. The $t\mu$ mesic atom penetrates the electron cloud of a deuterium molecule and collides with a deuterium nucleus. The $t\mu$ and d^- resonantly combine within the D_2 molecule to form the mesomolecule $(dt\mu)^+$. This molecule is an analog of H_2^+ except that its bond length is smaller by a factor equal to the muon-electron mass ratio $m_\mu/m_e = 207$. This confines the deuteron and triton within a distance of 5×10^{-11} cm, which allows penetration of the Coulomb potential barrier; fusion therefore occurs within a time on the order of 10^{-12} s, with the usual 17.6-MeV energy release. The muon is available for next cycle of fusion reactions unless it is captured by the α particle. On the average it is emitted with kinetic energy equal to the Rydberg energy it had around the nucleus, about 1 keV.

This catalyzed process repeats either until the muon is captured by an alpha particle after fusion or until it undergoes spontaneous decay. Muons that stick to alpha particles after fusion are eliminated from the chain reaction unless they are stripped by collisions with other atoms (D or T); this is the principal loss mechanism in MCF. The initial sticking probability ω_s^0 is a crucial parameter because it determines the number of fusions that a single muon can induce.

How can the muon be stripped from the alpha particle? The $\alpha\mu$ ions are formed with an energy of 3.5 MeV and are slowed to thermal energies by collisions with surrounding D_2 and DT molecules. The $\alpha\mu$ ion can be stripped in such collisions as long as its kinetic energy exceeds the appropriate threshold ($E_{\alpha\mu}^T \approx 10.5$ keV). The stripping (charge transfer plus ionization) may occur through several channels. Collisions of the $(\alpha\mu)_s$ with DT and D_2 molecules can result in charge transfer, ionization, or excitation of discrete $\alpha\mu^*$ levels. Stripping may also occur from excited states of the $\alpha\mu$ ion that are formed in the sticking process or in the collisional excitation.

The effective sticking probability ω_s^{eff} is given by $\omega_s^{\text{eff}} = \omega_s^0(1 - R)$, where R (the probability of muon stripping by charge

transfer or ionization) is given by $1/\exp(-I)$; we have

$$I = \int_{v_0}^v \frac{v' \sigma_s(v') dv'}{mS(v')}, \quad (76)$$

where v_0 is the threshold for charge transfer, $\sigma_s(v)$ is the sum of the charge transfer and ionization cross sections for the $\alpha\mu$ ion, $S(v)$ is the stopping power, v and m are the velocity and mass of the $\alpha\mu$ ion, and $v_1 = 5.83$ atomic units (corresponding to $E = 3.5$ MeV) is the velocity at which the $\alpha\mu$ ion is created in the fusion reaction. Inasmuch as R depends

exponentially on I , accurate calculation of R requires very accurate evaluation of σ_s in the energy range $E_{\alpha\mu}^1 \sim E \sim 3.5$ MeV.

Collisional excitation processes are required to explain the density dependence of fusion rate constants. Cross sections for these processes are difficult to measure because the lifetime of muon is so short. One approach is to measure the cross sections for similar electronic systems and scale them by taking into account appropriate mass difference, threshold energy, momentum transfer, etc. Another approach is to determine them theoretically.

We have calculated charge-transfer cross sections using the first Born approximation with Coulomb boundary conditions that take the asymptotic interaction into full account.¹⁸⁸ We call this the boundary-corrected Born (BCB) approximation. In standard Born approximation calculations, plane wave functions are employed; this is less accurate for these processes in the intermediate- and high-energy regimes.¹⁸⁹ We have used our first-order theory successfully^{189,190} for purely hydrogen-like targets, as is the case in the present problem, with projectile charge Z_p ranging from 1 to 6.

With this theory we have calculated state-to-state and total cross sections for muon transfer from $\alpha\mu_{1s}$ to $H^+(544)$ in the laboratory impact-energy range $E = 25$ to 1000 keV. [$H^+(544) = H(nlm = 100, \dots, 544)$]. Here "total" represents the cross sections summed over all bound states from the $H^+(544)$ calculation using the well-known Oppenheimer scaling law $\sigma_{tot} = \sigma_1 + \sigma_2 + \sigma_3 + \sigma_4 + 3.049\sigma_5$.

Our computed cross sections (Fig. 5-79) are in good agreement with the experimental data of Rinn et al.¹⁹¹ and Watts et al.¹⁹² in the energy range 50 to 180 keV. All scalings referred to in the figure are according to our exact scaling formula.¹⁸⁸ Beyond 180 keV neither experimental nor quantum coupled-channel, atomic basis¹⁹³ results are available, so the absolute accuracy of our results cannot be established. However, because our results are in good agreement with experimental results in the intermediate-energy range and because the Born approximation

Fig. 5-79. Scaled¹⁸⁸ charge-transfer cross sections for the reaction $\alpha\mu^+ + p \rightarrow \alpha + \mu$ vs proton impact energy E (laboratory system). Experimental data from Rinn et al.¹⁹¹ and Watts et al.¹⁹²; CTMC calculations from Cohen.¹⁹³

is a high-energy approximation, we believe that the accuracy of our results should be excellent in the high-energy regime. The results of Cohen,¹⁹⁴ obtained with the classical trajectory Monte Carlo (CTMC) method, agree less well with the scaled experimental data.

These charge-transfer cross sections can be used in kinetic models to predict the effective stripping probabilities.

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Circular Dichroism in Photoelectron Angular Distributions

Circular dichroism (CD) is a phenomenon in which a system responds differently when interacting with left-circularly polarized (lcp) or right-circularly polarized (rcp) light. Traditionally, CD has been observed by means of differential absorption of lcp and rcp light passing through solutions containing dissolved chiral molecules. The differential absorption arises because of interference between electric dipole (E1) and magnetic dipole (M1) interactions and demands intrinsic chirality of the system. The magnitude of the effect is therefore small.

Photoelectron angular distributions resulting from single- or multiple-photon ionization of oriented systems can exhibit dichroic behavior.¹⁹⁵ This circular dichroism in angular distributions (CDAD) exists in the electric dipole approximation and requires no intrinsic chirality and no spin-orbit coupling. The effect is therefore observable in atomic and molecular systems in the gas phase. The alignment necessary for observing CDAD can be created in a variety of ways, such as adsorption of atoms or molecules on surfaces, photoabsorption, photodissociation, or collisions. The left-right symmetry is broken by a careful choice of the directions of alignment, circularly polarized light propagation, and photoelectron detection (Fig. 5-80).

The vectors for lcp and rcp light are related by reflection in a plane containing the propagation direction. If the system is symmetric under this reflection,

Fig. 5-80. Experimental arrangement for CDAD measurement.

there will be no dichroic behavior. The total (angle-integrated) ionization signal therefore does not exhibit dichroic behavior. If one measures photoelectrons ejected at a particular angle, however, the left-right symmetry can be broken. Figure 5-80 makes it clear that the measurement of angular distributions in the plane containing the propagation direction and the alignment axis will exhibit no dichroic behavior, whereas the angular distribution out of this plane will exhibit dichroic behavior. The CDAD signal is maximum in a direction perpendicular to this plane.

We have performed *ab initio* calculations for photoionization of adsorbed atoms and diatomic molecular systems. These calculations show that the magnitude of the CDAD signal (measured as a difference in lcp and rcp angle-resolved cross sections) is comparable to the photoionization cross section itself.

The existence of CDAD has been verified.¹⁹⁶ Nitric oxide molecules, excited with linearly polarized light from various rotational states of the ground electronic ($^2\Pi$) state to the $A^2\Sigma^+$ state, were ionized by lcp or rcp light. The exciting and ionizing beams were co-propagating; the angular distributions were collected in a plane perpendicular to the propagation direction by rotating the polarization direction of the linearly polarized light. The ro-vibrational states of NO are unaligned in the gas phase. The absorption of photons in the transition to the $A^2\Sigma^+$ state creates an alignment in the $A^2\Sigma^+$ ro-vibrational states. Subsequent

Fig. 5-81. CDAD signal from the photoionization of oriented NO. The angle θ_k (Fig. 5-80) is that between the direction of polarization and the electron detectors.

ionization of these aligned states with lcp or rcp light exhibits CDAD.

Figure 5-81 shows a representative set of CDAD measurements for single-photon excitation and single-photon ionization ("1 + 1" measurements) and illustrates

their sensitivity to the alignment. The alignment parameters extracted by fitting the data to theoretical expressions describing the CDAD signal are in good agreement with our calculations.¹⁹⁶

CDAD is a clear signature of the existence of alignment. This raises the possibility of the use of CDAD to probe initial-state alignment. It would seem that the extraction of the alignment parameters from CDAD measurements would require prior knowledge of photoionization matrix elements and phase shifts, but this is not so. In fact, we have shown¹⁹⁵ that, for $(n + 1)$ CDAD experiments (n linearly polarized exciting photons, one lcp or rcp photon), the ratio of CDAD signals for two branches starting from a given initial state removes all the dependence on the photoionization matrix elements. The deduction of initial-state alignment from CDAD involves algebraic numbers that depend on the rotational quantum numbers only. This simplification makes the technique very attractive as a probe of state alignment. Experiments using this technique are under way to measure the alignment of NO created by photodissociation of methyl nitrite.

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High-Average-Power Solid State Lasers

6

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High-Average-Power Solid State Lasers

Introduction

J. Z. Holtz

In 1987 we continued a broad-based, aggressive R&D program aimed at developing the technologies necessary to make possible the use of solid state lasers that are capable of delivering medium- to high-average power in new and demanding applications. Our efforts were focused along the following major lines: development of laser and nonlinear optical materials, and of coatings for parasitic suppression and evanescent wave control; development of computational design tools; verification of computational models on thoroughly instrumented test beds; and application of selected aspects of this technology to specific missions.

In the laser materials area, our efforts were directed towards producing strong, low-loss laser glasses and large, high-quality garnet crystals. We worked with the major laser glass companies (Schott and Hoya) to develop compositions tailored for high-average-power operation. This work produced high optical quality glasses that are free of platinum inclusions and whose absorption loss is about $3 \times 10^{-3} \text{ cm}^{-1}$ in large castings and an order of magnitude lower in smaller castings. We have reduced to practice surface finishing and surface ion exchange techniques that increase the surface tensile strengths of these materials by factors of 5 to 10.

Our crystal program consisted of computational and experimental efforts aimed at understanding the physics, thermodynamics, and chemistry of large garnet crystal growth, and of experimental programs at Allied Signal Corp. dedicated to the growth of Nd:YAG and Nd:GSGG. These efforts have improved our understanding of the Czochralski crystal growth process and of its limits. Significant progress was made in growing flat-interface Nd:YAG, but cracking of large boules remains a problem. A number of large boules of Nd:GSGG were grown, and we are now concentrating on reducing the absorption loss in these crystals. A significant outgrowth of this research is the development of the moving melt zone technique for large crystal growth. This research effort, now supported in part by funds from the Laboratory's Institutional Research and Development Program, is proceeding rapidly.

Our laser experimental efforts were directed at understanding thermally induced wave front aberrations in zig-zag slabs, understanding fluid mechanics, heat transfer, and optical interactions in gas-cooled slabs, and conducting critical test-bed experiments with various electro-optic switch geometries. We now have a clear understanding of most of the physics and engineering issues involved and will integrate many of these components into an operating laser in 1988.

We have begun development of a solid state laser, using this technology, for submarine laser communication. The approach described in this section is that of designing and testing a semiconductor laser-pumped solid state laser that is conveniently frequency doubled to efficiently

obtain the required laser wavelengths. Development tasks include spectroscopic studies of a variety of rare earth ion-doped crystal and glass hosts, development of suitable semiconductor diode array pump sources, laser characterization studies, and modeling satellite system cost sensitivities as determined by laser transmitter performance for a variety of space-based applications.

Laser Materials Development

Laser Glasses

Nd:Glass Development. Elimination of platinum inclusions in phosphate glass is a critical step in the realization of full pulse energy on Nova. A central focus for our research in Nd:glass for high-average-power (HAP) lasers is the extension of platinum-free melting technology to the glasses whose properties are optimized for repetitive-pulse operation.

While under contract to LLNL during 1984-85, Hoya Optics and Schott Glass Technologies developed phosphate glasses with improved thermomechanical and laser properties. Hoya Optics continued to develop HAP-3, a silico-phosphate glass with about 2.5 times greater thermal stress resistance than the high-cross-section glasses used on Nova.

Platinum-inclusion-free melting of HAP-3, a co-funded effort between LLNL and Hoya Optics, demonstrated that low solubility of platinum in silico-phosphate glasses prevents routine platinum-free melting. Hoya Optics reduced the platinum-inclusion density to yield only a few Pt-free slabs ($30 \times 10 \times 1$ cm) out of twenty melts of 5 L each. Since this poor yield is unacceptable, we are renewing a composition study to identify better candidate compositions for Pt-inclusion-free melting.

This year, we reinitiated the compositional research at Schott Glass Technologies. Our specific goal was to achieve Pt-free glass (using the existing melting technology) with laser and thermomechanical properties equivalent to those of HAP-3. Following 70 test melts (0.5 L each), three promising glasses were tested in 2-L melting experiments. We chose advanced phosphate glass (APG-1)

for scaling to 40-L castings. Three castings ($80 \times 25 \times 6$ cm each) were made in October 1987. These were Pt-free, had good thermomechanical and laser properties, and were of good optical quality. Further melting refinements are required to lower the optical absorption at the laser wavelength. The current status of APG-1 is adequate for laser amplifier demonstrations.

Author: J. E. Marion

Strengthening of Laser Slabs. During operation, a steady state thermal gradient develops in amplifier slabs that gives tensile stresses on the slab surfaces approaching or exceeding the slab strength. Fracture of the slabs is a major limitation to operation at high average power.¹

We adopted a protocol for estimating the actual strength of slabs based on a Weibull statistical treatment.² Although our unexpected failures are more easily understood when we use this approach, our laser designs are still limited by the modest strength of the slabs. For that reason we now focus on improving slab strength so as to increase power output.

The thermal gradient and tensile surface stress for a given set of laser operation parameters depend on the thermomechanical properties of the amplifier material. In our first approach, we improved the power output by developing better materials, including single crystals and superior glass compositions. In our second approach, we improved the optical fabrication process for producing the slabs to reduce the severity of the resultant surface damage, which limits the strength. In our third approach, we induced surface compressive stresses through chemical ion exchange of the finished slabs to deactivate the microcracks created during optical fabrication. The tremendous potential of each of these methods has been demonstrated

on small samples, and each technique is now being applied separately to laser slabs for verification in the laser test bed.

The prospects for substantial improvements in the thermomechanical properties of laser glasses seem limited since each parameter (such as the thermal conductivity, thermal expansion, and fracture toughness) only varies within a fairly limited range. Nonetheless, our efforts with Schott Glass Technologies and Hoya Optics produced glasses that have laser properties similar to the previously available compositions, but have 2.5 times better thermomechanical properties, giving us a 250% increase in power output potential. We achieved this impressive advance by combining modest improvements in individual parameters. Recently, large castings were produced so that these new glasses can now be implemented into the laser test bed.

The second area of emphasis is in the reduction of the strength-limiting microcracks that occur in the slab surface as a result of optical fabrication. Small sample tests show that when we eliminate cracks by acid etching,³ strength increases over 100 times. Extraordinary strengths, approaching the theoretical limit, have been measured on glass and single crystal samples. Unfortunately, acid etching of the optical surfaces destroys the required flat surfaces. We used an alternative method to reduce the damage, large amounts of material are removed at each successively finer grinding and polishing step to eliminate the damage from the previous step.³ In small samples, strength increases tenfold. Using the improved finishing method, we demonstrated a twofold increase in the strength of our first full-sized slabs. With further work we expect fourfold to eightfold strength increases on full-sized slabs.

The third emphasis in our strengthening research is the deactivation of surface cracks by inducing a compressive stress field at the surface. A variety of methods to achieve surface compressive stresses are used in industry (e.g., thermal tempering of car windshields), but these all have the same limitation as acid

etching: inadequate surface quality after strengthening.

One technique, chemical ion exchange, shows promise. In this process, the slabs are treated in a salt bath in which small ions are leached from the surface of the glass and exchanged with larger ions. The result is a compressive layer at the surface. Preliminary fracture tests of ion-exchange-strengthened full-scale slabs showed a twofold improvement over nontreated slabs. Once this process is perfected, we expect that we will be able to induce a 200-MPa stress layer in the glasses with the best thermomechanical properties. This represents about a fourfold strengthening compared with untreated slabs.

We expect a tenfold improvement in power output by the use of new glasses with 2.5 times better intrinsic properties, a 200-MPa compressive layer, and a factor-of-2.5 improvement in strength by reducing surface damage.

Author: J. E. Marion

Quantum Efficiency and Relaxation Rate Measurements. The radiative and nonradiative relaxation rates of the Nd^{3+} upper laser level are required in the design codes for high-average-power laser devices. They are used to determine

Fig. 6-1. Radiative decay. Spontaneous emission of photons accompanies transitions from the $^4F_{3/2}$ (R) metastable state to lower lying energy levels.

Fig. 6-2. Multi-phonon emission. An isolated Nd^{3+} ion in the ${}^4\text{F}_{3/2}$ (R) upper laser level decays through interaction with the host material, producing high energy phonons.

Fig. 6-3. Cross relaxation. An excited Nd^{3+} ion shares its energy with a neighboring Nd^{3+} ion.

model energy storage times, laser efficiencies, and thermal energy deposition rates. To determine the radiative and nonradiative contributions to the excited state relaxation rate (without reliance on model calculations), two independent measurements are needed. For this purpose we measured the quantum efficiencies and the fluorescence decay rates of a large number of samples of laser glass.⁴⁻⁶

Figure 6-1 shows radiative decay, one of the four major relaxation processes for neodymium-doped laser materials. Figure 6-2 shows multiphonon emission, a nonradiative process that results from an interaction between isolated Nd^{3+} ions and the material. It has a decay rate that is independent of neodymium concentration. Figures 6-3 and 6-4 show how the rate of nonradiative relaxation increases with increasing neodymium concentration through ion-ion interactions.

The need for absolute measurement of the nonradiative relaxation rates was recently indicated when Mangir and Rockwell⁷ reported anomalously high heat generation in the process of flash-lamp pumping several phosphate laser glasses. They suggested that their results could be explained if the quantum efficiency was significantly lower than previously thought, thus implying that high nonradiative relaxation rates were responsible. The work performed here did not provide a completely satisfactory explanation for the observations of Mangir and Rockwell. However, the emission cross sections obtained from analysis of our experiments are up to 25% smaller than those used by Mangir and Rockwell to determine excited-state densities. Use of the lower cross sections and the higher nonradiative decay rates measured in our work would decrease the apparent discrepancy between their theory and experiment from a factor of 2 to about 1.5. The residual discrepancies could be caused by other unidentified artifacts in their experiments.

Experiments. Figure 6-5 shows the quantum efficiency measurement apparatus (Refs. 4-6) using a radiometrically calibrated integrating-sphere detection system. An argon ion laser operating at 514.5 nm was used to excite small samples placed on a pedestal at the center of the integrating sphere. Emissions in the ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{9/2}$ (R $\rightarrow$ Z) and ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ (R $\rightarrow$ Y) bands of Nd^{3+} were detected. The Judd-Ofelt theory⁴ was used to correct for the small amount (10%) of radiative emission that occurs in the ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{13/2}$ (R $\rightarrow$ X) and ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{15/2}$ (R $\rightarrow$ W)

bands at wavelengths where the S-1 photocathode is not sensitive.

Room temperature fluorescence decay rates were obtained from the samples in separate experiments. A pulsed dye laser was used for excitation at 580 nm. Fluorescence decay waveforms were recorded with a transient digitizer and analyzed by standard numerical techniques. An analytical method⁴ to correct the measured data for the effects of radiation trapping was also developed.

Quantum efficiency and fluorescence decay rate measurements were made on samples of the phosphate glasses LG-750, LG-760, LHG-5, LHG-8, Q-88, and Q-98, the silicate glass ED-2, and Nd:YAG. Figure 6-6 shows the quantum efficiencies obtained from samples of LG-750 and LG-760 plotted as a function of neodymium concentration. The solid curves shown in the figure are the results of calculations based on the predictive model discussed below.^{5,6}

Analysis of Relaxation Rates and Emission Cross Sections. For systems exhibiting single exponential decay, the fluorescence quantum efficiency η_f is equal to the ratio of the spontaneous photon emission rate (radiative decay rate) A_r to the total decay rate W_t , i.e.,

$$\eta_f = \frac{A_r}{W_t} \quad (1)$$

At low Nd^{3+} concentrations, the Nd^{3+} fluorescence decay in the phosphate glasses and in YAG is almost purely exponential; it is possible to determine the radiative decay rate from the measured fluorescence decay rate and quantum efficiency using

$$A_r = \eta_f W_t \quad (2)$$

The nonradiative decay rate W_{nr} can be defined as the difference between the

Fig. 6-4. Excitation migration. The excitation energy moves (hops) from one Nd^{3+} to another.

Fig. 6-5. Integrating-sphere quantum efficiency measurement apparatus. A rotatable mirror directs the source to the sample in sphere or to the thermopile power monitor.

Fig. 6-6. Intrinsic fluorescence quantum efficiencies for LG-750 and LG-760 samples plotted vs neodymium concentration. The solid curves are calculated values.¹

fluorescence decay rate and the radiative rate, and therefore

$$W_{nr} = (1 - \eta_f) W_r. \quad (3)$$

Equations (2) and (3) were used to obtain experimental values of the radiative and nonradiative decay rates for each sample.⁵ Radiative rates obtained by this method were consistently somewhat lower than those predicted by the Judd-Ofelt theory.⁸⁻¹⁰ The experimental radiative decay rates and the emission line shape functions $g(\lambda)$ were used to determine the cross section for stimulated σ^{em} emission as a function of wavelength λ using the relation^{4,10}

$$\sigma^{em}(\lambda) = \frac{\lambda^4 A_r}{8\pi c n^2} g(\lambda), \quad (4)$$

where n is the host material's index of refraction. The lower values for A_r yielded emission cross sections that were less than those based on the Judd-Ofelt theory by up to 25% (Refs. 4 and 5).

Analysis of Nonradiative Fluorescence Quenching in LG-750 and LG-760. For LG-750 and LG-760, the decrease in

quantum efficiency with increasing Nd^{3+} concentration was accurately described by a model in which the critical range for nonradiative energy transfer is given by^{4,11,12}

$$R_{DX}^6 = \frac{3c}{8\pi^4 n^2 A_r} \int \sigma_D^{em}(\lambda) \sigma_X^{abs}(\lambda) d\lambda, \quad (5)$$

where $X = D$ or A for donor-donor (Fig. 6-4) or donor-acceptor (Fig. 6-3) transfer, respectively, and σ_X^{em} and σ_X^{abs} are the emission and absorption cross sections for species X , respectively.

The measured $Z \rightarrow R$ absorption spectra and $R \rightarrow Z$ emission spectra were used in Eq. (5) to calculate the values $R_{DD} = 11.14 \text{ \AA}$ for LG-750, and $11.37 \text{ \AA}$ for LG-760 (Ref. 4). A similar calculation for the cross relaxation process using the $R \rightarrow W$ emission spectrum¹³ and the $Z \rightarrow W$ absorption spectrum, yielded $R_{DA} = 4.07 \text{ \AA}$ for LG-750. Because the $R \rightarrow W$ emission spectrum has not been measured for LG-760, it was not possible to determine R_{DA} for LG-760 by this method.

For LG-750, a zero concentration non-radiative relaxation rate (Fig. 6-2) of $W_0 = 216 \pm 150 \text{ s}^{-1}$ was obtained by a least squares fit between the quantum efficiencies predicted by our analytical model¹ and the measured values. The model described in Ref. 4 was then used to calculate the quantum efficiency as a function of Nd^{3+} concentration, using the parameters listed in Table 6-1. Figure 6-6 shows the results plotted as a solid curve. The agreement between the theoretical model and the experimental results is excellent.

For LG-760, it was necessary to determine both R_{DA} and W_0 by a least squares fit, as indicated in Table 6-1. Model calculations for LG-760 are also plotted in Fig. 6-6. The data indicate that there is weaker concentration quenching in LG-760 that arises from a smaller critical range for cross relaxation R_{DA} .

The samples of LG-750 and LG-760 used in this study generally had absorption coefficients on the order of 1 cm^{-1} at $3.5 \text{ \mu m}$ (attributable to water

contamination). This level of contamination can produce a nonradiative quenching rate on the order of 60 s^{-1} at low neodymium concentrations.¹⁴ This rate is roughly the same as the lower limit of experimental uncertainty in the values of W_0 shown in Table 6-1. The observed rate W_0 is actually the sum of rates due to multiphonon emission and impurity (e.g., water) quenching. Therefore, the upper limits of experimental uncertainty in W_0 represent an upper bound of about 350 s^{-1} on the actual multiphonon rate, and no lower bound can be accurately determined from the present results.

Conclusion. Measurements of quantum efficiencies and fluorescence decay rates were combined to determine both spontaneous radiative emission and nonradiative relaxation rates. The experimentally determined spontaneous emission rates and calibrated emission line shape functions were used to obtain cross sections for stimulated emission as a function of wavelength. Generally, good agreement was found between the measured radiative properties and the predictions of the Judd-Ofelt theory, although the theory appears to consistently overestimate the radiative emission rates and consequently the emission cross sections in all of the glasses by as much as 25%.

For both LG-750 and LG-760, the non-radiative relaxation rate at low Nd^{3+} concentrations was found to be $210 \pm 150 \text{ s}^{-1}$. Since the level of water contamination in these samples could account for roughly 60 s^{-1} of the observed rate, the results can only be used to establish an upper bound of about 350 s^{-1} on the multiphonon decay rate. Nevertheless, this is almost an order of magnitude lower than extrapolation from data on other ions might indicate.^{15,16}

The effects of concentration quenching were shown to be accurately described by Förster-Dexter^{11,12} nonradiative energy transfer theory when the hopping model of Burshtein^{17,18} is used to describe cross relaxation made possible by migration of the excitation energy. It was shown that the essential nonradiative energy transfer parameters can be determined from

Table 6-1. Intrinsic spectroscopic and energy transfer parameters for LG-750 and LG-760.

Parameter	LG-750	LG-760
Radiative decay rate, $A_r (\text{s}^{-1})$	2420	2560
Radiative lifetime, $\tau_r (\mu\text{s})$	414 ± 20	390 ± 20
Peak emission cross section, $\sigma_e (10^{-20} \text{ cm}^2)$	3.44 ± 0.2	4.14 ± 0.3
Index of refraction, $n (1050 \text{ nm})$	1.516	1.508
Donor-donor critical transfer range, $R_{DD} (\text{\AA})$	11.14	11.37
Cross relaxation critical range (Donor-acceptor), $R_{DA} (\text{\AA})$	4.07	3.52 (fit)
Zero concentration nonradiative rate, $W_0 (\text{s}^{-1})$	216 ± 150 (fit)	209 ± 150 (fit)

calibrated absorption and emission spectra. Such tools can be invaluable in the analysis of new laser materials when we need to determine whether quenching is intrinsic or is caused by a defect that can be eliminated through improved material synthesis.

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Laser Crystals

Nd:Cr:GSGG Growth. Neodymium:chromium:gadolinium scandium gallium garnet (Nd:Cr:GSGG) is among the best materials available for high-average-power (HAP) laser operation because of its exceptional laser efficiency. We developed the growth of large crystals of this material under a contract with the Allied Signal Corporation. Our goal was stable growth of $>10\text{-cm}$ -diam by $>20\text{-cm}$ long crystals and low absorption at the laser wavelength. Divalent calcium, Ca^{2+} , added to the melt promoted stable growth, but gave high absorption. Unfortunately, absorption ranges from about $1\% \text{ cm}^{-1}$ at the top of the crystals to about $6\% \text{ cm}^{-1}$ at the bottom (Fig. 6-7). For efficient laser operation, the loss must be further reduced to 0.5 to $1\% \text{ cm}^{-1}$ over the entire length of crystal.

Currently, we have discontinued the growth of Nd:Cr:GSGG. Our present inventory includes four boules, all grown with Ca and Ce additions, which are

Fig. 6-7. Schematic of Cr^{4+} loss profile at $1.06\ \mu\text{m}$ in typical Nd:Cr:GSGG crystals grown to date.

Fig. 6-8. Cr^{4+} absorption spectra.

available for laser experiments. The largest boule is 96 mm in diameter by 194 mm long. After cutting these boules into plates, we have four plates that are $5 \times 80 \times 194$ mm or larger and six plates that are $5 \times 50 \times 150$ mm or larger. One smaller plate from an earlier boule was fabricated into a zig-zag laser slab measuring $5 \times 50 \times 90$ mm. After heat treatment in a reducing atmosphere (1000°C , 6% H_2 in Ar, 12 hr), the slab had a total loss of about $1.5\% \text{ cm}^{-1}$, which is higher than our requirements. If reducing heat treatments can be perfected, we will have a sufficient GSGG inventory to build a laser amplifier head to demonstrate the potential of Nd:Cr:GSGG for efficient HAP laser operation.

One-Micron Absorption in Nd:Cr:GSGG.

We believe that the absorption in the GSGG grown under contract at Allied Signal Corporation is attributable to the

Cr^{4+} that occupies fourfold coordination sites in the garnet lattice. During the three years of this contract, we scaled GSGG crystal growth to large boule diameters. This scaling decreased the thermal gradients at the melt/crystal growth interface that led to growth instabilities and resulted in spiral growth morphology. Although Ca^{2+} additions to the melt controlled the spiraling, uncompensated Ca^{2+} ions and a slightly oxidizing growth atmosphere (necessary to prevent volatilization of gallium suboxide) induced the formation of undesirable Cr^{4+} .

To alleviate this problem, cerium oxide was added to the melts. Because of its electrochemistry, Ce changes its valence state from $3+$ to $4+$ (to compensate Ca^{2+}) more readily than Cr. Cerium in either valence state does not contribute to an absorptive loss at the laser lines or pump bands of GSGG. The addition of Ce resulted in material with losses $<10\% \text{ cm}^{-1}$, a tenfold improvement over previous boules. Figure 6-7 shows a representative Cr^{4+} loss profile at $1.06\ \mu\text{m}$. The spatial distribution of loss in GSGG depends on the segregation coefficients of the various dopants and on the annealing effects imposed by the growth atmosphere as the boules are grown.

Two spectral absorption features attributable to Cr^{4+} were seen in Ce:Cr:Nd:GSGG. We denote the primary center as CR4, which consists of a double peak in the near-IR region with accompanying bands in the red and blue-green spectral regions. CR4 absorptions can be intermixed with absorptions from a second center (denoted as CR4'), which is believed to arise from energy perturbations of Cr^{4+} due to point defect sites. Figure 6-8 shows absorption spectra for each center. Although CR4' also absorbs at the laser band, its peak is shifted further towards the IR and thus results in lower loss than CR4. Exposure to blue photons at room temperature transforms CR4 into CR4'; the reverse transformation occurs upon heating above about 375°C . Both CR4 and CR4' occupy one of the tetrahedrally coordinated sites in GSGG. Since these sites lack a center of symmetry, the intensity of transitions is

very high, such that even minute amounts of Cr^{1+} result in relatively strong absorption and high loss.

To further improve the material, we must subject the slabs to conditions and chemical reactions that eliminate or reduce the residual Cr^{1+} . The most effective results have been obtained by annealing small samples for 12 hr at 1300°C in a platinum/iridium tube furnace with a reducing atmosphere of a 1:1 ratio by volume of CO and CO_2 . The absorption loss in samples treated in this manner were studied using spectrophotometry, calorimetry, and loss meter measurements. All three methods indicate that the overall loss is less than 0.5% cm.

Scaling the heat treatments to the larger slabs has been difficult. While the absorption losses can be decreased by this approach, treatments on large samples often resulted in high losses due to creation of fine scattering sites or decorated dislocations. The treatment parameters that worked well on earlier samples treated in a platinum/iridium tube furnace were ineffective in decreasing the $1\text{-}\mu\text{m}$ absorption when used in an alumina tube furnace. This may have occurred because impurities in the alumina have increased the partial oxygen pressure of the treatment gas. Treatments in dilute hydrogen atmospheres yielded better results. A slab treated in 0.5% H_2 in Ar for 24 hr in a tungsten-lined furnace was operated as a laser. However, residual $1\text{-}\mu\text{m}$ loss is inhomogeneously distributed; the highest losses are near the surfaces. Figure 6-9 shows a loss profile for this slab.

We are uncertain as to how the scattering sites are formed. We believe that complete containment of the samples within a platinum or platinum/iridium tube furnace is probably an important element of the heat treatment process. Platinum may pass hydrogen from the ambient atmosphere while filtering out oxidizing gasses. Hydrogen is expected to form hydroxyl ions that participate in reducing Cr to the desirable trivalent state. Extraneous materials in the vessel (such as alumina or graphite) may lead to scattering and poor reduction in absorption. The scattering may arise from volatile impurities that contaminate the sample surfaces. Grain boundaries and

Fig. 6-9. Schematic of loss profile extrapolation for $50 \times 90\text{-mm}$ slab that was tested as a laser.

dislocations then provide ready channels for propagation of scatterers to the sample interior. A further possibility is that scattering sites form during heating or cooling through a critical temperature range. Our current research focuses on identifying appropriate heat treatment processes to reduce the loss to below $1\% \text{ cm}^{-1}$ in the large slabs. Then these slabs can be used for laser operation.

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Nd:YAG Growth. Allied-Signal, Inc. of Morristown, N.J. continues its work on the growth of flat-interface, neodymium-doped yttrium aluminum garnet (Nd:YAG) crystals for the Free Electron Laser (FEL) Program. Although advances have been made on a continuous pellet-feed method, some problems still remain. A more important problem is growing large-diameter, flat-interface boules that do not crack.

The master oscillator for the FEL Program requires a high-repetition rate, $\sim 1\text{-kHz}$ laser of moderate energy per pulse. The material of choice for this application is Nd:YAG because of its high emission cross section and good thermo-mechanical properties. Current plans call for zig-zag laser plates with $0.5 \times 8 \times 21 \text{ cm}$ of high optical quality. Advances in Nd:YAG crystal-growth technique are required in order to grow the 8.2-cm diam, core-free boules from which the plates can be fabricated.

Experimentation at Allied-Signal is directed toward defining the conditions required to grow core-free, flat-interface Nd:YAG. A second objective is to develop

a continuous melt-replenishment system that will maintain the proper neodymium concentration over the length of the crystal and will permit the growth of longer crystals.

The flat-interface, Czochralski (CZ) crystal-growth process currently under development uses crystal rotation rate to control interface shape. In conventional Nd:YAG growth, the crystal grows with a deep, convex interface having internal

Fig. 6-10. The schematic of the pellet-feed system shows a pellet falling down the standpipe.

Fig. 6-11. The cross section of a 3-cm Nd:YAG boule as seen through crossed polarizers shows the transition from deep to flat interface.

(211) facets. These facets cause an undesirable central core of highly stressed material. Thus, the optically usable width of a plate fabricated from such a boule is reduced to substantially less than half the boule diameter (usually about one-third). This requires growing a large, deep-interface boule of about 24 cm diam. Maximum diameter in commercial production is currently limited to 7.5 cm because of the increased tendency of large crystals to crack.

If we grow the crystal with only a slightly convex interface, the (211) facets do not form, and the full diameter of the boule may be used to fabricate plates. Increasing crystal rotation rate causes the fluid in the region of the growing interface to become more isothermal by convective heat transfer. (Note that the growth interface shape is defined by the melting point isotherm.) The transition from deep to flat interface occurs at rotation rates of 25 to 55 rpm, depending on boule diameter, crucible size, and furnace configuration. The transition must be made smoothly without introducing crystal defects (such as bubbles and dislocations) that will distort the laser beam and reduce crystal strength. Ultimately these defects can cause the crystal to crack. A further complication is introduced by neodymium, which may cause the interface to become unstable by a phenomenon called constitutional supercooling.¹⁹ Higher rotation and lower pull rates reduce this effect.

Allied-Signal established three CZ growth stations devoted exclusively to growing Nd:YAG. An uninterruptable power supply capable of supplying 120 kW for 20 min was installed. A digital computer control system that uses proportional, integral, and derivative (PID) diameter-error feedback to control furnace power was developed. A parallel-path experimental approach separately addressed the problems of continuous feed and flat interface. For continuous feed, 3-cm-diam crystals were grown from 7.6-cm-diam crucibles; for flat interface, 8.2-cm crystals were grown from 15.2-cm crucibles.

Figure 6-10 is a schematic of the pellet-feed system. Cylindrical Nd-doped pellets (5, 7, 14 mm long and 4 mm diam)

were dropped into a standpipe at a frequency sufficient to maintain the melt level in the crucible. The standpipe is located close to the crucible wall and fitted with a porous iridium cap. Although the pellet-feed technique maintained Nd concentration and melt level, undesirable micron-sized iridium inclusions were introduced into the growing crystal. These inclusions cannot be tolerated because they increase loss and become damage sites during pumping of the zig-zag laser crystal. It may be possible to contain these small iridium particles within the standpipe by reducing the porosity of the plug in the end. An alternative scheme of feeding Nd:YAG rod instead of pellets may eliminate the iridium inclusion problem by avoiding thermal shock to the melt.

Figure 6-11 is a polariscope photograph illustrating the interface shape along the axis of a 3-cm boule. The transition from deep to flat is continuous and occurs about one diameter length below the shoulder of the cone. The interface is flat for the remainder of the boule. Although some success was achieved growing flat-interface, core-free, 8.2-cm-diam Nd:YAG, all the boules cracked. The most difficult aspect of this task is performing the transition from deep to flat interface without introducing defects. We believe that boule cracking is caused by these defects.

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Large-Dimension Czochralski Crystal Growth.

We continued to study the growth of large-dimension garnets for laser hosts by the Czochralski (CZ) crystal-growth process. This culminated in the growth of gadolinium gallium garnet (GGG) crystals weighing up to 17 kg and measuring up to 23 cm long at a full diameter of 11 cm. The LLNL CZ growth station is a state-of-the-art research facility, capable of growing many large-dimension crystalline materials.

Our extensive experimental measurements and computational modeling (see "Czochralski Crystal Growth Modeling" later in this section) paid off this year in a demonstration of the growth of large-dimension laser crystals. We chose GGG

and neodymium-doped GGG as our test materials instead of gadolinium scandium gallium garnet (GSGG) because they are only slightly more difficult to grow and are a factor of 6 less expensive and faster growing.

The crystal-growth facilities and equipment as well as the CZ process are described in detail in Sec. 6 of the 1986 Laser Annual. Briefly, the blended GGG powder materials are isostatically compressed and loaded into an iridium crucible that is heated to the melting point of GGG (1750°C) by inductive coupling. A GGG single crystal grows from a rotating GGG seed that is lowered to the surface of the melt and slowly pulled. The crystal is weighed, and the diameter is calculated from its rate of weight increase and the pull rate. This value is compared with the desired diameter and is controlled by PID feedback to the inductive heating power.

Table 6-2 summarizes the results of the crystals grown during the year. We grew ten crystals—nine GGG and one Nd:GGG. The pull rates for GGG and Nd:GGG were 6 and 1 mm/hr, respectively. A constant rotation rate of 12 rpm was used for runs 3 to 7. All of these crystals grew with a deep, concave interface and cracked either during growth or during cool down. Crystals from runs 8 to 14 were grown by increasing the rotation rate during growth to flatten the crystal-melt interface. The lower rotation rate shown in the table is the rate used to dip the seed at the start of the run. The higher value is the rate used to grow the body of the crystal with a flat interface.

Table 6-2. GGG crystals grown during phase 2 testing.

Run	Diameter (cm)	Length (cm)	Weight (kg)	[Ca] (ppm)	Rotation (rpm)	Notes
3	7.0	0.5	0.4	0	12	Cupped
4	7.8	16.6	4.8	25	12	Cracked
5	9.4	11.3	6.5	25	12	Cracked
6	9.3	19.7	8.3	25	12	Cracked
7	10.2	24.7	9.9	25	12	Cracked
8	10.3	17.9	11.6	25	12-15	Flat
10	11.5	9.1	7.4	25	12-16.5	Spiral
12	11.0	22.6	17.0	25	12-15	Cracked
13	11.0	22.8	17.0	35	6-13.3	Flat
14 ^a	7.6	16.0	6.0	35	6-24	Spiral

^a Neodymium-doped (10^{20} ions/cm³).

Fig. 6-12. Largest undoped GGG boule grown at LLNL is 11 cm in diameter and weighs 17 kg.

Runs 8 and 13 were the most successful. Figure 6-12 shows the boule as grown from run 13. While there are some dislocations within the boule, the diameter control is excellent. Only one doped GGG crystal was grown at a Nd concentration of 10^{20} ions/cm³. While this boule eventually formed a spiral shape, its internal quality (with respect to stress, inclusions, and dislocations) is excellent. Spiraling may be reduced by growing at a lower rotation rate and increasing the Ca concentration in the melt.²⁰

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Czochralski Crystal Growth Modeling. Significant scaleup of garnet crystal growth depends on acquiring a fundamental and complete understanding of the controlling features of Czochralski crystal growth. Unfortunately, this growth process is hindered by the interaction of many nonlinear physical phenomena that complicate experiments and often defy intuition. Long-range radiative interactions in a complex geometry dominate heat transfer through the entire growth system. Heat transfer in the melt is governed by intense fluid flow driven by density and surface-tension gradients produced by the temperature field and by crystal rotation. Finally, the interplay of the growth environment and the evolving melt-crystal interface, crystal shape, and melt meniscus strongly affect the properties of the grown crystal.

Improvements in the quality of the end-product crystal are obtainable only if all of these effects are understood, and modeling is often the only way to independently probe the importance of these phenomena.

The mathematical description of these processes in a comprehensive model for CZ crystal growth is the thrust of the effort described here. The model will be a predictive tool to expand our understanding of classical CZ growth, to develop and optimize current CZ processes, and to determine the design of new crystal-growth processes.

In the first phase of model development, the hydrodynamic thermal-capillary model (HTCM)²¹ introduced the complexities of fluid mechanics to an existing model for heat transfer and interface shapes in CZ growth. Recently, the integrated process model (IPM)²² was developed to include features of the furnace environment that directly impact the CZ growth process. The IPM integrates features of the induction heating analysis of Gresho and Derby²³ to describe realistic crucible heating, the radiation enclosure analysis of Atherton et al.²⁴ to calculate heat transfer through the furnace, and the HTCM for description of heat transfer, fluid flow, and interface shape in the crucible, melt, and crystal. (See Refs. 21 and 22 for details of model development.)

Figure 6-13 shows the spatial distribution of inductive heat deposition in the crucible. The bottom and top edges of the crucible are heated strongly by the inductive field; most of the remaining energy is deposited into the crucible wall. References 23 and 25 explore this phenomenon in more detail.

The crucible heating result of Fig. 6-13 is used as an input to the heat transfer calculations in the integrated process model.²²

We are now in a position to directly compare model results with experimental measurements. These efforts are under way. In addition, the model is being used to explore the sensitivity of the process to operating parameters and to probe the phenomena that govern the process.

Through these efforts, we continue to expand our fundamental understanding of the CZ growth of oxide crystals.

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Damage Measurements at 1064 nm

We compiled a data base of laser-induced damage thresholds measured at moderate pulse-repetition frequencies (PRF) of 6 to 120 Hz at the repetition laser experiment (REPTILE) facility. The samples were irradiated with 1064-nm, 16-ns pulses focused to a spot size nominally 1 mm in diameter. We attained fluences exceeding 40 J/cm^2 . Most measurements were conducted at 120-Hz PRF, which we obtained by interleaving the pulses of four commercial lasers each operating at 30 Hz. We typically irradiated a sample for five min (36 000 shots) unless obvious damage was observed earlier by naked eye. We determined final damage by comparing pre- and post-shot photographs of the irradiated site taken with a Nomarski microscope at $100\times$ magnification.

We conducted over 200 damage tests on samples supplied by more than 20 sources both from LLNL and commercial vendors. These samples represent a broad spectrum of commercially available optics as well as evolutionary development in state-of-the-art research coatings and substrates. We separated them into seven categories of laser optics: antireflective (AR) coatings, highly reflective (HR) coatings, polarizers, single or multiple layers of the same thin-film material, metals, bare polished dielectric and crystalline surfaces, and bulk materials. Within each category we attempted to test samples covering the full range of attainable thresholds, but we cannot preclude the existence of samples with even higher thresholds.

Figure 6-14 summarizes tests conducted on AR coatings deposited on a

Fig. 6-13. Perspective plot of the rate of heat generation in the crucible produced by the inductive heating system. The vertical scale is linear.

variety of substrate materials: fused silica, BK-7, calcium fluoride, and several nonlinear crystals. The e-beam-deposited coatings consisted of multilayer designs utilizing the specified high-index material together with silica or magnesium fluoride. On the other hand, the sol-gel coatings, all fabricated at LLNL, were made of one or more layers of a porous material. In both cases, differences in thresholds of samples made of the same materials were attributed to variations in coating designs and fabrication procedures. The best samples made by either

of these coating processes had thresholds on the order of 30 J/cm².

We tested HR coatings of many designs, deposited on substrates of fused silica, BK-7, or silicon carbide. Most designs consisted of alternating layers of silica and the materials listed in Fig. 6-15. Laser damage thresholds were strongly dependent both on material combinations and their specific designs. For

e-beam coatings, titania/silica designs yielded both the highest thresholds, exceeding 36 J/cm², and some of the lowest. For sol-gel (porous) coatings made of these two materials or of other material combinations, thresholds were lower than those of counterpart films made by e-beam deposition. However, the data base is too small and the variety of samples tested too large to preclude any

Fig. 6-14. Laser damage thresholds of AR coatings measured with the REPTILE Laser under moderate average-power conditions, 60 to 120 Hz, using 16-ns, 1064-nm pulses.

Fig. 6-15. HR coatings, 60 to 120 Hz, using 16-ns, 1064-nm pulses.

particular material combination as a candidate for high-threshold reflectors.

We conducted polarizer damage tests primarily at 60 Hz with P-polarized pulses incident on the samples at their respective design angles that ranged from 55 to 66°. Most substrates were made either of fused silica or BK-7. The e-beam coating designs combined layers of silica with layers of the high-index materials specified in Fig. 6-16. The samples typically had higher densities of coating

defects than corresponding HR coatings. Although only a few of these defects actually contributed to the damage induced at the threshold fluence, once begun, both the magnitude and number of damage points grew rapidly under repeated irradiation. The sol-gel coatings consisted of only single layers of the materials shown deposited on fused silica substrates. These samples were relatively free of visible defects, but damage accumulated rapidly with repeated irradiation.

Fig. 6-16. Polarizers, 6 to 60 Hz, using 16-ns, 1064-nm pulses.

Fig. 6-17. Single and multiple layers, 60 to 120 Hz, using 16-ns, 1064-nm pulses.

Polarizers made with titania were especially susceptible to damage under these conditions. For identical high-index materials, both e-beam and sol-gel polarizers had comparable thresholds. We tested only a few e-beam and sol-gel samples with S-polarized pulses; to first order, the thresholds were comparable to the P-polarization measurements.

In the process of developing sol-gel coatings at LLNL, we tested a wide variety of single materials deposited as one or more layers on fused silica. Figure 6-17

shows these results (except for those made of silica, which are included with AR coatings). Thresholds of individual materials, even with multiple layers, were typically higher than those observed when materials were combined in alternating layers with other materials such as silica. Alumina sol-gel layers yielded the highest thresholds, comparable to those obtained with silica sol-gel AR coatings. With many sol-gel coatings, we observed a momentary flash of white light when irradiation began. Presumably

Fig. 6-18. Metallic reflectors, 120 Hz, using 16-ns, 1064-nm pulses.

Fig. 6-19. Bare polished surfaces, 6 to 120 Hz, using 16-ns, 1064-nm pulses.

this was caused by the burning of residual solvent in the porous coatings because we could detect no indication of damage to the coating after irradiation.

Eight vendors supplied an assortment of metallic reflectors that were fabricated either by polishing, electroplating, or diamond-turning. Figure 6-18 shows that metals, whether bare or coated with dielectric layers, had the lowest thresholds of all the categories of optical materials tested. Near the threshold, damage nucleated around observable artifacts and then grew no further. However, only slight increases in fluence produced significantly more massive damage. A single-layer dielectric overcoat or multilayer HR deposited over the metal always resulted in thresholds slightly higher than for those samples without the protective dielectrics. The highest threshold measured was 4.3 J/cm^2 for a diamond-turned copper substrate with a dielectric HR stack.

We examined surface damage to front or rear surfaces of bare polished substrates for four types of samples: fused silica, calcium fluoride, nonlinear crystals, and silicon carbide. Each type formed a well-defined grouping of thresholds with the spread governed primarily by the quality of polishing and the extent of previous handling (Fig. 6-19). There was no significant difference in thresholds

between the various nonlinear crystals, which included KDP, KD*P, and L-arginine phosphate (LAP). For many of the samples, especially the fused silica substrates, we failed to induce any surface damage at all because of limitations on available fluence during certain tests.

We examined many of these same substrates separately for damage within the bulk material itself (Fig. 6-20). Damage was first detected visually by increased scatter of a collinear He-Ne laser beam and was confirmed by detailed bulk microscopy. We found that in nonlinear crystals the bulk material damaged at fluences typically 30% lower than those causing damage to the surfaces. We observed the opposite effect with calcium fluoride for which no bulk damage was observed unless the surface damage became so massive that it propagated into the bulk material. Many bare and AR-coated fused silica substrates were tested without producing any bulk damage at fluences exceeding 36 J/cm^2 . We therefore show these results as indeterminate both for threshold and number of samples tested.

The REPTILE facility provides a rapid means of evaluating damage resistance of optical materials at 1064 nm. Its high PRF facilitates measurement of component lifetime and large-area testing. In general, our tests reveal no clear evidence of

Fig. 6-20. Bulk materials, 6 to 120 Hz, using 16-ns, 1064-nm pulses.

PRF-related effects at rates up to 120 Hz. We find this both from comparison with damage produced by single-shot tests at the same pulse duration and from large-area temperature rises of only 0.1 to 3.5°C measured with an infrared camera. Repeated irradiation merely accentuates some subtle damage in a few samples that may then grow to massive damage after several thousand shots. Because thermal effects due to the heating of μm -sized defects are not resolvable by our camera, they are not precluded as a possible damage mechanism.

Authors: F. Rainer and E. A. Hildum

Optical Characterization of Advanced Laser Materials

The goals of the high-average-power (HAP) laser program place significant constraints on the optical quality of all transparent laser components. As a result, we undertook a concurrent materials development program. In support of this development program, we established a laboratory to perform highly sensitive loss measurements on newly developed or processed materials. A key instrument used for materials characterization in this laboratory is the scanning optical lossmeter. This innovative device (see "Optical Loss Measurements with Cavity Lossmeter" in the 1986 Laser Annual) performs wavelength-dependent total loss measurements on highly transparent samples by a cavity ringdown technique.

Table 6-3. Some representative results of loss measurements.

Material	Loss (%/cm at 1064 nm)	
	Lossmeter	Calorimeter
GSGG		
Boule 5023-1	0.57	0.26
Boule 5023-2	0.63	0.32
Boule 5028-1	0.50	0.30
Boule 5028-2	0.40	0.34
KD*P (deuteration level)		
95.0%		0.070
97.0%		0.056
99.0%		0.042
99.2%		0.034
LiNbO ₃		
AL5100		0.078
AL101		0.133

Consequently, the result of this measurement is the net loss due to surface and bulk scatter and absorption losses.

As an aid to help differentiate scatter loss from absorption, we also established a laser calorimetry capability. This instrument measures absorption loss by monitoring the temperature rise of a thermally isolated sample that is experiencing a constant laser irradiation. Consequently, this technique measures only the losses that produce heat.

To better understand the role of scattering as a loss mechanism, we recently constructed an instrument to accurately measure bulk scatter. Preliminary data from this facility is just becoming available.

Table 6-3 shows some representative results of loss measurements performed on either new or specially processed materials being considered for high-average-power applications. Much of the data obtained in the laboratory is discussed in the detailed individual articles that follow.

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Nonlinear and Electro-Optic Materials Development

Linear Electro-Optic Effect in $\beta\text{-BaB}_2\text{O}_4$

The linear electro-optic (EO) effect is used primarily for fast-switching of optical pulses. Currently, electro-optic switching of HAP lasers is limited by the thermo-mechanical properties of the EO materials. Although beta-barium borate ($\beta\text{-BaB}_2\text{O}_4$) is considered an excellent material for second-harmonic generation in the ultraviolet, its potential as an EO material was unexplored. $\beta\text{-BaB}_2\text{O}_4$ might be an excellent candidate for use in a HAP laser EO switch because of its resistance to thermal fracture, its high damage threshold, and its wide range of transparency.²⁶ In order to evaluate its potential for use in such an application, the linear EO coefficients have been measured.

Fig. 6-21. Apparatus used for electro-optic measurement.

Figure 6-21 shows the apparatus used for the measurement. The Pockels cell was biased such that light with circular polarization was incident upon the sample. The compensator was inserted to correct for the static birefringence of the sample in several of the measurements, thus returning the light to a state of circular polarization.

For the unclamped measurement (the EO coefficient at zero stress), a Fluke H10B power supply capable of supplying ± 10 kV was connected across the faces of the crystals via sputtered gold electrodes. The change in light intensity as a function of applied voltage was measured using a 200-Hz chopper and lock-in amplifier (LIA), yielding the absolute unclamped EO coefficient, r^T .

The system was calibrated by measuring the unclamped coefficients of two common nonlinear optical materials, KH_2PO_4 (KDP) and LiNbO_3 .²⁷ The measured values for KDP and LiNbO_3 were $r_{xyz}^T = 9.9$ pm/V and $r_{xyz}^T = 7.2$ pm/V, respectively. These compare well to the most reliable values reported in Ref. 27, 10.3 pm/V and 6.8 pm/V. Three measurements were made per sample with a reproducibility of $\pm 1.5\%$, due to fluctuations in light intensity.

A 20-V peak-to-peak sine wave from a Hewlett-Packard function generator was applied to the sample for the clamped measurement (the EO coefficient at zero strain). The resulting modulation in light intensity, $\Delta I/I_0$, was detected using an EG & G 5202 LIA. The modulation frequency was scanned from 100 kHz to 13 MHz. We observed that the total

 Table 6-4. Measurement configurations for $\beta\text{-BaB}_2\text{O}_4$.

Sample	k	E	Γ^a
1	[010]	[101]	$\frac{2\pi l_{0101}}{\lambda} (n_e - n_o) - \left[\frac{(n_e)^3 r_{zzz} - (n_o)^3 r_{xxz}}{2\sqrt{2} l_{0101}} \right] V_{app}$
1	[101]	[010]	$\frac{2\pi l_{1011}}{\lambda} (n_e - n_o) + \frac{r_{xxx} V_{app}}{4 l_{1011}} [2(n_e)^3 + (n_o)^3]$
2	[001]	[010]	$\frac{2\pi l_{0011}}{\lambda l_{0101}} (n_o)^3 r_{xxx} V_{app}$

^a Derived using Eq. (50) in Ref. 30.

variation in modulated intensity, due to the photoelastic contribution²⁸ to the clamped EO coefficient in the frequency range of 3.2 to 13 MHz, was less than 8%. The data from 3.2 to 13 MHz was averaged and compared with that from a KDP standard, yielding a relative measurement of the clamped EO coefficient, r^S . For calibration purposes, r_{xyz}^S of KDP was compared in the transverse and longitudinal configurations,²⁹ using two different crystals. The resulting coefficients agreed with each other to within 1.5%.

Two samples of $\beta\text{-BaB}_2\text{O}_4$ were measured. Sample 1 was a thin slab with polished (101) and (010) faces and dimensions $l_y = 9.30$ mm and $l_{101} = 2.90$ mm. Sample 2 had polished (010) and (001) faces with dimensions $l_y = 8.12$ mm and $l_z = 5.04$ mm. Crystal faces upon which the modulating electric field was applied (see Table 6-4), were coated with approximately 260 Å of gold.

Table 6-4 indicates the geometry of the measurements.³⁰ E is the direction of the applied field $E_{app} = V_{app}/l$, k the direction

Table 6-5. Measured electro-optic coefficients^a for β -BaB₂O₄.

r	Value
r_{yyz}^S	$(0.24 \pm 0.03) r_{yz}^S$ (for KDP)
r_{yyy}^T	2.5 ± 0.1 pm/V
$r_{xxz}^S = r_{zzz} - (n_o/n_e)^3 r_{xxx}^S$	$(0.013 \pm 0.002) r_{yz}^S$ (for KDP)
r_{xx}^T	0.17 ± 0.02 pm/V

^a At $\lambda = 514.5$ nm and $T = 23^\circ\text{C}$.

Table 6-6. Fracture data for seven KD*P disks.

Sample	Breaking strength (MPa)
1	240
2	219
3	289
4	205
5	230
6	208
7	248

of the wave vector, Γ the phase difference of the light propagating along the two orthogonal eigenmodes through the length of the crystal, n_o and n_e are the ordinary and extraordinary indices of refraction, and n_e' is the extraordinary index of refraction with the given propagation vector $\mathbf{k}$. In calculating the value of the r -coefficients in Table 6-5, we used the values $n_o = 1.67$ and $n_e = 1.55$ at $\lambda = 514.5$ nm.²⁶

Using a value of $r_{yyz}^S = 8.6$ pm/V for KDP at 10 MHz,³¹ we obtain a value of $r_{yyz}^S = 2.1$ pm/V for β -BaB₂O₄ from Table 6-5. A prediction of the magnitude of $r_{yyz}^S = 2.7$ pm/V in Ref. 26, calculated from the measured second-harmonic generation coefficient d_{yyz} , agrees well with the measured value, indicating that the contribution to the magnitude of the EO coefficient is due mainly to the electronic nonlinearity. Assuming no ionic contribution to the EO coefficient in β -BaB₂O₄ and using Kleinmann's symmetry and the relation between the EO coefficient r^S and the second-harmonic coefficient d ,³² we would expect that

$$r_{xxz} = r_{xxz}.$$

For waves propagating down the optic axis of β -BaB₂O₄ with the applied field in the [010] direction, a plate with length $l_z = 2$ cm and $l_y = 0.5$ cm has a half-wave voltage of 14 kV. Thus β -BaB₂O₄ can be configured as a Pockels cell with reasonably low voltage for waves propagating parallel to the optic axis, where the static

birefringence is zero. With its high damage threshold and favorable thermomechanical properties,²⁶ β -BaB₂O₄ is an attractive material for HAP optical switches.³³

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Stress Fracture Tests on KD*P

We purchased ten 1-in.-diam disks of deuterated potassium dihydrogen phosphate (KD*P) from Cleveland Crystals. Each disk had one surface diamond-turned so that we could estimate the tensile fracture strength of diamond-turned KD*P Pockels cell crystals. Seven disks were fractured in testing, one was broken accidentally, and two were saved for future slow-crack-growth experiments. Table 6-6 shows the fracture data.

The Weibull extension-value distribution³⁴ that describes this data for determining the probability of fractures for a Pockels cell is

$$\phi = 1 - \exp \left[- \frac{A}{2 \times 10^{-7}} \left(\frac{\sigma}{247} \right)^{9.3} \right], \quad (6)$$

where ϕ is the fracture probability, A is the surface area of the KD*P component exposed to tensile stress (in m²), and σ is the tensile stress (in MPa). For a 10×2 -cm Pockels cell, this expression predicts that half the elements will break at 88 MPa. For $\phi = 0.1$ (10% will break), the stress is 56 MPa.

We are examining the possibility that slow crack growth could give delayed failure at lower stresses.³⁵ We use the fracture mechanics expression³⁴

$$\sigma_f = \frac{YK_c}{\sqrt{\pi a}} \quad (7)$$

to estimate the flaw depth; here σ_f is the strength, Y is a geometrical factor (1.12), K_c is the fracture toughness (0.10 MPa for KDP), and a is the flaw depth. For the tests shown in Table 6-6, the depth

averaged $0.06\text{ }\mu\text{m}$. This surprisingly shallow damage is probably related to significant plastic flow during diamond turning.

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Laser Component Development

Zig-Zag Slab Amplifiers

Wave-Front Validation with the B-Head Test Amplifier. We conducted experiments with the B-head test amplifier, providing wave-front measurement data, to validate our numerical simulation code, TECATE-BREW. In these experiments, we used two $32 \times 10 \times 1.5\text{ cm}$ silicate glass slabs: a zig-zag design and a straight-through design. Both slabs were fabricated from LSG-91H glass manufactured by Hoya Optics, Inc. The zig-zag slab was doped with 2.7 wt% Nd, and the straight-through slab was doped with 1.9 wt% Nd. The zig-zag slab was designed to have ten internal bounces and a 20-cm pump region centered on the slab length. The entrance and exit wedge angles of this slab were both 28° , which produced unit volume fill and essentially unit beam size.

To reduce the slab's angular sensitivity, we selected a zig-zag-slab length that allowed each ray to bounce an even, rather than odd, integer number of times at total internal reflection (TIR) surfaces. The condition of unit volume fill is achieved when, after refracting into the slab, all rays propagate parallel to the slab wedge faces after their first bounce from a TIR surface. The entire slab volume is then double passed by the beam as it overlaps itself on the zig-zag path. The straight-through slab was used to validate the numerical simulations without the complexity of zig-zag propagation.

Wave-Front Validation. We performed wave-front validations progressively in a set of carefully posed experiments. Purely static and purely thermal boundary conditions were validated first in the zig-zag slab without pumping, followed by the pumped zig-zag and straight-through slab validations.

Fig. 6-22. Validation of the zig-zag slab's wave-front distortion under purely thermal boundary conditions. T is temperature.

The wave-front distortion observed under purely static loading (i.e., without a thermal load) was validated in 1986, using the unpumped zig-zag slab in Ref. 36. In that experiment, the slab was loaded at its tips in a simple fixture designed to bend the slab. The resulting fringe pattern is very similar to the pattern calculated using the TECATE-BREW model.

The purely thermal experiment was more complex because of the difficulty in insulating the slab from the environment. In this experiment, a temperature difference was applied between the top and bottom of the unpumped zig-zag slab using chill bars as shown in Fig. 6-22(a). The

sides, or TIR surfaces, were insulated by a stagnant air layer, with the entrance and exit faces open to room air. Because this is not a perfect insulation scheme, we recorded the slab side temperature as a function of slab height to establish the amount of thermal flux leaving the sides [see Fig. 6-22(b)]. We assumed that the thermal impedance, or film coefficient, was uniform along the slab sides. We obtained reasonable agreement between the observed optical path difference inferred from the Fizeau fringe pattern and the calculated slab optical path difference (OPD), using a spatially uniform film coefficient of $7 \text{ W/m}^2 \cdot ^\circ\text{C}$ as shown in Fig. 6-22(c). This value gave the best fit for slab surface temperature. It is difficult to obtain a more accurate agreement between the calculated and observed OPD without undertaking an extremely detailed evaluation of spatial variations in the film coefficient. Unlike the experiments with water-cooled slabs, the thermal impedance between the slab sides

and the "isothermal" room in these air-cooled experiments was comparable to the thermal impedance between the top and bottom of the slab itself. We found a much closer agreement when the slab was cooled with water; however, under these experimental conditions, we were unable to influence the slab OPD over an aperture distance much larger than a slab thickness.

Optical performance was measured under flashlamp pumping conditions, using the assembly shown in Fig. 6-23. The slab was mounted in a central vane, which provided an elastomeric support, amplified spontaneous emission (ASE) suppression, and edge cooling of the slab. This central vane consisted of the slab bonded by a transparent elastomer (Urapol, $n = 1.47$) to a $1.06\text{-}\mu\text{m}$ ASE absorbing KG-3 glass (from Schott Glass Technologies, Inc.), which was bonded to a stainless-steel chill bar. This slab/chill-bar assembly was then bonded to polycarbonate extensions. The slab, elastomer, chill bar, and extensions were all flush-mounted forming the central vane (i.e., one side of the slab coolant channel).

The other side of the coolant channel consisted of a pump window (0.4 cm thick) fabricated from Schott BK-7 glass that was flush mounted in a polycarbonate side panel. One of these side panels was mounted on either side of the central vane. A large O-ring formed the seal between the central vane and the side panels, except for the region around the slab entrance and exit faces. These faces were sealed with an adhesion seal made of Urapol.

Cooling water entered the amplifier head through a plenum at the bottom of the assembly and exited the amplifier through a second plenum on the top of the assembly. The flow upward through the cooling channel ($1.5 \text{ mm} \times 29 \text{ cm}$) was turbulent for flow velocities $> 10 \text{ gpm}$. We also operated in the turbulent flow transition region near 5 gpm .

The pump cavity consisted of two single-lamp "Goodwin" reflectors,³⁶ one mounted on each side panel. The Ce:SiO_2 flashlamps were 1.0 cm in diameter, were filled with 300 Torr of xenon gas, and were electrically in series. A 1.5-cm-i.d.

Fig. 6-23. The B-head test amplifier used with the zig-zag slab to validate the TECATE-BREW results under flashlamp pumping conditions.

water cooling jacket on the lamp was also made of Ce:SiO_2 . The pump reflector surfaces were machined flat, buffed, coated with evaporated silver, and overcoated with an SiO_2 thin film for protection against oxidation. During operation, the flashlamps were simmered at ~ 120 mA and were pulsed by a single mesh, resonantly charged pulse-forming network ($3\tau \approx 600 \mu\text{s}$). The pulse repetition rate and energy were adjusted to control the average power.

To record wave-front distortions under thermal load, we put the amplifier assembly in a Fizeau interferometer cavity, which was mounted on a large isolation table. To avoid coupling the cooling system vibrations to the Fizeau cavity components, we mechanically isolated the amplifier from the interferometer table with a separate air suspension table. This system was operated with either a He-Ne laser source or a Nd:YAG source. In both cases, the Fizeau cavity length was adjusted to be a multiple of the laser resonator length to within a coherence length. This provided a stable interference pattern that was either recorded on film or was digitized using a charge-injection device (CID) camera.

These CID data were processed using a Fourier-domain signal-processing technique that provides an unambiguous measure of the phase distortion through the amplifier (see Sec. 3, "Nova Computer Systems" for details). Some of the early interferometric data was only recorded on film as conventional interferograms. In this article, the CID film data

are unavoidably mixed with these more easily understood phase-front plots.

Wave-front validation under flashlamp pumping conditions relies strongly on knowing the temperature distribution in the slab. Before this can be determined, the thermal source distribution in the slab must be described. The dominant thermal source is pump light absorption by the slab. There are, however, other significant sources of heat in the pump cavity that can couple to the slab through its boundaries. For example, heat deposited directly in the edge elastomer from the flashlamp can be conducted to the edge of the slab before reaching the coolant. This thermal source distribution, when combined with the thermal boundary conditions, drives the wave-front phase distortions in the slab. Therefore, the wave-front validation experiments performed under flashlamp pumping conditions verify both the TECATE-BREW and MIR3D pump-cavity models in a consistent sense.

The experimentally observed wave-front distortion under pumping conditions is shown in Fig. 6-24. The Fizeau fringe pattern has been reduced to single-pass beam phase as a function of position in the aperture. We see that the beam phase is dominated by astigmatism, i.e., focusing in a direction uncompensated by the zig-zag propagation. The TECATE-BREW/MIR3D simulation is also shown and contains many of the features seen in the observed wave front.

To obtain this level of agreement between the observed phase distortion and

Fig. 6-24. Measured and calculated wave-front distortion of the zig-zag-slab amplifier. The calculated wave-front distortion is dominated by non-uniformities in pumping. Measurement and calculation were both made at 3000 W of electrical input power.

our simulation, we included the thermal source in the edge elastomer. This source is driven by the overlap between the infrared emission of the flashlamps below the lamp's coolant-water infrared cutoff at $1.2\text{ }\mu\text{m}$ and by the infrared absorption edge of the Urapol polyurethane elastomer at $1.1\text{ }\mu\text{m}$. We also derated the reflector reflectivity from 97 to 80% to produce the smooth focusing effect along the long dimension of the aperture.

We are concerned that there may be some contribution to the observed wave-front phase distortion from a thermal source redistribution mechanism that is not currently modeled (e.g., fluorescence repumping). Nevertheless, the agreement between the observed and calculated beam phase is quite good, as shown by the magnitude of the phase distortion and by the reproduction of higher spatial frequency features known to be in the flashlamp-reflector design.

Wave-Front Correction. In view of the strong dependence of wave-front phase aberrations upon details of the thermal sources, the most direct way to correct the wave-front distortion observed in our small B-head test amplifier is to make the thermal source distribution uniform. This could be accomplished by proper design of the flashlamp reflector, accounting for any thermal source redistribution effects. We believe that small variations in pump uniformity will cause unacceptable variations in OPD, placing impractical limitations on the fabrication of the flashlamp reflectors.

Because we require only that the path-length-integrated OPD and gain are

spatially uniform over the aperture, we developed a method for correcting small OPD and gain errors caused by the reflector design. One method of correcting the zig-zag slab's wave-front distortion caused by nonuniformities in pumping is to purposefully modify the spatial distribution of the pumping at the ends of the slab's pump region. We demonstrated this with an empirical procedure in our test amplifier (Fig. 6-25). Four reflective pump masks were positioned between the slab coolant window and the flashlamp/reflector assembly; two on each side of the slab. Initially these masks had a simple parabolic shape. By iterating between the wave-front distortion changes and small modifications to the mask shape, we reduced the wave-front distortion to that shown in Fig. 6-25. We subsequently modeled this masking technique, using the empirically derived mask shape, and obtained similar results. We expect even better agreement as we improve the model for the pump cavity. The existence of a simple method for correcting the zig-zag slab's wave-front distortions, however empirical at this time, is encouraging.

Straight-Through-Slab Comparison.

We modeled the straight-through slab and compared the computed results with data obtained in laboratory measurements. Wave-front phase measurements revealed the efficacy of the zig-zag geometry and showed good agreement with the TECATE-BREW/MIR3D code results. These data were taken at a relatively low pump power, corresponding to $\sim 25\%$ of the pump power used in the zig-zag experiments so that the strongly focused

Fig. 6-25. Wave-front distortion in the zig-zag slab can be corrected by using masks in the pump cavity to modify the pumping profile. Both measurements were made at 3000 W of electrical input power.

straight-through interferogram could be resolved. In the absence of zig-zag propagation, effects of surface deformation are limited to refraction at the entrance and exit faces of the straight-through slab, which is negligible in comparison with the thermal lensing. Figure 6-26 shows the depolarization data for the straight-through slab.

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Gas-Cooled Disk Amplifiers

Introduction. The geometry of a typical gas-cooled disk amplifier is similar to that of a conventional solid state disk amplifier. Using this geometry for average-power applications requires two types of flow regime: A flow channel to cool the active-medium slab and a drift region to condition the flow between slab assemblies. In 1987, the goal of the gas-cooled disk amplifier program was to

understand the gas dynamics, heat transfer, and optical interactions of the gas flows with the plane wave front of a probe beam. We experimented on compressible channel flow with and without heat transfer and developed flow codes that accurately describe the results. Experiments characterized the scattering of coherent light by channel flow and drift-region turbulence and show that for properly chosen parameters this scattering loss can be made negligible.

We made measurements on a flow channel test bed in which the gas stream is confined in a narrow gap formed by the laser slab and a flow separation window. Figure 6-27 shows the channel flow geometry, which will generally consist of the following:

- A nozzle that feeds the gas into the flow channel.
- A development region upstream of the laser slab that can hydrodynamically and thermally condition the cooling gas.
- The laser slab region where heat is removed.

Fig. 6-26. (a) Measured and calculated wave-front distortion for the straight-through slab at ~ 750 W of electrical input power. (b) Measured and calculated depolarization at ~ 3000 W of electrical input power.

Fig. 6-27. Flow channel test bed.

- A downstream relaxation zone.

- The downstream diffuser system for pressure recovery.

Our primary interest is the laser slab region; however, the upstream and downstream sections are of importance in determining all flow properties at this location.

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Bulk Flow Behavior.

Introduction. Whether developing or fully developed, flow in a thin channel is boundary-layer in character and is directed predominantly along the channel axis. Figure 6-28 shows how characteristic flow parameters evolve as the flow proceeds down the channel.³⁷ At the exit of an ideal nozzle, the viscous boundary layer is very thin, and the velocity profile is nearly uniform. At this point, frictional forces at the wall start to retard the flow, resulting in the formation of viscous boundary layers that penetrate into the core flow. The viscous development continues until penetration of the wall effects is complete, and the profile becomes fully developed. Because of this viscous action, the static pressure decreases as the flow proceeds down the channel.

The temperature profile is influenced by shear-generated energy in high-velocity-gradient regions. In the absence

of heat transfer at the walls, this heating causes a slight temperature rise in the fluid within the viscous boundary layer. At the slab, energy removal forms a thermal boundary layer where the surface temperature is much higher than the bulk fluid temperature; steep temperature gradients exist in the gas near the slab surface. Like the viscous boundary layer, this thermal boundary layer penetrates across the channel until something resembling a fully developed profile is achieved. As the thermal boundary layer thickens, gradients near the wall decrease. The viscous profile is symmetric, while the thermal profile is asymmetric because heat is only being removed from the laser slab side of the channel.

The bulk character of the flow has a pronounced effect on distortions in the system optical components. Optical distortions can result from cooling-induced temperature gradients and from pressure imbalances due to a drop in channel pressure. We studied the bulk flow and thermal characteristics in the cooling channel flow. We developed computer models to perform quick and inexpensive numerical experiments to explore system design options. Using the flow issues test bed (FIT-1), we gathered data to validate the numerical models.

Models. Computer models help quantify the bulk character of the cooling-channel flow and provide details of heat transfer at the laser slab surface. We used three types of models:

1. Analytical solutions to the one-dimensional flow equations for special flow conditions.
2. Numerical integration of the one-dimensional flow equations, using experimental correlations for boundary effects.
3. Numerical integration of the two-dimensional Navier-Stokes equations using high-order turbulence modeling.

The one-dimensional equations of motion are amenable to analytical solution only for special conditions, of which two are of interest in gas-cooled slab configurations. One (Fanno flow) results if the channel flow area is constant and friction is the dominant effect on the flow. The other, the so-called tapered channel, results if the channel flow area is allowed to adjust so as to maintain a constant

Mach number under the combined effects of friction and energy addition. The very slight taper angle that results eliminates the flow acceleration caused static pressure drop for moderate Mach numbers (>0.2). Both geometric conditions, which were addressed in preliminary parametric investigations of flow and heat transfer in the cooling channels,^{38,39} continue to be used in system optimization studies. (See Ref. 38 for details of the models).

Other flow conditions require numerical integration of more complex equations. The simplest general solution results from integration of the steady, one-dimensional equations of motion. Ordinary differential equations describing conservation of mass, momentum, and energy for the very general case that includes flow area change, frictional drag, and energy addition can be written in terms of average flow characteristics (velocity, temperature, pressure, etc.). Wall effects are treated using experimental correlations for the frictional drag and convective heat transfer. This type of solution gives a reasonable estimate of the bulk character of the flow, but it is deficient in that the experimental correlations for wall drag and heat transfer assume fully developed symmetric profiles. There are significant lengths of channel where the flow is not fully developed, and the thermal profile is asymmetric (see Fig. 6-28).

Understanding the flow and flow development requires solution to the more complex two-dimensional flow equations including high-order turbulence modeling. We used the two-dimensional boundary-layer program TEXSTAN^{40,41} to explore the effects of asymmetric heating and to quantify the effect of thermal preconditioning upstream of the slab. TEXSTAN integrates the steady, compressible, boundary layer form of the Navier-Stokes equations for mass, momentum, and energy conservation. We used a two-equation turbulence model, formulated in terms of the turbulent kinetic energy and turbulent dissipation function. In addition to details of the thermal and viscous profiles, these more refined calculations help determine the validity of the simplified analytical models.

Fig. 6-28. Flow development in the cooling channel.

Parametric Investigations. We performed parametric investigations to quantify trade-offs in system design parameters.^{38,39} These studies primarily looked at the effects of channel thickness, flow Mach number, and flow pressure on slab cooling effectiveness and system power requirements. Figure 6-29 plots two quantities, the slab temperature gradient and the ratio of the compressor power per unit surface area of slab and the removed thermal heat flux vs flow Mach number for various pressures. The same cooling effectiveness (represented by the slab temperature gradient) can be achieved using either a pressure of 1 atm and a Mach number of 0.25 or a pressure of 5 atm and a flow Mach number of 0.03 (line 1' → 2'). However, the compressor power required to remove each watt of thermal load is reduced by nearly a factor of 30 for the latter case (line 1' → 2'). Trends such as these drive us to explore systems with increasingly higher pressure and lower flow Mach numbers.⁴²

Bulk Flow Characterization—

Measurements and Calculations. Extensively instrumented experiments provided information on the bulk character of the channel flow and data for the validation of various numerical approaches.

Fig. 6-29. The effect of Mach number and pressure on the cooling effectiveness and compressor power requirements for a 30-cm aperture system with thermal heat loading of 2 W/cm^2 .

The static pressure in the channel, which is associated with window pressure distortion, and the surface and gas temperatures, which influence thermal distortion of the slab and separation window, are of special interest. Because such distortion of optical components degrades the beam profile, it will have to be minimized.

Static pressures along the channel were measured using static pressure taps and a Bourdon tube gage.⁴³ As the flow travels through the nozzle and down the channel, the static pressure will vary significantly. In the nozzle, the rapid contraction converts much of the internal energy to kinetic energy, with negligible reduction in the stagnation properties (temperature and pressure). As a result, the static temperature and pressure both drop, while the flow Mach number increases. In the channel, frictional drag causes an irreversible reduction in the stagnation pressure. The static pressure drops accordingly. Thus, comparison with measured static pressures is a good test for any numerical model.

Figure 6-30 shows an example of the measured and predicted static pressure in the FIT-1 fixture for a constant-area

channel and a tapered channel. The associated flow Mach numbers are also shown. Eighteen pressure taps were located within the channel, upstream of the nozzle, and downstream in the diffuser. The constant-area calculations used the analytical solution and the solution to the one-dimensional flow equations. The tapered channel requires complete integration of the one-dimensional flow equations. In all instances, the frictional pressure drop in the flow channel is modeled accurately. Furthermore, this figure demonstrates the effect that a very slight channel taper has on reducing the static pressure drop at moderate Mach numbers, thus reducing pressure imbalance distortions in the optical components.

Two issues must be considered with respect to temperature:

1. The surface temperature, which as a measure of the slab temperature influences system gain and optical distortions.
2. The thermal boundary-layer temperature profile, which pertains to scattering losses within the cooling gas. Experiments were conducted using the FIT-1 fixture with an electrically heated indium tin oxide resistive coating at the

inside surface of one of the windows.³⁴

Figure 6-31(a) shows the probe locations relative to the heated window. Figure 6-31(b) shows a miniature thermistor mounted to a hot-wire probe support. A micrometer positioner traversed the probe across the channel. The temperature of the inside surface of the heated window was inferred from outside surface temperatures measured with a thermal imaging camera. There is some uncertainty in this measurement because we cannot precisely quantify the effects of conduction within the slab on the temperature profile.

Figure 6-32 gives a representative comparison of TEXSTAN-calculated probe and surface temperatures to those measured for flow in a 3-mm-thick straight channel. In interpreting the temperature measured by the intrusive thermal probe, it is important to note that flow stagnates at the leading edge of any blunt object. Thus, the probe does not necessarily see the static temperature of the gas but a temperature more closely associated with stagnation conditions. Thermal conduction within the gas reduces the measured temperature somewhat below the stagnation temperature. The probe temperature T_p is related to the static temperature T_s by

$$T_p = T_s + r\Delta T_d, \quad (8)$$

where r is the adiabatic recovery factor, given approximately by the cube root of

▲ Fig. 6-30. Measured (data points) and calculated (curves) static pressures in a straight and a tapered (0.5°) channel.

Fig. 6-31. Thermal probe locations and probe configuration: (a) probe locations relative to the heated window; (b) probe geometry.

Fig. 6-32. Probe temperatures for a Mach-0.3 flow in a 3-mm channel at a pressure of 0.189 MPa and a surface heat flux of 1.67 W/cm².

Fig. 6-33. Calculated slab surface temperature profiles and observed temperature for a surface heat flux of 1.67 W/cm².

the Prandtl number (0.89 for nitrogen), and ΔT_d is the dynamic temperature rise. The probe temperature would be identical to the stagnation temperature if the recovery factor were unity. The additional lines on the ordinates of Fig. 6-32 give values for the calculated and measured surface temperatures. The profile temperatures agree to within 2°C (simply an offset); the calculated surface temperatures were as much as 5°C less than the measured value. We consider this to be excellent agreement, particularly considering the uncertainty in the inside surface temperature. Figure 6-33 shows the TEXSTAN-calculated slab surface temperature profile, neglecting conduction in the window and nonuniformities in the resistive film thickness. Also shown is the surface temperature (predicted by the simplified analytical solutions and

the one-dimensional integration) and the measured surface temperatures. Figure 6-33 further demonstrates the importance of proper consideration of thermal boundary-layer development. Only after a relatively long development length does the TEXSTAN-calculated gradient approach the value associated with simple caloric heating. The surface temperature gradient in the simplified algorithms, which assume that the thermal boundary layer is everywhere fully developed, is dominated by caloric heating of the gas. This has caused us to consider thermally preconditioning the flow upstream of the slab, using resistive heating, to eliminate these strong cooling variations.⁴⁵ Both methods give estimates that agree well with the experimental value in the downstream portion of the slab, particularly in view of the uncertainty in the measured inside surface temperature.

These results show the accuracy of our numerical approaches and demonstrate our ability to predict system operating characteristics; we will use this ability to design future experiments and to investigate configurations that cannot presently be addressed experimentally.

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Optical Diagnostics of Gas Flow.

Introduction. As outlined in the preceding article, an extracting beam must traverse two flow regimes. The first is the flow channel, where a thin channel of high speed gas removes up to a few W/cm² from a heated plate. The second is the roof-shaped region between slabs (called the drift region), where the gas flow is conditioned in such a way that beam distortions are minimized, while a high reflectivity of pump-light-reflecting surfaces is maintained. In either regime, light will be scattered at high enough angles to be lost to downstream optics. Since the gain per plate in a gas-cooled disk amplifier system is not high, these scattering losses must be well understood and minimized.

Turbulent scattering, like all scattering, arises from index of refraction fluctuations in the medium traversed by the probe beam. Since the refractive index in a gas depends on temperature and pressure, fluctuations of these quantities will cause scattering.

In turbulent flow cooling, pressure fluctuations are caused by turbulent velocity fluctuations. Pressure pulses from a variety of sources, particularly boundary-layer instabilities in the diffuser, occur at wavelengths too long to contribute to turbulent scattering. Gas-cooling of hot surfaces takes place across steep temperature gradients close to the solid surface. Turbulent flow turns these temperature gradients into temperature fluctuations. Hence, the scattered intensity will be increased if surface heat is removed.

We are funding a study aimed at formulating a theory describing these phenomena with a semi-analytical turbulence model. From it, we expect relevant scaling laws.

We investigated¹⁶ the problem experimentally by using two techniques: a modified Schlieren technique and a cavity lossmeter¹⁷ (manufactured by Deacon Research of Palo Alto, California).

In the Schlieren technique, the up-collimated beam from a 2 ω Nd:YAG laser is directed through the flow channel and brought to a focus. This image is then sufficiently magnified so that a mask

can be shaped and cut. The mask covers the diffraction pattern arising from the aperture that the flow channel presents to the beam.

Figure 6-34 shows the light forward-scattered by the turbulent features in the flow will be readily visible in the flow, which manifests itself as a readily visible halo. Because this halo is visible with the unaided eye, we can observe changes in intensity when flow parameters are varied. Since the focal length of the focusing lens is known, the extent of the halo allows a determination of the shape and size of the average scattering feature. The scattering occurs into an elliptical cone of 0.7×1.3 mrad and is fairly insensitive to variations in Mach number. This indicates that the scatterers are geometry-dominated, which is expected for flow in a high-aspect-ratio channel. We also see that a typical eddy has an elliptical shape, elongated by about a factor of 2 in the flow direction. The eddy dimensions for a 3-mm channel are about 1.3 mm long $\times$ 0.7 mm wide. This is in reasonable agreement with the von Karman constant that typical channel flow eddy sizes are of order one-fourth of the channel thickness.

For quantitative measurement, the scattered light that appears as the halo around the mask is collected in a photo-detector and referenced to the incident intensity.

Fig. 6-34. Flow at Mach 0.6 shows the elliptical halo of light scattered by the turbulent features in the flow.

Fig. 6-35. Scattered intensity vs Mach number for 2-, 3-, and 5-mm-thick flow channels as measured with the modified Schlieren technique.

Fig. 6-36. Relative scattered intensity vs Mach number for 2-, 3-, and 5-mm-thick flow channels as measured with the lossmeter.

By following the scattered light and reconstructing an image of the flow channel with it, we obtain the usual Schlieren picture (i.e., a map of the refractive index gradients). These pictures show that the eddies are uniformly distributed across the aperture and that macroscopic flow instabilities and nonuniformities are absent in a flow channel with sufficient pre-conditioning length.

Figure 6-35 shows the scattered intensity vs Mach number for various channel thicknesses when no heat is removed from the slab. The scattered intensity scales with the fourth power of the Mach number. If we take a vertical slice through these data at any fixed Mach number, we obtain the relationship between scattered intensity and channel thickness. From the Schlieren measurements, we find that $L^{1.4}$ scaling (where L is the channel thickness) provides a good fit to the

data. To obtain a calibration for the absolute values of the scattered intensity, the scattered light is measured for varying mask sizes. From this, the value for zero mask size is extrapolated. Since the scattered intensity is at most of order 1%, the uncertainty in the calibration is around $\pm 35\%$.

In the lossmeter technique, we observe the temporal decay of a laser pulse between two very high reflectivity mirrors. Inserting a small loss into the cavity changes the decay time. From this change, we can deduce the magnitude of the loss. To measure turbulent scattering in the channel flow, we placed the flow channel in the center of a confocal cavity. The externally injected Q-switched pulse from a 2ω Nd:YAG laser is observed to decay at a rate dependent on the structure of the scatterers in the flow channel. This structure will, of course, depend on flow parameters, such as Mach number, pressure, and channel thickness. Unlike the Schlieren technique, this technique uses signal-averaging, which makes it possible to obtain very accurate measurements if operating conditions are stable.

Figure 6-36 shows the results obtained with the lossmeter technique. Again, the scattered intensity scales with the fourth power of the Mach number. Deducing the channel thickness scaling from this plot shows that the scattered intensity scales as L , rather than as $L^{1.4}$ as observed with the Schlieren technique. This is the first time the lossmeter technique was used to observe losses due to extreme forward scattering. We are confident that as we understand the technique better such apparent differences will be reconciled.

Since the scattering from randomly distributed density fluctuations scales with $(dp/p)^2$, and since dp/p itself is proportional to the square of the Mach number, the scaling of the scattered intensity with the fourth power of the Mach number is not surprising. If the scatterers are small compared to the traversed path length L , a scaling proportional to L is expected for small losses. Deviations from this linear law can be expected to arise from eddy sizes comparable to the channel thickness since, in this case, no averaging takes place.^{19,41}

Heat removal was simulated by an electrically heated resistive indium tin oxide film applied to the inside of one flow channel window. The film, which had 85% transmission, showed no degradation for surface heat fluxes of up to 4.7 W/cm^2 . As expected, the thermal fluctuations now contribute to the total scattered light. The shape and extent of the scattered light halo observed in the focus does not change noticeably as the film is heated.

Figure 6-37 shows by what factor the amount of scattered light is increased when heat is applied for a 3-mm channel. Since a slower flow implies higher surface temperatures, it is not surprising that the case for a Mach number of 0.3 shows a greater thermal scattering enhancement than a flow with a Mach number of 0.5. The lossmeter technique could not be applied with the heating film present since its 15% transmission loss is much greater than the losses due to turbulent scattering.

As indicated in the introduction, the drift region now must be conditioned to minimize optical distortions in the space between the slabs. Since the gas must enter and leave through the pump-light reflector surfaces, we approached the problem by replacing the plane reflectors with jet arrays of very high solidity; the area ratio of holes to solid surface reached 98%. Our flow visualization experiments indicate that the jet entrainment and the pairing instability will set up a high-intensity background turbulence at low average rates of flow. The high turbulent intensity is needed to decrease the feature size of the average refractive index fluctuation in the drift region. Near-laminar flow would maintain the large-scale temperature gradients, which will be present at the reflector surfaces because of flashlamp heating.

To measure turbulent scattering in the drift region,⁵⁰ we used much the same Schlieren setup as described above, but in a double-pass configuration. This doubles the scattering angle, which makes it easier to observe the halo of scattered light around the diffraction pattern of the beam-limiting aperture. Figure 6-38 shows the scattered intensity vs temperature difference, where the center of the jet array

Fig. 6-37. Scattering enhancement factor due to surface heat flux for two Mach numbers. Absolute scattered intensities are smaller for lower Mach numbers.

Fig. 6-38. Scattered intensity in the drift region vs center-to-edge temperature difference for various flow rates.

was heated with respect to the edges. The scattering is essentially independent of flow rate. At such low Mach numbers, it is no surprise that temperature-generated refractive-index fluctuations dominate fluctuations generated by velocity fluctuations or compressibility effects. In practice, the jet array will be water-cooled, which will reduce the temperature gradients across the reflector surface to a very few degrees. This will reduce the scattering losses in the drift region to an acceptable level.

The turbulent scattering measurements made so far have given us a good understanding of the magnitude of the effect and of its most important scaling laws. We have shown that under eventual operating conditions for an average power disk amplifier the turbulent scattering losses can be made small enough, particularly if He is used as a cooling gas.

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Pockels Cells

Introduction. Because we hope to scale our electro-optic switches into zig-zag and/or gas-cooled slab lasers operating at the hundreds of watts to multi-kilowatt level, most of our efforts in 1987 were directed towards extending their power-handling capabilities. Although we are investigating quartz as an electro-optic switch material, the greater part of our activity centered around the practical utilization of potassium di-deuterium phosphate (KD*P), since this is the only switch material available at scale and with a low half-wave voltage. Two drawbacks, however, limit the performance of KD*P in high-average-power switches: its intrinsic optical absorption and its low fracture strength. Though the optical absorption is only about 0.5% per cm, it rapidly leads to thermal gradients that exceed the thin-plate fracture temperature of approximately 7°C.

Activity on the rectangular ring electrode (RRE) Pockels cell, developed at LLNL in 1986, focused on improving the first generation of this longitudinal-field KD*P switch architecture, and drew to a close in 1987. We identified several problem areas, which are discussed in the first article of this subsection.

Since crystalline quartz has both negligible optical absorption and high resistance to thermal fracture, we set out this year to determine its suitability in a high-average-power electro-optic switch. To this end we fabricated a quartz Pockels cell and measured its electro-optic coefficients, as discussed in "Quartz Electro-Optic Switch Design." This material is ideal for a switch expected to operate at high optical power, but its power supply requirements preclude it from playing a near-term role in switches that must be operated at moderate-to-high repetition rates.

We began work on a novel KD*P longitudinal-switch design employing plasma electrodes. Because it applies an electric field between glow-discharge plasma electrodes, which are transparent and conducting and are in intimate contact with the KD*P, this approach allows the use of very thin pieces of KD*P,

minimizing the effects of optical absorption on switch performance. We have activated a 10-cm × 1.5-cm × 5-mm thick, single-shot, plasma-electrode Q switch, as discussed in "Plasma Electrode Design."

Since our most promising switch material, KD*P, is very sensitive to thermal conditions, we must evaluate the performance of our switch designs under the type of loading conditions we expect to subject them to in actual use. The goal is to design a switch that will not degrade the performance of the laser; this implies that the wave front must propagate through the switch undisturbed. To evaluate how closely our designs actually come to this performance goal, in 1987 we began the assembly of an experimental facility to quantify the degradation of the wave front while the switch is under full optical load.

Author: M. A. Norton

Rectangular Ring Electrode Design.

During 1987 we continued our development of and support for the rectangular ring electrode Pockels cell for the HAP regenerative amplifier. The RRE architecture, shown in Fig. 6-39, is an extension of longitudinal-field Pockels cell technology.⁵¹ A rectangular KD*P crystal is brought into optical contact with a dielectric-coated metal mirror that acts as both a cavity mirror for the laser system and as an electrical ground plane for the electro-optic switch. In this two-pass switch geometry, a longitudinal electrical field is established along the direction of the beam between the ground plane and a rectangular electrode around the perimeter of the crystal.

The electrode dimensions and the overall crystal length-to-width (L/W) ratio are optimized to maximize the uniformity of the electric field and hence improve the extinction ratio of the Pockels cell between its "on" and "off" states. The electric field symmetry provided by the conductive mirror allows the crystal L/W ratio to be halved from that of a conventional two-electrode longitudinal-field Pockels cell. This reduced crystal thickness allows a fourfold improvement in the average-power

performance of the RRE Pockels cell over that of a conventional longitudinal KD*P modulator.

We addressed a number of performance issues for the RRE architecture during the year. One of the most critical issues was the nonuniformity of the gap between the crystal and the conductive mirror. When an electric field is established in the crystal, a voltage drop occurs across the gap between the crystal and the mirror. Even though this gap is small since the two optical surfaces are in direct contact, fabrication tolerances can cause a gap variation on the order of a few microns between the two components. Because the relative dielectric constant of KD*P ($\epsilon_r = 48$) (Ref. 52) is much higher than that of air, this results in a voltage drop in the gap of tens of volts per micron. A variation across the crystal aperture in the total voltage coupled into the crystal (4.1 kV) can have a serious effect on the uniformity of the electro-optic effect in the crystal and degrade the performance of the switch. We have reduced this problem to a manageable level with tighter flatness specifications for the crystal and the mirror and with improved assembly techniques.

Another problem that we have encountered has been electrical breakdown and tracking between the electrode and the mirror along the surface of the crystal. In the original RRE architecture, this straight-line arcing path was to be protected by a layer of urethane elastomer potted around the perimeter of the crystal. Unfortunately, poor adhesion of the elastomer to the crystal resulted in eventual breakdown of the potting material and the appearance of surface tracking paths between the crystal and the mirror. After extended use, the Pockels cells were unable to withstand the application of the 4.1-kV two-pass half-wave voltage.

In a search of available potting materials, we failed to find one that exhibited good adhesion to KD*P. We have therefore been forced to address this problem in other ways. Movement of the lower edge of the crystal electrode away from the mirror increased the arcing path length with only a small penalty in increased E-field nonuniformity. We rounded the corners of the rectangular

Fig. 6-39. Rectangular ring electrode Pockels cell.

aperture with a 5-mm radius, which reduced field enhancement at these points and improved Pockels cell operating lifetimes. But a more permanent solution lies in the design of a crystal geometry with one or more small steps in the edge of the crystal, which will break up the straight-line arcing path in our RRE design. It is likely that any newly designed longitudinal Pockels cell architectures would use this method to prevent electrical breakdown.

Optical interference between the rear face of the KD*P crystal and the conductive mirror has also been a problem in the RRE Pockels cell. We apply a sol-gel AR coating to the rear face of the crystal to eliminate etalon effects between this face and the mirror (the front surface of the crystal is AR coated and wedged). The high aspect ratio of the HAP regen aperture (10:1) made the spin coating of sol-gel onto the optical faces of the crystal particularly difficult; by reformulating the alcohol carrier used for the sol and by improving fixturing techniques, we were able to apply a uniform sol-gel coating with a transmission peak at 1053 nm

(Ref. 53). Fixturing techniques that maximize the parallelism between the rear face of the crystal and the mirror help reduce the contrast ratio of the fringes generated between the two surfaces (as well as improving the E-field uniformity as previously discussed).

While these measures have improved the reliability of the single-shot RRE Pockels cells installed on the HAP regen, there may be fundamental limitations to the scaling of the RRE beyond the level of a few hundred watts. The crystal-fracture-limited power handling ability $P_{\max}$ of an RRE switch in a regenerative amplifier cavity is given by the formula

$$P_{\max} = \frac{8KA \Delta T_f}{2.7\alpha t^2} = \frac{62A}{t^2} \text{ (watts)}, \quad (9)$$

where K is thermal conductivity of the crystal (0.02 W/cm K), A beam aperture, ΔT_f estimated fracture temperature of the crystal⁵⁴ (6.5°C), α optical absorption of the crystal (0.005 cm⁻¹), and t crystal thickness, which is equivalent to length.

It is interesting to note that increasing the width of the aperture lowers the switch's power-handling ability due to the $1/t^2$ dependence and the lower bound placed on the crystal L/W ratio by field uniformity considerations. The RRE architecture must be scaled along the length of the aperture in order to maximize its average power capability. This argues for the use of high aspect ratio apertures. The RRE is limited in peak power as well as by the damage threshold of the dielectric-coated metal mirror (typically 3 J/cm² for copper).⁵⁵ These limitations in the peak power and average power handling capability of the RRE architecture have forced us to look at other electro-optic switch architectures.

Author: L. F. Weaver

Quartz Electro-Optic Switch Design.

As part of our work on new electro-optic switch architectures, we have investigated quartz as an electro-optic material. Crystalline quartz offers several advantages: the measured optical absorption of our samples is very low at 1.06 μm ($\alpha = 7.0 \times 10^{-5} \text{ cm}^{-1}$) (Ref. 56), so that thermal loading due to absorption is not an

issue for high-average-power operation. The dielectric loss factor for quartz is also very low, resulting in negligible electrical losses in the crystal. Quartz is a very robust material, with none of the thermal stress-fracture problems associated with KD*P.

The chief drawback of quartz is that its electro-optic coefficients are very much lower than those of KD*P or other commonly used materials. We calculated that a practical quartz switch would require electrical driving voltages of hundreds of kilovolts for proper operation; we set up a quartz Pockels cell demonstration experiment to determine exactly what driving voltages would be required. Our Pockels cell used a 4- × 4- × 1-cm synthetic quartz crystal placed in a special electrode structure that prevented electrical breakdown around the edge of the crystal. We employed a transverse field geometry, with the E field applied perpendicular to the direction of optical propagation. The required half-wave retardation voltage at wavelength λ is given by the formula

$$V_{\lambda/2} = \frac{\lambda d}{2l(n_0^3 r_{\text{eff}})}, \quad (10)$$

where d is the crystal width (between electrodes), l the total crystal beam-propagation length, n_0 and r_{eff} the effective electro-optic coefficient for a particular crystal orientation.

We placed the assembly in a pressure vessel filled with 90 psi of SF₆ gas and pulse charged it from a 200-kV Marx bank pulse generator. The RC decay of the voltage on the crystal was monitored with a high-bandwidth, liquid-electrolyte voltage divider.

We double-passed optical beams of 633- and 1064-nm wavelengths through the quartz crystal, as shown in Fig. 6-40, and measured the electro-optic retardation introduced by the pulses for each of the two possible quartz crystal switch orientations (optical propagation along the y or z axis). The full half-wave voltage was reached at 633 nm with the beam oriented along the crystal y axis. At 1064 nm, we found $n_0^3 r_{\text{eff}}$ to have a value of 0.39 pm/V for the y axis orientation and 0.18 pm/V for the z axis orientation. In

Fig. 6-40. Experimental two-phase setup used to measure the half-wave voltage of quartz.

contrast, the corresponding parameter for KD*P at 1064 nm is 40.6 pm/V (Ref. 57).

Using these values, we have estimated that the driving voltage required for an electro-optic quartz switch would be 230 kV (allowing for losses in the electrode structure). While high-repetition-rate, 250-kV pulse generators do not exist at the present time, the development of technologies able to provide such pulses will make quartz electro-optic switches very attractive for high-average-power laser systems.

Author: L. F. Weaver

Plasma Electrode Design.

Introduction. The plasma electrode Pockels cell (PEPC) is a novel design for the electrode geometry of a longitudinally excited Pockels cell. Both crystal faces are completely covered by transparent conducting electrodes, which are created as needed by ionizing a volume of low-pressure gas adjacent to either side of the crystal. The PEPC design results in a longitudinal thermal gradient and a longitudinal electric field.

The PEPC was first demonstrated at a small aperture (2 cm × 2 cm) in a single-shot mode at LLNL in 1983 by Goldhar and Hennesian.⁵⁸ They later demonstrated⁵⁹ a large-aperture (26 cm × 26 cm), single-shot PEPC with a variety of gases operating in the 5- to 30-Torr range. During 1987 we activated a 1.5-cm × 10-cm PEPC with KD*P as the electro-optic switch material.

Because the electrodes in the PEPC are transparent and uniform across the face of the crystal, it is possible to use very thin crystals and still retain the low switching voltage of longitudinally excited KD*P. In fact, the thickness of the crystals can be determined solely by mechanical fabrication limitations and by the dielectric strength of the material. This means that the longitudinal thermal gradients for a given average power will be the lowest achievable in any KD*P architecture.

Following the analysis of the thermal aspects of electro-optic switches by Eimerl,⁶⁰ we can compare the theoretical power-handling ability of a cooled plasma electrode Pockels cell with that of a cooled rectangular ring electrode Pockels cell for the same aperture. We take the aperture to be 1.5 cm × 10 cm and consider a 2-mm thickness for the PEPC and a 1.5-cm thickness for the RRE. It is likely that thermal stress fracture resulting from optical absorption would be the average-power-limiting mechanism of the switches. For the crystal geometry we have chosen, the PEPC has the advantage by a factor of ~50 over the RRE since the power-handling scales as $1/t^2$, where t is the crystal thickness. We project that the PEPC might scale to the multikilowatt level as a switch operating at 100 Hz.

Concept. A schematic of the PEPC is shown in Fig. 6-41. The electro-optic switch material separates two electrically isolated chambers, each of which

Fig. 6-41. Geometry of the plasma electrode Pockels cell.

contains a pair of electrodes. The chambers contain helium gas at low pressure in the 2- to 100-Torr range. The switching voltage is applied between two bar electrodes on either side of the crystal. Just prior to the application of the switching voltage, the pairs of electrodes on either side of the crystal are used to apply voltage pulses, which break down the gas in each chamber: this makes the gas conducting so that the switching voltage can be applied. The cathode on each side is segmented into three pieces to help promote a uniform discharge, while the anode on either side is a single bar.

We spin-coated a z-cut, 5-mm thick KD*P crystal with a sol-gel AR coating and mounted it in a Lucite holder, which slips into the dual-chambered Lucite assembly. The two chambers are connected to an oil diffusion pump via long lengths of polyethylene tubing, which provide electrical isolation; after rough evacuation they are filled with a few torr of helium, which is kept flowing through the chambers at a low rate.

Performance. We measured the optical performance of the cell between two crossed polarizers with a fast photodiode

at several points in the aperture. The PEPC turns on in <40 ns and stays on for approximately $3 \mu\text{s}$. The 40-ns turn-on-time is not a limitation of the PEPC but is determined by the switching time of the driver. We made measurements at six widely separated points across the aperture and found the transmission uniformity to be better than a few percent when the helium gas is at 5 Torr.

Conclusions. The PEPC as presently configured is operating as a single-shot Q switch with more than adequate turn-on time and with good transmission uniformity.

To operate the PEPC at its full potential as a high-average-power cavity switch, the crystal faces will need to be cooled. This is most effectively accomplished by flowing the coolant gas directly across the faces. If a second set of windows are introduced⁵⁹ to separate the coolant from the plasma, then the plasma would be placed on the outside of the coolant chamber. For this case we calculate that half-wave voltages greater than 150 kV would be required for any realistic design of coolant chamber and windows. Thus we project that success in operating the PEPC at its full potential will come only if the plasma and the coolant gas are one and the same. To utilize the plasma as the coolant, we will need to operate the plasma at pressures higher than 5 Torr. To this end we have begun experiments in the 50- to 100-Torr range to look at the transmission uniformity; the preliminary results indicate uniformity of better than 5%.

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Optical Loading Experiment. An inherent problem in optical switching within high-average-power lasers involves the thermal effects resulting from the small but significant optical absorption in the electro-optic switch material. Absorption of the laser pulse heats the crystal, and this heating can in turn distort the wave front. The optical loading experiment (OLE) is an ongoing effort to characterize this distortion.

Fig. 6-42. Wavefront deformation of symmetrically cooled KDP under optical load.

As a two-part system, OLE simulates high-average-power absorptive heating and interferometrically measures the induced distortion of the wave front. The first major component of OLE is a Nd:YAG welding laser that, at $1.06 \mu\text{m}$, provides the necessary optical intensity to simulate the placement of the optical component in a high-average-power laser. The second component of OLE consists of a two-arm Michelson interferometer in which the part to be tested is placed. The interferometer operates with a low-power HeNe laser. During the laser shot a CIE camera records the interferogram, which is transferred to wave-front reconstruction software resident on the Nova VAX. The resulting interference pattern is then processed to give a picture of the wave-front distortion caused by thermal loading.

Our system delivered as much as 9 W/cm^2 to a small area of a sample of symmetrically cooled KDP. Even at this relatively low intensity, the wave-front distortion maps the intensity distribution of the incident beam (Fig. 6-42). In KDP, which has an absorption approximately ten times that of KD*P, there was approximately one wave of distortion for every 3 W/cm^2 of incident optical power difference across the face of the crystal.

With the arrival next year of the new high-power YAG laser for optical loading, OLE will be used to investigate the performance of new switch designs for use at high optical power densities. Additionally, since OLE will record both the thermal source function (the beam spatial

profile) and the response of the electro-optic switch, this data will be used as input to our thermo-optical modeling to benchmark the extension of the codes to anisotropic materials. The OLE facility will then be permanently emplaced as an optical proving ground for all new materials and components destined for use in high-average-power systems.

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Pulse Shaping for the FEL Master Oscillator

High-average-power solid state lasers find application in a variety of missions, many of which involve driving a nonlinear process, such as harmonic generation and Raman conversion. As nonlinear processes can exhibit exaggerated dependence on input intensity, a temporal flat-topped output is dictated for maximum efficiency. The gain-saturation behavior of solid state lasers requires that a temporally shaped pulse be input to the amplifier in such a way that a flat-topped output pulse is obtained. The most desirable implementation would be a continuously variable pulse shape controlled by a computer. What follows is a description of such a system.

Our computer-controlled pulse shaping system is shown in Fig. 6-43. The laser

Fig. 6-43. Computer-controlled pulse shaping for the FEL master oscillator.

pulse originates in a long-pulse (~ 200 ns) oscillator. A portion of this pulse is shaped by a Pockels cell modulator. The result is diagnosed by a photodiode and an oscilloscope. The Pockels cell is driven by a high-voltage, high-peak-power amplifier, which is in turn driven by a computer-controlled waveform generator. A wide range of pulse shapes is possible because we use an arbitrary waveform generator. An example waveform is shown in Fig. 6-43.

Previous pulse shaping systems employed in ICF research^{61,62} have been required to shape pulses with durations on the order of 1 ns. These systems have relied on photoconductive switches and variable impedance transmission lines to accomplish this task. While this approach is necessary and expedient for the ICF application, it is not a good match for the longer pulse lengths required for average-power applications. Longer pulses also suggest less stressing requirements on time response and the possibility of employing a fast, arbitrary waveform generator followed by a high-power linear amplifier. This amplifier would drive a commercially available Pockels cell modulator for shaping the optical pulse.

The baseline pulse length for the FEL master oscillator (FEL MO) is 50 ns. It would be desirable for the waveform generator to make at least ten updates during that time, indicating an update rate of at least 200 MHz. The waveform generator must also have a jitter of less than 0.5 ns with respect to an asynchronous trigger from the master timing system. We could find no commercial unit that would meet these requirements, so we investigated the possibility of designing a custom waveform generator. Our design study revealed that the state-of-the-art in digital to analog converters (DACs), which are the heart of a waveform generator, is being driven by the computer graphics industry. The fastest commercially available DACs are being designed for 1-V video output levels, using emitter-coupled logic (ECL) technology. We found an 8-bit DAC with a 300-MHz update rate and a 0.8-V output into 50 Ω available from Brooktree. They also offered a 256- $\times$ 8-bit pipelined static random access memory (RAM), which ran at 250 MHz and was compatible with the DAC. Next we investigated the ECL required to support the DAC and the RAM and found that 100K was a good choice. Interestingly, 100K ECL has a practical

upper clock speed limit on the order of 250 MHz, particularly when wire-wrap construction is used, as we had planned. These facts all pointed to a reasonable update rate of 250 MHz.

We proposed to meet the jitter requirement of less than 0.5 ns by using a trigger coherent oscillator. Berkeley Nuclonics offers such a device that operates at 125 MHz. We chose to use that device and frequency-double it to generate our master clock. The resulting waveform generator is controlled by custom software running on an IBM PC AT and interfaced through a 24-bit parallel port.

The amplifier requirements are also determined by the baseline pulse length of 50 ns. We would like an output of at least 2 kV into 50 or 75 Ω for an input of 0.8 V with a rise time of 5 ns, which represents a very challenging design problem. Our design is based on power MOSFET transistors (FETs); the devices we used were developed in a cooperative effort between LLNL and Los Alamos.⁶³ They consist of two IRF 440 chips in a custom microwave package with two source leads, one for the gate drive and one for the output. This allows a considerably shorter rise time. The outputs of five FETs are combined with a transmission line transformer that has a 75- Ω output impedance. The input to the amplifier is in the range of 0 to 0.8 V into a 50- Ω impedance. A PNP inverting amplifier stage amplifies this signal to a maximum of 15 V. It is then buffered and fanned out by a voltage follower stage. This stage feeds five current amplifier stages, which drive the gates of the five output stages. The amplifier has an output of 1800 V into 75 Ω , which is approximately 40 kW; this corresponds to a power gain on the order of 65 dB. The rise time is about 6 ns. The resulting amplifier has excellent stability and is remarkably free from oscillations in the output.

The overall system has been demonstrated in the laboratory and meets the present pulse shaping requirement for the FEL master oscillator.

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Faraday Rotators for the FEL Master Oscillator

Introduction. Faraday rotators are used in laser systems for polarization switching and optical isolation. We have used permanent-magnet Faraday rotators in our conceptual designs for the FEL master oscillator. Our Faraday rotator design incorporates practical, readily available magnetic materials and was analyzed⁶⁴ with a validated magnetics code, the 2-D Poisson equation solver PE2D.⁶⁵

Our primary design criteria were to achieve a rejection ratio of at least 100 and to operate at the required time-averaged optical power. This article addresses issues relevant to the required rejection ratio, but not operation at the required average optical power.

Point Design. Figure 6-44 shows the magnet design. The pole pieces are made of Permendur; the permanent-magnet sections are Nd:Fe:B, magnetized in the direction of optical propagation. This design assumes FR-5 rotator glass; a

Fig. 6-44. Faraday rotator design. The rectangular aperture is well suited to zig-zag lasers.

Fig. 6-45. Magnetic flux lines (top and side views) determined with PE2D analysis code.

Fig. 6-46. Integrated field uniformity across the aperture for several axial positions of the 5-cm-long FR-5 rotator glass. The rotator can be tuned by moving the glass axially.

material with a higher Verdet constant V and lower absorption than FR-5 glass would be desirable.

The magnetic flux lines in the top view of the magnet (Fig. 6-45) are parallel and fairly evenly spaced in the region occupied by the rotator glass, as if they originated and terminated at virtual poles. As described below, this "virtual pole" analogy assisted us in analyzing the uniformity of the flux lines in the vertical direction.

The uniformity of the integral $\int B_z dz$ of the magnetic flux density along the optical axis in the region occupied by the rotator glass was evaluated for several axial positions of the glass. Figure 6-46 shows the results of this evaluation, and the nominal required value of the integral. The integrated flux is quite uniform over the area occupied by the rotator glass. If the clear aperture extended from the center of the aperture to ± 0.4 cm (allowing for a misalignment of either the glass or the beam, since the half width of the beam is 0.25 cm), the integrated fractional field variation would be less than 1%. This is true for a range of axial positions of the glass, indicating that it would be reasonable to tune the rotator by changing the axial position of the glass. The integrated field for the centered position is 4% higher than the nominal required value, so there is some design margin. In practice, the length of the glass would be chosen on the basis of actual magnet performance.

It is also necessary to predict the field variation as a function of vertical displacement from the center of the magnet. To do this with a 2-D code such as PE2D, we use the analogy of virtual poles, in which the flux lines passing through the rotator glass originate and terminate

at poles of permanent magnet material. Figure 6-45 shows this view, looking normal to the direction of laser beam propagation. The magnetic flux lines are quite parallel and uniformly spaced in the region occupied by the rotator glass.

The uniformity of the integrated flux in the vertical direction is good. For a clear aperture extending ± 4 cm vertically from the center of the poles, the fractional variation is $\sim 2.5\%$. Below we combine this variation with the variation across the aperture in the top view and with other estimated errors to predict the performance of the rotator as an isolator.

Performance. Our measure of Faraday rotator performance is its rejection ratio R when it is used as an isolator; we have

$$R = \left[T_0 + \sin^2 \pi E/4 \right]^{-1}, \quad (11)$$

where T_0 is the transmission of the polarizers when crossed and E is the error, expressed as a fraction of desired product BIV of magnetic flux density, path length in the glass, and Verdet constant.

We now consider other possible sources of error such as nonuniformity of the magnetic field and temperature variations in the glass and in the magnet. These are differential errors, which cannot be tuned out. We assume that absolute errors in B , l , and V can be tuned out.

The Verdet constant of the rotator glass is inversely proportional to the absolute temperature.⁶⁶ If we assume a temperature difference of 10°C across the aperture, V varies by $\sim 3.3\%$. The magnetic induction of the permanent magnet material varies⁶⁷ with temperature by about $-0.1\% ^\circ\text{C}^{-1}$, so a temperature variation of 10°C within the permanent magnet material implies a 1% variation in the flux across the aperture.

We recall that the fractional variations in the magnetic flux in the horizontal and vertical directions are 1 and 2.5%, respectively. Combined, the four errors imply a variation of less than 8%, which corresponds to a rejection ratio of at least 100 for a crossed-polarizer transmission $T_0 = 0.5\%$. For $T_0 = 0.2$ and 0.1% , the rejection ratios will be approximately 170 and 200, respectively. This meets our design requirement for rejection ratio, as long as T_0 for the polarizers is less than 0.5%.

Shielding. It is prudent to provide magnetic shielding around this component to prevent external magnetic materials such as iron from perturbing the rotator field and to prevent the rotator magnet from interacting with external devices. A shield has not been designed: the boundaries used in the PE2D analysis indicate that a box of soft iron sheet 3 to 6 mm thick, with its sides 8 cm from the sides of the magnet, should be adequate.

Forces. The magnetic field pushes the right and left sides of the magnet apart with a force F given (in S.I. units) by⁶⁸

$$F = \frac{B^2 A}{2\mu_0}, \quad (12)$$

where B is the magnetic flux density, A is the area of the magnet sides, and μ_0 is the permeability of free space. If we take the flux in the center region to be 0.8 T (which is conservative), the resulting force will be 3100 N, or about 700 lb. Dealing with such a force will require care in design, assembly, and disassembly, but should not be a major problem.

Magnetization. The best solution to magnetization is to procure the magnet as an assembled unit. Otherwise, we will have to build our own magnetization facility. Data sheets on the Nd:Fe:B material⁶⁹ indicate that the required magnetizing field is 30 to 40 kOe. This should not represent a major problem, because a magnetizing coil would be similar in size and field to the large Faraday rotators in the Nova laser system.⁷⁰

Summary. We have designed and analyzed a permanent-magnet Faraday rotator suitable for use in the FEL master oscillator. The design meets the rejection ratio requirement of 100.

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Phase Conjugation by Stimulated Brillouin Scattering

In an experimental study of phase conjugation by stimulated Brillouin scattering (SBS), we demonstrated that a conjugate beam with a clean near-field intensity distribution could be obtained by using an SBS medium with a low nonlinear refractive index. We obtained

substantial correction for up to three waves of astigmatism.

Phase conjugation is a promising technique for correcting phase error in laser beams. Propagating a diffraction-limited optical wave through an imperfect optical medium produces a distorted output wave with local electric field amplitude $|E(\mathbf{r})|$ and local phase $\phi(\mathbf{r})$. If there existed a mirror capable of producing the counter-propagating conjugate wave with phase $\phi_{\text{conj}}(\mathbf{r}) = -\phi(\mathbf{r})$, each ray in this conjugate wave would retrace its path back through the imperfect medium, and the conjugate wave would emerge as a counter-propagating replica of the diffraction-limited input wave. The simplest conjugating mirror consists of a lens and an SBS medium. While SBS provides gain for all backscattered, Stokes-shifted Brillouin waves, greatest amplification is experienced by the scattered wave whose phase fronts exactly match those of the incident wave.

Several mechanisms detract from beam correction by conjugation. The SBS gain is intensity dependent and may be too low to provide appreciable reflectance for fractions of the beam that are brought to focus well off axis. The reflected beam may contain both conjugate and nonconjugate beams and may be modulated by interference between these beams. Other nonlinear effects, such as self-focusing, compete with the stimulated scattering.

We believe, in fact, that self-focusing was the primary cause of the failure of a CS₂ SBS conjugator that was fielded in 1986 slab laser experiments.

To examine the influence of self-focusing, we studied phase conjugation by SBS in two liquids, CS₂ and CCl₄, in fused silica, and in methane at 30-atm pressure. The strength of self-focusing is determined by the nonlinear refractive index coefficient γ , which is defined relative to the linear refractive index n_0 by the equation $n = n_0 + \gamma I$, and is usually stated in units of $10^{-16} \text{ cm}^2/\text{W}$. In these units, for the media CS₂, CCl₄, SiO₂, and methane at 30 atm, γ values are 320, 10, 2.7, and 1.4, respectively. For the liquids, values of γ are those measured⁷¹ with pulses having a duration of ~ 10 ns. The value of γ for methane was computed from the value of χ^3 given by Ward and Bigio.⁷² The value of γ for SiO₂ was measured at LLNL with 1-ns, 1064-nm pulses.

In our conjugation experiments we used 1053-nm pulses having 13-ns square waveforms in a collimated Gaussian beam with a Gaussian radius of 7.3 mm. Ripple depth for this beam was $<5\%$ (see Fig. 6-47). For both the input and reflected beams, we measured the pulse energy and recorded the near- and far-field distributions and the temporal waveform.

Injecting this beam into a conjugator consisting of a lens with focal length of

Fig. 6-47. Near-field intensity distribution in the input beam: (a) photograph and (b) lineout.

Fig. 6-48. Near field of beam reflected by a CS_2 conjugator. The input beam did not contain added aberration: (a) photograph and (b) lineout.

Fig. 6-49. Near field of beam reflected by a CCl_4 conjugator. The input beam did not contain added aberration: (a) photograph and (b) lineout.

59 cm and a cylindrical volume of CS_2 with a length of 20 cm produced a reflected beam with severe small-scale modulation (see Fig. 6-48). This modulation was not significantly altered by variation of the input energy from 2 to 100 times the SBS threshold of ~ 0.5 mJ, or by configuration changes in which the position of the beam waist was varied from just inside the cell window to deep within the CS_2 . Use of a phase plate to increase the divergence of the input beam did not eliminate the modulation.

In contrast, beams reflected by CCl_4 were free of small-scale modulation (see Fig. 6-49). Because the modulation was eliminated by substitution of CCl_4 , whose γ value is a factor of 32 times lower than that for CS_2 , we believe the modulation observed in the CS_2 experiments can be attributed to self focusing.

However, when aberration consisting of 1.5 to 4.5 waves of astigmatism was

added to the beam entering the CCl_4 , the reflected beam contained a fringe pattern oriented perpendicular to the upstream line focus produced by focusing the astigmatic beam (see Fig. 6-50). At input powers >5 times the SBS threshold, the reflected beam also contained a second fringe pattern oriented orthogonal to the fringes observed at low power. Moving the downstream line focus outside the CCl_4 cell eliminated this second fringe pattern.

The modulation depth in the fringes on the reflected beam was reduced when either silica or methane was substituted for the CCl_4 . Figure 6-51 shows the beam obtained from silica at 3 times threshold with 3 waves of input: fringing is negligible. However, increasing either the input power or the input astigmatism caused an increase in the depth of modulation of beams reflected from either silica or methane.

Fig. 6-50. Beam reflected by a CCl₄ conjugator. Input energy was 3 times the SBS threshold, and the beam contained 3 waves of added astigmatism: (a) photograph and (b) lineout.

Fig. 6-51. Near field of beam reflected by SBS in silica. Input energy was 3 times the SBS threshold, and the beam contained 3 waves of added astigmatism: (a) photograph and (b) lineout.

Table 6-7. Comparison of conjugator performance. Data is specific to an input, 13-ns, 1053-nm pulse with input energy equal to 3 times the threshold (E_{th}) for SBS and with 3 waves of added astigmatism.

	CCl ₄	SiO ₂	CH ₄ (30 atm)
Near-field modulation depth (at $3E_{th}$)	$\pm 40\%$	$\pm 10 - 13\%$	$\pm 10 - 13\%$
Threshold (mJ)	12	30	30
Operating fluence (mJ) (desire < 100 mJ)	36	90	90
Reflectance (at $3E_{th}$; desire > 0.25)	0.42	0.38	0.38
Far-field correction (at $3E_{th}$)	$< 2 \times$ D.L.	$< 2 \times$ D.L.	$< 2 \times$ D.L.

Therefore, this fringing appears to be a generic characteristic of beams obtained by SBS with an astigmatic input beam. Because substitution of SBS media with lower nonlinear refractive index reduces the modulation depth, self focusing may

also contribute to the formation of the fringes. A model that accounts for all of our observations is not presently available. Our results for materials other than CS₂ are summarized in Table 6-7. Stated modulation depths are measured peak-to-valley excursions divided by average amplitude. Threshold in an SBS experiment is defined to be the apparent zero in a plot of reflected energy vs input energy. We arbitrarily selected energies of 3 times the threshold value as a minimum operating point; better dynamic range is achieved by operating at higher input energies. The apparent equivalence of SBS reflectance for all three materials stems from having stated the reflectance measured at an input energy of 3 times threshold. Material parameters are

Fig. 6-52. Pulse reshaping by SBS in silica and methane. Input energy was 3 times the SBS threshold, and the beam contained 3 waves of added astigmatism.

therefore buried in the determination of threshold energy. While we did not make detailed measurements of the far field, our data show that the divergence of the reflected beams was not more than twice the diffraction limit.

Since we studied only one waveform, a 13-ns square pulse, we cannot make a general statement about fidelity of waveform reproduction. Examples of waveform reshaping are given in Fig. 6-52. The primary effect seems to be a shortening of pulse duration caused by the time required to form the acoustic grating.

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Four-Phase Interferometer for Measurements of Optical Aberrations

In many circumstances, the aberration characteristics of optical components can be measured before installation and thereafter be regarded as fixed. In the context of high-average-power lasers, however, this is not necessarily the case. Thermal gradients present in a high-average-power component, even after it has reached a state of thermal equilibrium, cause optical aberration. The magnitude and transverse variations of thermally induced aberrations can be minimized by optimum design. "Wave-Front Validation with the B-Head Test

Amplifier," earlier in this subsection, describes Fizeau interferometer measurements of the zig-zag amplifier "B" head in its current state of optimization. "Multidimensional Signal Processing" in Sec. 3 explains a Fourier-domain bandpass processing approach for analyzing the interferometric data.

Although it is important to measure component characteristics during the development of a high-average-power laser system, in the final analysis it is the quality of the system output beam that indicates the success of aberration control. One way to measure the wave front of a beam is to generate a high-quality reference from a sample of the beam to be measured and interfere the two. The interferograms from such a self-referencing interferometer can be analyzed unambiguously as described in Sec. 3, "Multidimensional Signal Processing," if a large tilt is introduced in the instrument. However, the many-fringe interferogram so generated is difficult for an operator to interpret directly.

Another way to analyze interferometer outputs is to add phase shifting to the instrument. When one arm of an interferometer is shifted in phase by a known amount and in a known direction, ambiguities about the sign of wave front irregularities can be eliminated. Phase shifting interferometers using continuous-wave (cw) lasers are commercially available for rapid and accurate measurement of optical components and cw beams. Comparable instruments for measurement of the wave front of a single laser pulse, however, are not on the

Fig. 6-53. Optical schematic of a Mach-Zehnder interferometer with large radial shear and optical processing to generate four phase shifted interferograms. The four images are multiplexed onto a single camera.

Fig. 6-54. Four phase shifted interferograms of the same beam from the four-phase interferometer.

Fig. 6-55. The wave front as derived from image processing and subsequent evaluation of Eq. (13) for the breadboard interferometer.

improved wave front against which to compare the initial beam.

In one output path of the interferometer, a 45° rotator and a polarizer divide both the signal and reference beams equally between two separate images on the camera. This generates two interferograms, one polarized horizontally, the other vertically. The former has 180° more phase delay between the reference and signal light than does the latter. A similar rearrangement of phases and polarizations is accomplished in the other output path, in this case with a quarter-wave plate in addition to the 45° rotator. The result is that the phase delays between the reference and signal light differ by 180° in the second two interference patterns and also differ by 90° from their counterparts in the first output path. Thus, the four interferograms, designated A through D, have relative phase shifts between the signal and reference beams of 0°, 90°, 180°, and 270° respectively. The four interference patterns can be equally well obtained from a single short pulse or a cw source.

The phase difference between the signal and reference beams at every point on the wave front is given identically by

$$\tan \Delta\phi = \left[\frac{(1 - R_{sh})R_{sv}}{R_{sh}(1 - R_{sv})} \right]^{1/2} \frac{D - B}{A - C} \quad (13)$$

where A through D are the intensity values from the four interferograms, R is reflectivity, and the subscripts "sh" and "sv" refer to the nonpolarizing beam splitter for horizontal and vertical polarizations, respectively. This result is independent of transverse intensity and polarization variations.

We have built a breadboard interferometer using "on hand" optics to explore the practical limitations of this design, and we have written video image processing software to determine intensity values for substitution in Eq. (13). Figure 6-54 is a set of four interferograms showing the 90° phase shifts between them. In initial tests we used a magnification of 5 and obtained peak-to-valley residual aberrations in the instrument of less than 1/10 wave with an rms deviation from flatness of less than 1/40

Fig. 6-56. An optical layout that implements the design of Fig. 6-53 with optical relays, apertures, and additional polarizers added.

wave, which is representative of the quality of the optics we used.

Figure 6-55 shows the processed data from such a measurement. Low polarization leakage, careful orientation of the quarter-wave plate, precise registration of the four images, and accurate measurements of the splitter reflectivities are important for proper operation. However, it is a property of the design that as the aberration magnitude goes to zero, the error from these four sources also goes to zero. Camera noise, spatially nonuniform response, and nonlinearities also affect performance.

Figure 6-56 shows an optical layout that incorporates several improvements to our original design. Added polarizers ensure polarization purity, and the

Fig. 6-57. Four-phase interferometer images of a high-aspect-ratio rectangular beam such as might be generated by a zig-zag amplifier.

entering beam is optically relayed onto apertures that have been added in each of the interferometer arms. These apertures prevent image overlap in the camera, and optical relays in each arm eliminate the diffraction rings that the

Fig. 6-58. Top view of the optical path taken by a beam as it propagates through a solid state zig-zag amplifier slab. All parts of the beam sample the thermal gradients induced by optical pumping and face cooling.

Fig. 6-59. Three-dimensional representation of the measured, thermally induced optical aberrations in a small, zig-zag, glass amplifier, under repetition-rated pumping conditions.

apertures would otherwise cause. As Fig. 6-57 demonstrates, the instrument is not limited to the examination of circular beams; four phase-shifted interferograms of a high-aspect-ratio rectangular beam can also be generated.

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Deformable Mirror for Wave Front Correction

Development of adaptive optics to compensate for optical aberrations in the atmosphere has proceeded for the last 15 to 20 years. Although they are very expensive, it is now possible to obtain deformable mirrors of ever increasing

aperture, stroke, and bandwidth for that application. However, we are working on laser systems with wave front correction requirements that are significantly different from those related to atmospheric propagation, different with respect to bandwidth, aperture shape, amplitude, and spatial resolution. We have developed a rectangular mirror for aberration correction in a zig-zag, solid state laser amplifier.

Zig-Zag Amplifier Aberrations. The primary source of aberrations in solid state laser amplifiers is the heating associated with optical pumping, although heating due to absorption of part of the beam itself can also be a factor. A major effort in the design of high-average-power, solid state lasers is to minimize these thermally induced aberrations, and the zig-zag amplifier architecture illustrated in Fig. 6-58 is particularly effective in this respect.⁷⁶ Under continuous pumping conditions, a beam that follows the zig-zag path through a slab amplifier is considerably less aberrated than one that propagates straight through. This is because along the zig-zag path all parts of the beam pass through the refractive index gradients that develop across the thin dimension of the slab. Along a straight path, on the other hand, some rays would sample only the center of the slab and others only the edges.

This significant improvement notwithstanding, some residual thermally induced aberrations remain. Figure 6-59 shows the optical quality of a small, zig-zag, glass amplifier measured with a Fizeau interferometer under repetition-rated pumping conditions. In this three-dimensional representation of the distortion in the amplifier, slowly varying

curvature is evident along both the long and the narrow dimensions of the slab.

Rectangular Deformable Mirror. We are working on a rectangular deformable mirror with an approximately 12:1 aspect ratio, to be used in series with a zig-zag glass or crystal amplifier. The mirror is deformed along its long axis by parallel rows of piston actuators referenced to a stiff backplate, and along its short axis by the application of moments, as illustrated schematically in Fig. 6-60. The application of moments at the edge of the substrate to obtain the narrow-dimension curvature is an extension of earlier work by Sawicki.⁷⁷ Figure 6-61 is an artist's conception of the completed mirror, which is designed to the following specifications:

- Aperture: 25×2 cm.
- Actuator spacing: 2.5 cm along the length.
- Surface figure: fabricated flat to $0.1 \mu\text{m}$.
- Correction range: $\pm 10 \mu\text{m}$ along the length of the mirror, $\pm 1 \mu\text{m}$ across the width of the aperture.
- Bandwidth: 10 Hz.
- Cooling: none.

The substrate is fused silica with a high-reflectivity, high-damage-threshold dielectric coating for $1\text{-}\mu\text{m}$ light. Fused silica is also used for the backplate, piezoelectric translator (PZT) support plate, and base, to minimize problems with differential thermal expansion. The actuators for the mirror are PZTs with a $20\text{-}\mu\text{m}$ range.

Finite Element Analysis. The substrate thickness and other dimensions of the mirror substrate must be chosen carefully to obtain the specified performance. If the substrate is too thin, initial figure accuracy will be difficult to achieve, and the mirror will be too sensitive to gravity and other mounting stresses. If it is too thick, the force requirements will be too great for the PZT and attachment interfaces. The legs to which the PZTs attach must be long enough to provide leverage for bending the substrate in the narrow dimension. However, they should not be made longer than necessary because that will make it more difficult to prevent the piston PZTs along the sides from also introducing moments.

Fig. 6-60. Rectangular deformable mirror architecture. In the long dimension, actuators displace the mirror perpendicular to its surface at the edges of the substrate to change its shape. Curvature is induced in the narrow dimension by moments, which are also applied at the edges.

To explore these and other design questions, we have begun finite element modeling of the mirror. Initial calculations are for a section of the substrate with forces applied only in an idealized fashion at the PZT connection points. Subsequent modeling will include gravity, actuator interfaces, and the mirror's stiff backplate. Figure 6-62 shows the calculated deformation of the mirror section when it is pushed up at two central PZT locations and pulled down at each end. The calculation shows that in this simple case a force of 5 lb at each actuator location will result in a smooth curve with an amplitude of $10 \mu\text{m}$. It is also evident

Fig. 6-61. Artist's conception of the assembled rectangular deformable mirror.

from Fig. 6-62 that even though the forces are applied at eight discrete points, the substrate is thick enough to respond smoothly. In fact, in the part of the mirror that will reflect the beam, the contour lines are almost straight. One characteristic of this design is that it naturally produces smooth variations of figure in the beam aperture, even when there are irregularities in the application of the forces or moments, because the actuator interfaces are far from the aperture.⁷⁷

Figure 6-63 shows the calculated result of pushing inward equally on each of the legs on this section of the mirror. The force required to achieve $1\text{ }\mu\text{m}$ of curvature across the narrow dimension of the beam is 20 lb. When equal forces are applied to the four opposing pairs of legs, antilastic bending causes the ends to rise higher than the middle, but this can be countered by small piston forces if desired. In fact, with a control algorithm that calculates desired curvature changes in the mirror's narrow dimension and desired position changes in the long dimension, quite complicated phase errors can be corrected.

The finite element calculations also determine the distribution and amplitude of stress in the substrate. For the cases illustrated in Figs. 6-62 and 6-63, the maximum stresses are less than 1000 psi.

Fig. 6-62. Calculated deformation of a section of the rectangular mirror with a 5-lb force on each piston PZT, as indicated by the arrows. The contours are spaced at $1.3\text{-}\mu\text{m}$ intervals.

Fig. 6-63. Calculated deformation of a section of the rectangular mirror with a 20-lb force on each moment PZT, as indicated by the arrows. The contours are spaced at $1.8\text{-}\mu\text{m}$ intervals.

Substrate Fabrication. We chose fused silica for the deformable mirror surface because a very high optical damage threshold for a laser wavelength of $1\text{ }\mu\text{m}$ can be achieved with dielectric coatings on this substance (or on glass). Making the entire substrate out of fused silica (or glass) is simpler than constructing a layered structure. On the other hand, the back side of the substrate must be ground or otherwise shaped to form the legs to which the PZT's attach. Because fused silica is a brittle material, care must be taken not to leave significant subsurface damage or the part will fracture easily under stress.

We have chosen to use ultrasonic machining, sometimes called impact grinding, to shape the back surface.⁷⁸ This process uses a metal tool that has been machined to match the desired surface features. The tool is then vibrated ultrasonically and advanced toward the substrate while a slurry of grinding compound is flowed between the surfaces. The tool transfers its vibrations to the grinding compound particles, which in turn remove material from the substrate. The required accuracy can be attained even at quite high material removal rates. Because this process removes material by successive fracturing of the surface, the breaking strength of the

substrate is reduced. However, chemical etching of the machined part to remove the damaged layer leads to a final surface with strength higher than standard optically polished surfaces and many times higher than the 1000 psi required.⁷⁹

Piezoelectric Actuators. PZT's are often used as actuators for deformable mirrors because they are relatively compact, capable of exerting large forces, and operable at high frequency. Unfortunately, most piezoelectric materials exhibit significant hysteresis when they are cycled. With additional iterations of the control loop, hysteresis can be overcome, but this means a more costly control system or slower operation; it would be better to eliminate the hysteresis. The PZT's we are using have a built-in strain gauge that measures the actual displacement and provides an error signal for a local closed loop. This loop keeps the motion of the PZT proportional to the control voltage and all but eliminates the hysteresis.

Figure 6-64 shows the measured departure from linearity when one of these PZT's is driven from zero to $18\text{ }\mu\text{m}$ and back to zero at 10 Hz. The maximum deviation from linear response is about 100 nm or $0.1\text{ }\mu\text{m}$, which is just over 0.5%. Measurements on a number of actuators demonstrate that this performance

Fig. 6-64. Measured departure from linearity for a PZT with internal strain gauge feedback, operating at 10 Hz over a range of 18 μm . The width of the line indicates measurement accuracy.

is typical and that at frequencies as high as 100 Hz, the departure from linearity for the reduced-amplitude excursions one might make at this bandwidth is still only a few percent. The force capability of the actuators is ± 70 lb, more than enough to achieve the desired $\pm 10\text{-}\mu\text{m}$ range in the long direction and $\pm 1\text{-}\mu\text{m}$ in the narrow direction.

Conclusion. We have described a rectangular, deformable, fused-silica mirror designed for the correction of thermal aberrations typical of a solid state, zig-zag amplifier. This mirror has an intended operating frequency of 10 Hz and a range of 10 μm . Actuators are spaced 2.5 cm apart in the long direction. Our finite element analysis model of the mirror has verified its performance with respect to correction range, surface smoothness, induced stresses, and required actuator forces. We have tested PZT actuators that have built-in closed loop compensation for hysteresis and have confirmed that they provide highly linear response.

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Laser Design Methodology

Slab-Laser Modeling

Slab-Laser Optical Design. We seek to synthesize, in thermo-optical designs of zig-zag slab lasers, combinations of material properties and geometrical

configurations that minimize optical beam aberrations. The fundamental physical problem encountered in these designs is the sensitive dependence of beam quality on the physical distribution of thermal sources and sinks and on thermomechanical boundary conditions. Edge regions of slabs are especially sensitive, and much of our design effort is directed at resolving problems in the edge regions of glass and crystalline slab materials.

Several problems arise in the edge regions. First, thermal and optical source functions within a slab vary in space near pump boundaries, even when pump intensities are spatially uniform at the illuminated slab surfaces. Second, amplified spontaneous emission (ASE) must be absorbed at slab edges to prevent gain reduction and the growth of parasitic modes within the slab. The absorption of ASE in claddings gives rise to thermal source functions, which must be accounted for in the design so as to minimize beam aberrations. Third, the soft elastomers used to mount slabs in stress-free states at their boundary surfaces can be thermo-optically significant, depending on their geometrical configuration and on their thermal conductivities and radiative absorption properties.

We have obtained 2-D inverse solutions⁸⁰ of the heat equation to characterize thermal fluxes that yield constant slab surface deformations and minimal beam phase aberrations. We have also obtained 2-D and 3-D code results^{81,82} that interpret experimental data and that identify the determinants and the morphology of optical beam aberrations in zig-zag slabs.⁸³ (Also see "Wave-Front Validation with the B-Head Test Amplifier," earlier in this section.) Here we present 3-D modeling results that are used in the design of practical ASE-clad zig-zag amplifiers and describe additions to previous code physics capabilities.

Glass Zig-Zag Slabs. Figure 6-65 shows a baseline configuration that has been used in theoretical and experimental studies of high-average-power glass zig-zag slab lasers. We assume for simplicity that flashlamp pump intensities I_s at illuminated slab surfaces are uniform in space and constant in time. The 3-D pump energy absorption distribution

function $\mathcal{Q}(x, y, z)$ in the slab defines both optical and thermal source functions. Along a photon path denoted by arc length s , we note that $\mathcal{Q}(s)$ is given by

$$\mathcal{Q}(s) = N\rho_D[BC \exp(-C\rho_D s) + (1 - B)D \exp(-D\rho_D s)], \quad (14)$$

where ρ_D is the laser ion dopant density (m^{-3}); B , C (m^2), and D (m^2) are fitting parameters for the efficiency of storing laser energy in solid media⁸⁴; and N is a normalizing factor for pump power absorption. $\mathcal{Q}(x, y, z)$ is obtained either from the MIR3D code,⁸⁵ which traces rays in the flashlamp pump cavity, or from analytical expressions that describe radiation transport in the slab from given illumination sources at the slab total internal reflection (TIR) surfaces.

The peak stored optical energy density ρ (J/m^3) in the slab is related to the absorbed pump power density P_{abs} (W/m^3) by

$$\rho = P_{\text{abs}} Q \delta / f, \quad (15)$$

where

Q = quantum efficiency = λ_p / λ_L ,
 λ_p = average wavelength of pump light,
 λ_L = average wavelength,
 δ = efficiency of storing optical energy, inclusive of ion state decays,
 f = pulse repetition frequency.

The ratio χ of heat to peak stored optical energy in the slab is given by⁸⁶

$$\chi = \frac{1}{\delta} \left[\left(\frac{1}{Q} - \frac{\lambda_L}{\lambda_F} \frac{\tau_D}{\tau_R} \right) + A \right], \quad (16a)$$

or

$$\chi = \chi_0 + A/\delta, \quad (16b)$$

where

λ_F = average wavelength of fluorescence,
 τ_D = fluorescent lifetime of doped material,
 τ_R = radiative lifetime at low doping,

Fig. 6-65. Elastomer and ASE-clad zig-zag slab.

A = added heat (in units of laser photon energy) in the slab caused by absorbing impurity sites, cavity sources of IR, transient ion sites, etc.

The thermal source function Q_A (W/m^3) in an absorber cladding of thickness t_A in the x direction is given by

$$Q_A = \beta I_A f_A \exp(-\beta x_A) \times [1 + \exp[-2\beta(t_A - x_A)]], \quad (17)$$

where I_A is the total flux of photons emitted spontaneously in the laser slab. β is the macroscopic absorption cross section (m^{-1}) for fluorescent photons, f_A is the fraction of spontaneously emitted photons that reaches the ASE cladding, and x_A is a local coordinate in the ASE cladding in the direction normal to its thickness. The exponential term in braces in Eq. (17) represents absorption of photons that are reflected back into the cladding from its external surface. In experimental code validations⁸² it was found that the elastomer used to mount our glass slabs is a weak absorber of pump light. The measured absorption spectrum was used in computations of the elastomer source function Q_{el} .

It is useful to consider first an idealized example in which the side edges of a $1.5 \times 10 \times 32$ -cm silicate glass (LSG-91H)

Table 6-8. Properties of baseline LSG-91H silicate glass zig-zag slab.

Slab dimensions: $1.5 \times 10 \times 32$ cm
 Slab wedge angle: 28°
 Pump dimensions: 10×20.3 cm
 Nd dopant density: $\rho_D = 2.7 \times 10^{26} \text{ m}^{-3}$
 $t_E = 1$ cm
 $t_A = 0.3$ cm; $\beta = 400 \text{ m}^{-1}$
 $B = 0.691$
 $C = 1.38 \times 10^{-25} \text{ m}^2$
 $D = 1.04 \times 10^{-24} \text{ m}^2$
 $Q = 0.6$
 $\delta = 0.55$
 $f = 1$ Hz
 $\lambda_L = \lambda_F = 1.06 \text{ }\mu\text{m}$
 $\tau_D = 286 \text{ }\mu\text{s}$
 $\tau_R = 415 \text{ }\mu\text{s}$
 $A = 0$ ($\chi = \chi_0 = 1.7$)
 $n_0 = 1.56$
 $dn/dT = 2.7 \times 10^{-6} \text{ K}^{-1}$
 $q_{11} = 0.22 \times 10^{-12} \text{ Pa}^{-1}$
 $q_{12} = 1.38 \times 10^{-12} \text{ Pa}^{-1}$

Thermomechanical properties

Glass

$k = 1.03 \text{ W m}^{-1} \text{ K}^{-1}$
 $\alpha_T = 8.4 \times 10^{-6} \text{ K}^{-1}$
 $E = 8.7 \times 10^{10} \text{ Pa}$
 $\nu = 0.237$

Elastomer

$k = 0.2 \text{ W m}^{-1} \text{ K}^{-1}$
 $\alpha_T = 1.5 \times 10^{-5} \text{ K}^{-1}$
 $E = 1 \times 10^6 \text{ Pa}$
 $\nu = 0.48$

Boundary conditions

$T_{\text{coolant}} = T_{\text{air}} = T_{\text{ref}} = 20^\circ\text{C}$
 $T_{\text{Dirichlet}} = 20^\circ\text{C} = \text{fixed temperature of ASE cladding exterior surface}$
 $h_{\text{TIR}} = 3.3 \times 10^4 \text{ W m}^{-2} \text{ K}^{-1} = \text{convective heat transfer (film)}$
 coefficient for heat removal from slab TIR surfaces
 $h_{\text{wedge}} = 10 \text{ W m}^{-2} \text{ K}^{-1} = \text{film coefficient for heat removal from slab}$
 wedge faces to ambient air

slab are perfectly insulated and the wedge faces are effectively insulated by ambient air. The peak stored optical energy is $\rho = 4.9 \times 10^5 \text{ J}$ at 1 Hz; slab TIR surfaces are cooled by turbulent water at 20°C with a convective heat transfer coefficient $h = 3.3 \times 10^4 \text{ W m}^{-2} \text{ K}^{-1}$; and $\chi = \chi_0 = 1.7$ (corresponding to $A = 0$). Table 6-8 summarizes the properties of this slab.

Figure 6-66 presents surface deformations computed for this slab with the BREW code, and temperature contours and beam fringe patterns computed with the TECATE code for the central planes are indicated. The center slab temperature is 40°C , and the maximum tensile stress $\sigma_{xx} = 15 \text{ MPa}$ occurs on the central portion of the TIR surfaces under the pump.

The value of χ is uncertain by perhaps a factor of 2. If we take $A = 1$ (corresponding to $\chi = 2.14$), the center-to-surface temperature decrement and the peak tensile stresses are approximately

twice the values indicated above for $A = 0$. The nominal tensile strength of this glass is approximately 90 MPa.

The transverse fringes in Fig. 6-66 are caused primarily by beam reflections from the deformed TIR surfaces (which are concave as viewed by the beam) and by refraction through the wedge faces (which are concave at the entrance and convex at the exit as viewed by the beam).

A compensation occurs in the components of the deformed-surface optical path difference (OPD). The optical path lengths of the zig-zag beam traversing between TIR surfaces are shorter near the slab edges than in the thermally expanded central region. The displacements normal to TIR surfaces at the slab center are approximately $0.3 \text{ }\mu\text{m}$ greater than at the slab edges. The OPD is compounded N_R times, where $N_R \approx 6$ is the number of beam reflections on the pumped TIR surface regions. The sign of the OPD leads to a converging phase profile.

In the x - z plane, the tip-to-tip slab length is approximately $4 \text{ }\mu\text{m}$ greater near the slab edges than at the slab center. The OPD caused by these wedge deformations results in a diverging phase profile, thus offsetting the OPDs that are incurred because of the TIR-surface deformations.

The shapes of the surface deformations are governed by details of the coupling between thermally generated forces in the slab. These forces are sensitive to several factors, including the dimensions of pumped and unpumped regions, the pump source distribution, the thermal boundary conditions, and the thermal source functions in media contiguous to the slab. We have confirmed numerically the intuitively obvious fact that a bare rectangular slab with a constant thermal source function throughout its volume will have convex deformations on all its surfaces. Our calculations provide us with detailed descriptions of surface curvature in more complex cases.

In practice, a zig-zag slab (Fig. 6-65) is in contact with an elastomer that conducts significant thermal fluxes to or from the slab. We therefore seek to match the thermal source functions in

Fig. 6-66. Temperatures, OPDs, and slab deformations for baseline silicate glass zig-zag slab.

the elastomer and ASE cladding to the source function in the slab edge regions, which is somewhat lower than in the bulk of the slab. Our calculations indicate that, by appropriate choice of composition and geometry, elastomer and ASE thermal source functions can be made to compensate beam phase aberrations. A simplified model of pump light and fluorescence radiative fluxes was used to estimate the ASE and elastomer thermal fluxes in these computations. Experiments that validate these model source functions in an actual zig-zag slab have not yet been performed. As will be seen in the next example, alternative design configurations can be used to eliminate the effects of the uncertain source function Q_{el} , so further optimization results are not presented here for the glass slab configuration shown in Fig. 6-65.

Crystalline Zig-Zag Slabs. We next consider a ten-bounce GSGG zig-zag slab that is to produce high beam quality on a 0.5×5 -cm aperture. The edge design of this slab is constrained by the fact that the refractive index of GSGG ($n = 1.942$) exceeds that of available elastomers. Fresnel reflection of ASE at a

GSGG-elastomer interface must therefore be avoided so that ASE can be transported out of the slab to an absorber that suppresses parasitic oscillations. An effective configuration is obtained for GSGG slabs by exchanging the positions of elastomer and ASE absorbers from the orientations shown in Fig. 6-65. In this new configuration, fluorescence is absorbed immediately by index-matched GSGG ASE cladding on the slab edges, and pump light does not strike the elastomer, so that Q_{el} need not be considered.

Table 6-9 gives the specifications of this baseline GSGG slab. Boundary conditions are identical to those in the glass slab baseline case. The peak stored optical energy density is taken to be 5×10^5 J/m³, and average-power operating conditions are attained at a pulse repetition frequency of 56 Hz.

Figure 6-67 gives computed temperatures, surface displacements (greatly magnified), and beam phase (fringes at $1.06 \mu\text{m}$, single pass). Optical path differences are less than $1 \mu\text{m}$ on a 0.5×5 cm area of the slab aperture for uniform pump intensity at the TIR surfaces. This result was achieved by optimizing pump

and slab dimensions while accounting for the given ASE cladding thickness and absorption coefficient β .

Closer examination of ray-tracing diagnostic data indicates that $(dn/dT)\Delta T$ is

Table 6-9. Properties of baseline Nd:Cr:GSGG zig-zag slab.

Slab dimensions: $0.5 \times 5.6 \times 8.78$ cm
Slab wedge angle: 32°
Pump dimensions: 5.6×4.8 cm
Nd dopant density: $2 \times 10^{26} \text{ m}^{-3}$
$t_A = 0.1$ cm; $\beta = 1200 \text{ m}^{-1}$
$B = 0.954$
$C = 2.99 \times 10^{-25} \text{ m}^2$
$D = 2.85 \times 10^{-23} \text{ m}^2$
$Q = 0.6$
$\delta = 0.84$
$f = 56$ Hz
$\lambda_L = \lambda_F = 1.06 \text{ } \mu\text{m}$
$\tau_D = 242 \text{ } \mu\text{s}$
$\tau_R = 281 \text{ } \mu\text{s}$
$A = 1$ ($\chi = 2.14$)
$n_0 = 1.942$
$dn/dT = 10.1 \times 10^{-6} \text{ K}^{-1}$
$q_{11} = -0.103 \times 10^{-12} \text{ Pa}^{-1}$
$q_{12} = 0.078 \times 10^{-12} \text{ Pa}^{-1}$
$q_{44} = -0.828 \times 10^{-12} \text{ Pa}^{-1}$
GSGG thermomechanical properties
$k = 7.01 \text{ W m}^{-1} \text{ K}^{-1}$
$\alpha_T = 7.5 \times 10^{-6} \text{ K}^{-1}$
$E = 2.1 \times 10^{11} \text{ Pa}$
$\nu = 0.28$

the dominant contribution to transverse OPDs, where $dn/dT = 10.1 \times 10^{-6} \text{ K}^{-1}$ (for GSGG) is approximately 3 times greater than the value in silicate glass. Surface deformations also contribute significantly to the computed OPDs in Fig. 6-67; $\mathbf{q} \cdot \boldsymbol{\sigma}$ contributions are small. Attempts to extend the pump region to within 0.4 cm of the nearest wedge faces were unsuccessful, because longitudinal OPDs approached or exceeded $1 \text{ } \mu\text{m}$ over a significant portion of the slab aperture.

The sensitivity of OPDs to ASE cladding parameters was evaluated further. For the pump and slab dimensions in Table 6-9, less intense ASE thermal sources (lower β , higher t_A , with $\beta t_A = \text{const}$) produce significant transverse OPDs extending well into the uniform gain region of the slab aperture. More generally, our computations indicate that certain other combinations of slab, pump, and ASE cladding dimensions and optical properties can also be optimized to yield small OPDs at maximum power.

Peak tensile stresses in the baseline example were $\sigma_{zz} = 54 \text{ MPa}$, on the external edge surfaces of the ASE cladding.

Fig. 6-67. Temperatures, OPDs, and slab deformations for baseline GSGG zig-zag slab.

These surfaces were maintained at 20°C and were assumed to be free of mechanical forces. Peak tensile stresses on the convectively cooled TIR surfaces were $\sigma_{xx} \approx \sigma_{zz} = 38$ MPa. The nominal tensile strength of GSGG is approximately 200 MPa, so the pumping power can be scaled linearly from these baseline values to any desired margin of safety.

It is important to note that the ASE claddings in this system are assumed to be GSGG, and that slab/cladding bonds must be strong enough to withstand the maximum tensile stresses attained on the TIR surfaces. If the GSGG claddings were replaced by claddings with the thermal and mechanical properties of KG-3 glass, our computations indicate that optical beam quality would be degraded and that premature stress fracture failures would occur.

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Depolarization Modeling.

Introduction. Current designs of high-average-power solid state lasers incorporate electro-optical components such as Pockels cell switches and harmonic frequency converters in addition to the laser amplifier, that are sensitive to beam polarization. Depolarization of electromagnetic waves can cause significant losses in these lasers. To understand and control depolarization in thermally stressed zig-zag laser slabs and in electro-optical components, we have implemented new physics subroutines in the 3-D BREW code.⁸⁷ In this article we describe the physics model used in the subroutines and present computed and experimental beam propagation and depolarization results for a flashlamp-pumped, convectively cooled glass laser slab.

Physics Model. When light propagates through isotropic solids under stress or through anisotropic crystalline solids, the speed of propagation depends on the direction of propagation and on the polarization. For a given direction of propagation, two phase velocities $v_{1,2} = c/n_{1,2}$ are generally associated with mutually orthogonal components of the electromagnetic

displacement field vector $\mathbf{D}$, where c is the speed of light in vacuum and n_1 and n_2 are indices of refraction associated with orthogonal displacement field components. The polarization state is obtained by solving an eigenvalue problem for $\mathbf{D}$, in which the eigenvectors $\mathbf{D}_1$ and $\mathbf{D}_2$ are always perpendicular to the direction of wave propagation $\mathbf{k}$. This 3×3 matrix problem can be reduced to 2×2 , because the third component of $\mathbf{D}$ is always zero.⁸⁸ We then work in the orthogonal frame defined by $(\mathbf{D}_1, \mathbf{D}_2, \mathbf{k})$. The wave equation for $\mathbf{D}$ can be written as

$$\left(\mathbf{M} - \frac{1}{n^2} \mathbf{I} \right) \cdot \mathbf{D} = 0, \quad (18)$$

where

$$\mathbf{M} = \begin{bmatrix} n_0^{-2} + (\Pi\sigma)_{11} & (\Pi\sigma)_{12} \\ (\Pi\sigma)_{21} & n_0^{-2} + (\Pi\sigma)_{22} \end{bmatrix} \quad (19)$$

is the impermeability matrix. The stress tensor σ is of second rank; the fourth-rank photoelastic tensor Π is a material property. In our examples we have

$$n_0 = n^0 + \frac{dn}{dT}(T - T_0), \quad (20)$$

where n^0 is the refractive index of the unperturbed material at reference temperature T_0 and dn/dT is the linear temperature coefficient of the refractive index. BREW neglects higher-order refractive effects.

The polarization vectors of the normal modes of Eq. (18) are eigenvectors of $\mathbf{M}$ with eigenvalues $1/n_{1,2}^2$, and we define $n_{1,2} = n_0 + \Delta n_{1,2}$. A matrix $\mathbf{N} = \mathbf{M}^{-1/2}$ can be defined to be the refractive index matrix, which is given by the approximation

$$\begin{aligned} \mathbf{N} &\approx \begin{bmatrix} n_0 & 0 \\ 0 & n_0 \end{bmatrix} - \frac{n_0^3}{2} \begin{bmatrix} (\Pi\sigma)_{11} & (\Pi\sigma)_{21} \\ (\Pi\sigma)_{12} & (\Pi\sigma)_{22} \end{bmatrix} \\ &= \begin{bmatrix} a & c \\ c & b \end{bmatrix} \end{aligned} \quad (21)$$

Fig. 6-68. Geometry of polarization calculations in a zig-zag laser slab.

for a material that is isotropic in the absence of perturbations. The matrix $\mathbf{N}$ is diagonalized by a similarity transformation:

$$\mathbf{N}' = \mathbf{R}^{-1}(q) \mathbf{N} \mathbf{R}(q) \equiv \begin{bmatrix} n_1 & 0 \\ 0 & n_2 \end{bmatrix}, \quad (22)$$

where $\mathbf{R}$ is the rotation matrix

$$\mathbf{R} = \begin{bmatrix} \cos q & \sin q \\ -\sin q & \cos q \end{bmatrix}, \quad (23)$$

and where

$$q = \frac{1}{2} \tan^{-1} \frac{2c}{b-a}, \quad (24)$$

A displacement field with wavelength λ evolves from $\mathbf{D}(0)$ to $\mathbf{D}(l)$ during propagation along a path of length l according to

$$\begin{aligned} \mathbf{D}(l) &= \exp\left(-i \frac{2\pi}{\lambda} \mathbf{N} l\right) \mathbf{D}(0) \\ &= \mathbf{R}(q) \begin{bmatrix} \exp\left(-i \frac{2\pi}{\lambda} n_1 l\right) & 0 \\ 0 & \exp\left(-i \frac{2\pi}{\lambda} n_2 l\right) \end{bmatrix} \\ &\times \mathbf{R}^{-1}(q) \mathbf{D}(0) \equiv \mathbf{J} \cdot \mathbf{D}(0); \end{aligned} \quad (25)$$

$\mathbf{J}$ is often called the Jones matrix.

Computational Model. Numerical and/or analytical solutions of beam phase and polarization aberrations in medium-average-power zig-zag slab lasers are complicated by two major factors. First, thermal stresses and refractive indices in the slab are spatially variable. Electromagnetic waves therefore propagate along curved trajectories according to the eikonal equation

$$\frac{d}{ds} \left(n \frac{d\mathbf{r}}{ds} \right) = \nabla n(\mathbf{r}), \quad (26)$$

where $\mathbf{r}$ is a position vector and s is the arc length of propagation. Second, thermally distorted slab surfaces require highly resolved tracking of ray refraction at slab entrance and exit surfaces and of ray reflections at zig-zag slab TIR surfaces. Since this physical problem is intractable by analytic methods, a 3-D numerical ray-tracing method is used in BREW.

Figure 6-68 shows some significant aspects of the BREW calculations. Rays of an incident plane wave are traced between reference planes R_1 and R_2 . The polarizations of the incident and emergent fields are designated in terms of their s and p components at R_1 and R_2 , respectively. Numerical computations in

the slab are executed on a 3-D parallelepiped grid mesh. Mesh elements are referred to as grid cells, and corners of grid cells as grid nodes. Slab temperatures, stress tensor components, and surface displacement vectors (with components u , v , w) are computed at all nodal points; these quantities are all null outside the slab.

An incident ray is propagated in air from R_1 to the left wedge face of the slab, where it is refracted into the slab. Propagation within the slab is carried out along those contiguous cells sampled by the ray in its zig-zag path. Double images have not been observed in current applications, so we use the average index of refraction n (determined from the average stress σ) in each grid cell to compute ray trajectories. The calculations are iterated to ensure that the mutually dependent propagation directions and refractive indices are determined self-consistently as the rays propagate along their trajectories through successive grid cells. Path lengths extend to deformed TIR surface and wedge face coordinates. Phase shifts at reflection points are included, although they can usually be ignored in these designs, in which all rays have the same number of TIR reflections.

In the example considered here, all incident ray directions at R_1 are perpendicular to the x and z' axes. During propagation within the slab, rays may deviate from planes that are parallel to y - z . BREW monitors this x -walkoff, but its effects are usually small and are not taken into further account. The refractive index eigenvalues $n_{1,2}$ and the Jones matrix for polarization components are evaluated in each cell and cumulatively updated for each ray according to Eqs. (18) through (26). Jones matrices for each cell are not maintained in core memory beyond their point of application. Beam output intensities are evaluated at R_2 , and intensity isocontours at R_2 are plotted for analysis and for comparison with experimental data.

Results. Optical path differences and beam polarizations were measured in a 1.5- $\times$ 10- $\times$ 32-cm straight-through LSG-91H glass slab mounted in the B-head test bed. The OPD computations for this experiment have been reported previously,³⁰ and the polarization data

Fig. 6-69. Computed and measured beam polarizations from a straight-through LSG-91H glass slab. Incident beam is purely p -wave; pump power = 1600 W(e).

are given in "Wave-Front Validation with B-Head Test Amplifier," earlier in this section.

The slab was pumped at 1600 W(e). Its TIR faces were cooled convectively by turbulent water at 20°C. The incident electric field was linearly polarized entirely in the p component. Figure 6-69 shows observed and computed isocontours of the output intensity of the s component at R_2 . The losses due to depolarization at this power are less than 1% and are confined to less than 25% of the slab aperture area.

We have also validated BREW calculations of beam depolarization in a slab subjected to purely mechanical stress loads. Depolarization is a nonlinear process in solids under stress; our results indicate that the polarized intensity contours are sensitive measures of the stress profiles in slabs. Depolarization is thus a sensitive measure of thermal source functions in zig-zag slab lasers.

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Reflector Modeling. Measured gain in our slab lasers rolls off near the slab edges. Calculations of illumination using our ray-tracing code MIR3D predict much less edge rolloff than we measured. Because fluorescent losses and resonant line trapping may account for this discrepancy, we are investigating these processes but have not yet resolved the rolloff issue. A computer model of these processes is being planned so that reflector designs can compensate for rolloff.

We tested one method of correcting for edge rolloff. It consisted of pump masks or "shades" under the reflector that allow less illumination at the slab middle and more illumination at the slab edges. The MIR3D code was modified to incorporate these shades (see Fig. 6-70). The computer simulations that linked this

modified MIR3D output to our heat and stress codes were quite successful in modeling the results of adding these shades.

Another method of partially correcting for edge rolloff is to design the water channel windows with edge reflectors. This design provides more illumination of the slab's edges and also reduces the reflector cavity losses. Clearly, a 100% reflective mirror plane that extended one side wall of a "birdhouse" reflector all the way through the slab would effectively replicate the entire reflector cavity. (See Fig. 7-109 in Ref. 90 for details of the birdhouse flashlamp-reflector-slab configuration.) In fact, the MIR3D code uses just such a plane of symmetry to calculate a "double birdhouse" reflector such as our C-head amplifier. Theoretically, slab illumination can be made flat across this plane of symmetry, as indeed the C-head calculations show. However, if that plane of symmetry did not extend all the way through the slab (i.e., if it extended only to the cooling channel window), then illumination would drop off near that plane. The farther the edge reflector is from the slab, the higher the losses and, consequently, the lower the illumination at the edge. Compound this by lowering the reflectivity of such a plane of symmetry from 100% to a realistic value, and illumination near the plane decreases even more. The preceding is a fair description of an actual reflector side wall in a single-lamp birdhouse configuration and of its consequent tendency toward edge rolloff.

Building good quality edge reflectors into the water channel windows is one way to alleviate edge rolloff. We have designed and constructed a slab laser with these edge reflectors: the gadolinium scandium gallium garnet (GSGG) crystal laser. Although laboratory tests showed some decrease in edge rolloff, there were other difficulties. One of these difficulties may be the result of the fabrication tolerances, especially at the sharp edges where the two birdhouse roofs are joined in multiple lamp designs. Reflector performance is surprisingly sensitive at this joining, as observed in the GSGG crystal and also in the C-head amplifier. The

Fig. 6-70. Calculated relative gains of the B-head amplifier across a 100-mm slab width.

Fig. 6-71. Calculated relative gains of the C-head amplifier across a 250-mm slab width.

MIR3D code was modified to simulate imperfect welds along the joining of these adjacent roof planes.

Figure 6-71 compares the calculated illumination patterns for a perfect C-head reflector with those for a simulated imperfect (almost as-built) reflector. We expect a similar result (peaks under each lamp and valleys under each inside roof point) for the GSGG crystal laser, which has a quad birdhouse reflector.

A possible solution to this fabrication tolerance constraint is to construct reflectors using solid quartz instead of welded metal planes. One such reflector has been fabricated and is being tested. However, because it was designed before the edge rolloff problem was identified as critical, it will require shading to compensate for the rolloff. A bonus when using solid quartz construction is that a separate water channel may not be necessary. This naturally extends the reflector sides very close to the slab, making edge pumping easier. Furthermore, deterioration of the reflective surfaces over time is greatly reduced.

In summary, future reflector designs should incorporate solid quartz construction, should exclude water channel windows, and should compensate for edge rolloff.

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Control of Parasitics.

Background. Parasitic oscillations arise in laser systems when optical gain is greater than optical loss along some undesired path in the system. Optical power builds up along that path and can become high enough to deplete stored energy or to damage portions of the system. Because gain is required for proper laser operation, parasitics must be controlled by adequate losses along all possible parasitic paths.

Such losses typically arise from reflections that attenuate or change the direction of the parasitic. The parasitic threshold can be increased by reducing the reflectivity or changing the reflection geometry. Reflectivity can be reduced by placing an absorber at the point of reflection, by matching the refractive index of

Fig. 6-72. Typical zig-zag laser slab of rectangular cross section (A-A). The ends are wedged to allow the laser beam to enter and exit.

Fig. 6-73. Oscillation path inside the slab on the surface A-A of the preceding figure. If the ratio n_1/n_0 of refractive indices inside and outside the slab is greater than $\sqrt{2}$, such paths are lossless.

the absorber to that of the laser medium (if the parasitic is inside the medium), and by placing antireflection coatings on interfaces.

It might also be possible to fill the laser medium with a saturating absorber that stops parasitic oscillation but bleaches to allow the laser beam to pass with low loss; no such absorber has been identified for solid state laser systems.

Parasitics in Zig-Zag Slabs. Rectangular slabs such as those used in the high-average-power project are vulnerable to parasitic oscillations that take place entirely within a single slab. For example, if a slab of rectangular cross section (Fig. 6-72) is placed in air with no absorbers on the flat surfaces, lossless oscillations can take place in any plane normal to the propagation axis if the slab index n_1 is greater than $\sqrt{2}$. This is so because the angle θ_t for total internal reflection (TIR) is

$$\theta_t = \sin^{-1} \left(\frac{n_0}{n_1} \right), \quad (27)$$

where n_1 and n_0 are the refractive indices inside and outside the slab, respectively. When $\theta_t = 45^\circ$, lossless oscillations are possible (Fig. 6-73). The index ratio is then

$$\frac{n_0}{n_1} = \sin 45^\circ = \frac{1}{\sqrt{2}}, \quad (28)$$

Fig. 6-74. Laser slab with edge cladding to suppress parasitics and with coolant flowing past pumped faces.

and if $n_o = 1$ we have $n_i = \sqrt{2}$. Since most laser media of interest have refractive indices greater than 1.414, it is clear that we must take measures to protect energy storage slabs against parasitic oscillations. (Simple free-lasing systems never store enough energy to have enough gain for parasitics, but their spatial and temporal beam quality is low.)

It is difficult to analyze all possible parasitic paths in even a relatively simple geometry such as that of a zig-zag slab. A path can be at any point in the slab, oriented in any direction, so we must search in five dimensions—three spatial and two directional. If we split each dimension into only 100 parts, we must track 10^{10} paths! And with such coarse subdivision we are likely to miss a parasitic that occupies only a small part of the phase space. For example, a parasitic might oscillate only over an angular range of a few tens or hundreds of micro-radians if its spatial size were great enough to keep diffraction low; finding it would require angular subdivision into 10^5 or 10^6 parts.

We also need some way of deciding when we have found a parasitic. The requirement that a ray path close on itself is often used, but in fact parasitic oscillation can take place without such closure. We can see why by considering a mid-plane oscillation at 45° , which does not close on itself unless the ratio of the lengths of the sides of the bounding rectangle is a rational number. When the slab has tilted sides or wedged ends, the paths have a fractal character that leads to extreme complexity.

How, then, are we to proceed? One way is to use brute force: with a fast

computer, trace 10^{10} rays far enough to get an idea of their gain and loss (see "Calculations of Amplified Spontaneous Emission Rates in Laser Disks," in Sec. 5). Save the worst rays (those with the lowest loss per unit gain length), and estimate diffraction effects by finding the area on a plane normal to each such ray over which parallel rays have similar gain and loss. By assuming that all dangerous parasitics pass through a plane cutting the slab (perhaps the mid-plane of Fig. 6-73), we can reduce the dimensionality of the phase space that must be searched to 4 and need only trace 10^8 rays, but the computational effort is still considerable.

Another way to proceed is to simplify the problem. If we can solve a simpler problem in which the parasitics are likely to be worse than in the actual slab, and if the parasitic threshold in the simpler case is above the normal operating gain, we have reasonable assurance that the actual slab will be free of parasitics. This is the approach used here.

Our simplification consists of assuming that the oscillations occur in a zig-zag slab of rectangular cross section with input and output ends having the same wedge angle (Fig. 6-74). Because the laser beam enters and exits through the wedged ends, the only parasitic control that can be applied there is a coating that maximizes the transmission of the slab wavelength for both polarizations at all angles. (Such a coating cannot, however, change the angle at which TIR begins nor the near-100% reflectivity above that angle.) Because pump light must enter through the large flat surfaces of the slab and because the slab is cooled on these surfaces, the parasitic controls available there are (1) a coolant whose refractive index is high enough to increase the TIR angle to a safe value, and perhaps (2) mild, laser-wavelength-only absorption in the coolant.

Because of the minimal parasitic control available from the wedged surfaces and large surfaces of the slab, most of the control must be achieved on the narrow edge surfaces. A parasitic control layer must be added to these surfaces that will absorb as much of the laser wavelength light as possible, for both

Fig. 6-75. If the edges are parallel to each other and perpendicular to the pumped faces, we need only keep track of the ray locations projected onto a plane parallel to the edges.

polarizations and at all angles of incidence. (The layer must also resist the large thermal load of the amplified fluorescence that it absorbs, but we do not consider the layer's thermal design here.)

The best possible parasitic control layer is a good absorber of the laser wavelength and has the same refractive index as the slab material. If the indices are not matched, Fresnel reflection at the interface will return some of the parasitic power back into the slab before it can be absorbed. Worse, if the index of the edge cladding is lower than that of the slab, rays far enough from normal incidence to undergo TIR will be returned to the slab with no loss at all.

Simplification. When the slab edge planes are parallel to each other and perpendicular to the large planes, the ray paths are greatly simplified. Any reflection off the edges changes only the ray component normal to the edge planes. This component therefore has only two values, which differ only in sign because reflections off the large planes or wedge planes do not change its magnitude.

Thus we need not keep track of this component on each bounce, and can do all our ray tracing in a plane parallel to the edge planes (Fig. 6-75). We also see that the worst possible rays are those that strike the edges at just the TIR angle, because such rays suffer no loss during edge reflection, have the longest gain paths for lossless rays, and strike the potentially lossy large planes and wedge planes at the largest angle off normal, thus suffering the lowest possible loss. If we consider only such TIR-angle rays, then any ray's starting point can be specified by its height h on a reference plane and its angle θ with respect to that plane (Fig. 6-76).

We therefore have a set of simplified worst-case rays that require only two dimensions for their specification rather

Fig. 6-76. With the simplifications of Fig. 6-75, the starting position of a worst-case ray (one that strikes the edge at just the TIR angle) is specified by its height h on a reference plane and its angle θ with respect to that plane.

than the original four. For 100 points per dimension, we must trace 10^4 rays rather than 10^8 , and we can do the problem on a desktop computer.

Calculation of Worst-Case Parasitics.

We find the parasitic threshold for edge-TIR-angle rays in a rectangular slab as follows. We specify a reference plane at one end of the slab, at the point where the wedge begins. We consider rays starting at various heights on this plane, and for each height we consider rays starting at all possible angles pointing into the wedge end. Each such ray represents all rays starting at the same height with the same angle, since at the edges all such rays undergo lossless reflections that change only the ray component perpendicular to the edges. All such rays therefore have the same projection on a plane parallel to the edges.

The rays are all started at the TIR angle to the edges, since they will then have the lowest possible loss. If the refractive index of the edge cladding is higher than that of the slab, TIR does not occur, and the rays are started with no component toward the edges.

Each ray is traced through the wedge end by bouncing it off the flat continuation of the pumped surface, if that will be the first surface it strikes. If this is not a TIR reflection, Fresnel's reflectivity corresponding to the refractive indices of the slab and coolant is used (this is the worst case). The ray is now guaranteed to hit the wedged surface. The calculation

Fig. 6-77. Typical plot of g_{th} from simplified worst-case calculation for 30 passes in a typical GSGG slab with an intermediate-index edge cladding. Lowest threshold occurs for $h/T = 0.5$ and $\theta = -6.03^\circ$.

next enters a loop in which intersections with the flat and wedged surfaces alternate, with tests at each surface to see if the ray will leave the wedge. At each reflection, TIR (100%) or Fresnel s reflectivity is used, with the outside refractive index being that of coolant or air.

Once the ray leaves the wedged end, it bounces down the slab until it reaches the other end. Rather than calculate each bounce separately, it is quicker to determine the number of bounces and their reflectivity (which is the same for each bounce) and do the whole process in one step. If we calculate to an intersection with a reference plane at the entry to the other wedge, the tracing can continue as though the ray had just started at this plane with its new height and angle.

Suppose we have followed a ray for N passes from one reference plane to the other. The j th pass will have a total reflectivity R_j that is the product of all the (equal) individual reflectivities at each bounce. The total reflectivity after N passes is then

$$R_{tot} = R_1 R_2 R_3 \cdots R_N = \prod_{j=1}^N R_j. \quad (29)$$

If the path length through the gain region of the slab in the j th pass (presumed to be in the center portion of the slab, not the wedges) is l_j , if the gain coefficient is α , and if diffraction effects are negligible (again, the worst case), the condition for the threshold of power growth on the ray is

$$R_{tot} \exp[\alpha(l_1 + l_2 + l_3 + \cdots + l_N)] = R_{tot} \exp\left(\alpha \sum_j l_j\right) = 1. \quad (30)$$

The threshold gain coefficient is therefore given by

$$\alpha_{th} = \frac{-\ln\left(\prod_j R_j\right)}{\sum_j l_j} = \frac{-\sum \ln R_j}{\sum l_j}. \quad (31)$$

If the gain region fills the whole cross section of the slab, then the gain of a ray depends only on its direction cosine γ

Fig. 6-78. Expanded 2° region around $\theta = -6^\circ$ for the calculations of Fig. 6-77.

along the length of the slab and is given by $\alpha L/\gamma_p$, where L is the length of the gain region along the slab. Thus we have

$$\alpha_{th} L = g_{th} = \frac{-\sum \ln R_j}{\sum \frac{1}{\gamma_j}}, \quad (32)$$

where we can identify g_{th} as the one-pass gain along a ray proceeding straight down the length of the slab, at the point where the parasitic ray crosses threshold.

Results. Figure 6-77 shows a typical plot of g_{th} from the simplified worst-case calculation just described. We followed rays for 30 passes in a slab with dimensions and refractive indices appropriate to gadolinium scandium gallium garnet with an intermediate-index edge cladding. Values of h/T (where T is the slab thickness) ranged from 0.1 to 0.9. The variation of threshold gain with initial angle θ is very complicated. The lowest-threshold case is for $h/T = 0.5$ and $\theta = -6.03^\circ$.

If we expand a 2° region around $\theta = -6^\circ$, we get an almost equally complicated

plot (Fig. 6-78). In a 0.2° region (Fig. 6-79), the threshold gain jumps up and down for changes of θ of 0.001° or less. The lowest-threshold region for a given initial ray height is about 0.06° wide; its position varies slowly with h , giving the broad region of lowest threshold shown in Fig. 6-79.

To see why the structure of the parasitic threshold is so extremely complicated, consider what happens after one pass along the length of the slab. As we change the initial angle of the ray, the point at which it strikes the wedge at the other end changes, and this in turn changes the angle at which the ray returns. The return angle changes abruptly whenever the return ray switches from a flat-surface bounce to a wedge-surface bounce just before it starts back. If the slab length is S and its thickness is T , such a switch occurs for an angle change of order T/S . When the ray returns to its starting end, more abrupt changes take place, but now the angle change involved is of order $T/2S$, because the lever arm is twice as long. The actual ray path per pass is longer than S , so we expect abrupt threshold changes after N passes for initial angle changes smaller than T/NS . In fact, T/S for Fig. 6-79 is about

1/14, and for 30 passes we would expect a feature size of somewhat less than 0.14° , which is just what we observe. As we follow more and more passes, the threshold structure becomes more

and more complicated (Fig. 6-80). It is, in fact, a fractal structure—one that becomes more and more complicated as we examine it more and more closely. In an actual slab, the parasitic will smear out

Fig. 6-79. Expanded 0.2° region around $\theta = -6^\circ$ for the calculations of Fig. 6-77. Threshold gain jumps up and down for changes of θ of 0.001° or less. Lowest-threshold region for a given h is $\sim 0.06^\circ$ wide; θ for this region varies slowly with h , giving the broad region of lowest threshold shown.

Fig. 6-80. For more passes (here, 60), threshold structure becomes more complicated.

by diffraction and will sample the average gain, loss, and geometrical behaviors corresponding to a range of initial angles. We can therefore get a reasonable estimate of worst-case thresholds by tracing rays for a pass count that gives a diffraction spread equal to the feature size; this is why 30 passes were used in the example.

The abrupt threshold jumps with initial ray height and angle mean that we cannot use standard search methods to find worst-case rays by looking close to known bad cases for worse cases. The complicated structure also means that there are unclosed ray trajectories that can have average thresholds as low as those of closed trajectories; it is even possible for an unclosed trajectory to have a lower threshold than a closed one for enough passes that a parasitic can grow to dangerous strength without ever repeating its path.

All these conclusions about complicated dependence on initial conditions, difficulty of worst-case search, and unclosed paths apply to full 3-D cases as well as to the worst-case 2-D cases we have been considering.

Figure 6-81 shows the result of running the angle/height worst-case search for a slab while decreasing the refractive index of the edge cladding from a matched value to a low value. As the index drops below the matched value, the parasitic threshold gain falls off rapidly. A slower drop then takes place, followed by another abrupt drop to zero. Below an edge cladding index of about 1.59, lossless parasitic oscillations are possible. This plot allows us to choose an edge cladding that will be safe against parasitics for any desired laser gain.

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Fig. 6-81. Worst-case parasitic gain as refractive index n of the edge cladding varies from a low value to a matched value. Below $n \sim 1.59$, lossless parasitic oscillations are possible.

zig-zag glass amplifier described in the 1986 Laser Annual as the "C-head."⁹¹ It has since been rebuilt with a new LHG-5 slab and is used for laser research and development. Our main thrust in 1987 was to characterize the test oscillator beam, identify sources of beam degradation, and develop techniques to optimize beam quality. In addition, we measured the gain performance and energy storage of the new glass for comparison with the previous system.

Several diagnostics were brought on line for this work. Figure 6-82 shows the regenerative cavity and associated diagnostics, which include an interferometer, a calorimeter, near- and far-field cameras, alignment sensors, and fast diodes for making temporal measurements. The data obtained on system performance and the diagnostics themselves have application to other multipass amplifier systems now being planned.

There are several differences between the amplifier used in these tests and that reported on previously. Our slab was doped higher (6.5 vs 5.0 at.%); we did not use coated lamps, antireflection-coated flow channel windows, or a flashlamp prepulse. In addition, we fired the pulse-forming networks at 500 μ s instead of 250 μ s.

Figure 6-83 shows the results of our small-signal gain measurements, which are plotted for comparison with previous C-head gain data.⁹² The present results are about 30 to 40% below the previous measurements; comparison of these two pieces of data helps quantify the effect of the differences between the two

Laser Systems and Applications

Free-Electron Laser

Test Oscillator Performance. The test oscillator is a regenerative laser at the heart of which is a 70- $\times$ 25- $\times$ 2.12-cm

Fig. 6-82. Optical schematic of the laser system used in the phase conjugation experiment and the subsequent beam quality measurements. The portion of the system ahead of the apodized aperture includes a long pulse oscillator and a double-passed rod amplifier.

amplifiers. We determined the stored energy using the relation

$$E_s = \ln(g) \frac{hcA}{(\lambda\sigma)}, \quad (33)$$

where g is the small-signal gain, A is the beam aperture, $\lambda = 1.054 \mu\text{m}$, and $\sigma = 4.1 \times 10^{-20} \text{ cm}^2$. The storage efficiency reached a maximum value of 5.4% when the flashlamp pump energy was 6 kJ. Optimizing the features mentioned above would, of course, increase the storage efficiency.

We carried out measurements of the amplifier phase front using a Fizeau interferometer. We put one mirror in the P^* Pockels cell location (Fig. 6-82) and inserted a splitter in the beam at the input to the slab. Interferograms were recorded with the near-field CID camera shown in Fig. 6-82. We made several measurements,

including phase-front vs flashlamp power (Fig. 6-84) and phase-front vs slab temperature gradient. The results shown in Fig. 6-84 indicate that there are temperature differences across the slab (in the coolant flow direction) of 1 to 2°C depending on the level of pumping. Elimination of these temperature gradients and improvement of the amplifier phase front will require improved pumping uniformity, reflector cavity masking to compensate for residual nonuniformity, and using laser glass less sensitive to thermal gradients.

During these phase measurements, we also recorded the far-field beam quality. Both the incident beam and the amplifier return beam were monitored on the far-field CID camera shown in Fig. 6-82. As expected, the return beam was sharply degraded as the phase front errors increased. With the unpumped slab double

passed, the far-field spot was a classic $\sin^2 x/x^2$ function, about 1.7 times the diffraction limit in width. As the pumping level was increased, the far-field return gradually degraded into a more or less unanalyzable smear. Correction of the amplifier phase front will, of course, help correct the far-field spot as well.

To achieve high-quality beams, we will need to correct residual aberrations. Present CCl_4 phase conjugators seem to do a good job of correcting about 3 to 4 waves of aberration⁹³; for the larger phase errors expected to occur at higher pumping rates and many passes, it may be necessary to include an adaptive optic in the cavity instead of allowing the aberration to accumulate over many passes before the beam is reflected from the phase conjugator. Adaptive optics can correct low temporal and spatial frequency aberrations.⁹⁴ We can then control the residual higher frequency aberrations by phase conjugation.

A typical mode of operation with the test oscillator is to have the pulse pass through the cavity 18 times before extracting it. After the first nine of these 18 passes, the P^* Pockels cell switches the beam to a phase conjugation cell. With sufficient drive, this conjugator will reflect 20 to 50% of the beam back into the cavity, where it is amplified during the the last nine passes through the system. Controlling beam quality during this operation requires careful relaying, spatial filtering, and beam apodization, and a very high-quality total internal reflection (TIR) surface in the amplifier. Each of these features exerts its own set of demands on the system. For example, in order to accomplish the spatial filtering, the alignment system must be capable of sufficient accuracy to pass the beam through the filter pinhole 18 times. And, of course, the cavity optics must be stable enough to maintain this alignment for a usable period of time.

Figure 6-85 is a photograph of the cavity output beam with no phase conjugation. For this photograph we relayed the input aperture to the Pockels cell planes. In addition, we apodized the input aperture to eliminate diffraction at the

Fig. 6-83. Small-signal gain measurements for the C-head slab. The SSA model is for the case with prepulse and uncoated lamps. The 1986 data were taken with prepulse and coated lamps. The 1987 data were taken with no prepulse and uncoated lamps.

Fig. 6-84. C-head phase-front distortion vs flashlamp power. The data were taken at nominally constant pulse energy; the power level was increased by increasing the pulse rate.

Fig. 6-85. Beam photograph taken at the output of the test oscillator. The exposure for this unmagnified beam was about 30 mJ/cm^2 . There was a $900\text{-}\mu\text{m}$ -diam pinhole in the spatial filter, and the input aperture was apodized.

amplifier input (which was not at a relay plane). The spatial filter pinhole was 7.5 times larger than the diffraction limit. Severe modulation of the beam, seen in this photograph, was the result in part of degradation of the amplifier TIR surface caused by contact with the cooling water, and of diffraction due to clipping at the slab aperture. Additional sources of beam degradation may be present but are difficult to identify in the presence of such severe modulation. This illustrates the necessity of a very high-quality slab in a system that interacts with the beam many times (288 times in this example). In order to control this aspect of beam quality, it will be necessary to preserve the TIR surface with an evanescent-wave coating and perhaps to limit the number of passes through the cavity.

During operation of the test oscillator this year, it has become apparent that beam quality is impacted by several factors in a multipass system. The quality of optical surfaces becomes critical in that imperfections contribute small aberrations in the pulse on each pass. Thermo-optical effects can produce phase errors. Diffraction must be dealt with even though the remedies may complicate the system and compromise the amplifier's clear aperture. Development of techniques to control beam quality is crucial to optimize the performance of systems like the test oscillator.

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Submarine Laser Communications

Introduction.

General Considerations. Over the last 10 years general system studies have helped evaluate the desirability of using 455- and 459-nm radiation for underwater communication applications.⁹⁵ These wavelengths are optimum for two reasons: First, they are in the optical transmission window of seawater; and,

second, they match up with the cesium atomic-resonance filter, which is a proven narrow-bandwidth filter with a high angular acceptance. The transmission window of seawater actually lies in a range of about 430 to 490 nm. However, in spite of much effort to find other filter/receiver schemes,⁹⁶ the cesium filter at 455 and 459 nm still offers the best noise rejection and therefore sets the frequency requirement. On the basis of these considerations and the technology available at the time, the Block 1 system (the initial laser transmitter system) was proposed.

The Block 1 system achieves the desired wavelength by Raman-shifting the output of an XeCl laser with lead vapor. The overall efficiency is projected to be about 1%, and the estimated cost of a 10-yr mission is \$6 billion. This includes the costs necessary to build, test, launch, and maintain a four-satellite constellation.⁹⁷ In spite of the high cost, submarine laser communications (SLC) mission objectives were considered of major importance, and the Block 1 proposal was funded.

Block 2 (Advanced Transmitter System). During the last several years, semiconductor laser technology has experienced major improvements in device efficiency, output fluence, and lifetime; consequently, semiconductors are very attractive as long-lived pump sources for solid state lasers. These developments, combined with previous solid state laser technology, led us to propose an advanced SLC transmitter which we felt would substantially reduce life cycle costs and substantially improve communication efficiency. An all-solid-state design would be lighter, would have a superior operating lifetime, and would offer billion dollar returns on the research and development investment. In addition, we felt that this effort would advance solid state laser technology and would demonstrate its viability for other Navy/DOD/industry applications. We proposed to assess the critical technologies, to establish the criteria for system selection, and, if justified, to develop a full-scale advanced transmitter system (Block 2) that would meet all technical

requirements. When our proposal to evaluate this further was accepted, we began to explore the issues.

The first task was to assess all possible host/dopant/frequency-conversion schemes within the performance envelope of the SLC application. The desired properties follow:

- Dominant spectral gain feature at 911 and 919 nm or 1366 and 1378 nm for simple frequency conversion.

- Stimulated emission cross section of $\sim 1 \times 10^{-19} \text{ cm}^2$ for efficient operation at low dopings.

- Metastable lifetime greater than a few hundred microns to allow economical use of diode pumps.

- Excitation pathways permitting efficient pumping with long-lived sources, such as AlGaAs diode arrays.

- Crystal host scalable to dimensions typically $1 \times 4 \times 20 \text{ cm}$.

We evaluated many candidate active ions, including Nd-, Pm-, Ti-, and Cr-based systems. We considered frequency-doubling and tripling crystals, as well as Raman shifters, as possible elements. We examined tunable systems and fixed-wavelength systems. We ranked them according to overall system performance and cost to develop and deploy.⁹⁸

We concluded that a promethium-based solid state laser would be a factor of 3 more efficient and smaller than the neodymium-based systems. Furthermore, it would be 6 times more efficient than the XeCl + Pb baseline system, and

would be simpler, lighter, more compact, and longer lived. Taken together, these factors result in a significant reduction in cost over the Block 1 system.

Block 2 Technical Assessment of Pm SLC. Several technical issues had to be addressed before a low-risk design could be projected. First, because no one had made a laser using promethium, we had to demonstrate that it would lase. Second, even if a laser were demonstrated, it had to operate efficiently at the required wavelength. We had to confirm that the radiation effects on the host lattice and on the diode pump sources were manageable. Furthermore, the diode array pump sources currently being produced are matched to the neodymium pump bands at 808 nm; the shorter wavelength material needed for promethium had to be demonstrated and lifetime tested. Finally, the actual cost of obtaining ^{145}Pm had to be determined. In our judgment, a successful promethium Block 2 laser system could be demonstrated and each of these issues would be favorably resolved.

We set up an experimental program to evaluate these technical issues. We began to synthesize ^{147}Pm -doped crystals and glasses and to characterize their emission and absorption properties. Our goal was to obtain extensive crystalline and glass host data on Pm^{3+} and to verify our projection that a Pm laser would be compatible with the Block 1 receiver. We initiated a contract with Oak Ridge National Laboratory to fabricate these

Fig. 6-86. Schematic summary of the general architecture of the promethium Block 2 system and the corresponding technical issues.

samples. In addition, we demonstrated laser performance and began to quantify the fundamental laser parameters (gain, loss, storage lifetime, etc.) so that we could project a point design based on measured performance. We began to evaluate performance and growth issues surrounding the harmonic doubler. We measured the appropriate neutron capture cross sections and estimated "not to exceed" costs for ^{144}Sm enrichment, ^{145}Sm production, and ^{145}Pm chemical separation. We evaluated γ -induced optical losses in suitable hosts at doses approximating mission dose. We contracted with diode manufacturers to verify the efficiency and lifetime of the short wavelength diode pumps. Subsequently, we evaluated the benefits of a Pm-based SLC in terms of size, weight, lifetime, and the impact on overall system costs. In summary, Fig. 6-86 illustrates the general architecture we were considering and the technical issues we felt needed further resolution.

During 1987, we evaluated these high-risk technical issues in detail, and we resolved them favorably. We can confidently project a design that will meet the mission requirements and result in significant cost reduction from Block 1 life cycle system costs.

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Characterization of Laser Materials.

Introduction. Great effort was made in 1987 to spectroscopically analyze host materials and build and demonstrate a promethium laser that could match up to the Block 1 receiver technology at 459 nm. The proposal for a promethium-based solid state laser for SLC was based on two premises. First, Pm, the analog of neodymium, would make an excellent laser, operating at around 920 nm. Second, because of the nephelauxetic effect, a suitable host could probably be found at the right wavelength among the many

Fig. 6-87. Level diagrams of Nd^{3+} and Pm^{3+} . The Nd^{3+} $^4\text{F}_{3/2} \rightarrow ^4\text{I}_{11/2}$ transition at $1.06 \mu\text{m}$ is the analog of the Pm^{3+} $^5\text{F}_1 \rightarrow ^5\text{I}_5$ transition at $0.92 \mu\text{m}$.

Fig. 6-88. The nephelauxetic effect for the $\text{Nd}^{3+} \cdot {}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ transition. One data point in $\text{Pm}^{3+} \cdot \text{YVO}_4$ is shown. Crystals and glasses are arranged in the order of increasing covalency of the ligand bond.

fluoride crystals and glasses. During 1987, an experimental program was set up to validate these projections and to identify a suitable host. The most important host characteristics are as follows:

- A dominant spectral gain feature at 911 or 919 nm.
- A stimulated emission cross section of $\sim 1 \times 10^{-19} \text{ cm}^2$.
- A metastable lifetime of greater than a few hundred μs .
- A crystal host scalable in dimensions to approximately $1 \times 4 \times 20 \text{ cm}$.
- Low absorption from color centers formed by nuclear decay by-products.

Judd-Ofelt Theory. The possibility of using Pm^{3+} as the active ion in a four-level, room-temperature laser was first recognized by Krupke⁹⁹ in 1972. He suggested that the Pm ion could be viewed as the analog of the neodymium ion and that its relevant optical properties (transition rates, branching ratios, excited state lifetimes, etc.) could be extrapolated from measurements on neodymium in the same host. Using a Judd-Ofelt^{100,101} analysis with intensity parameters extrapolated from other RE^{3+} ions in YAG, Krupke concluded that the ${}^5\text{F}_1 \rightarrow {}^5\text{I}_5$ transition of $\text{Pm}^{3+}:\text{YAG}$ at $\lambda = 0.92 \mu\text{m}$ is the analog of the ${}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ transition of $\text{Nd}^{3+}:\text{YAG}$ at $\lambda = 1.06 \mu\text{m}$; aside from its radioactivity, Pm should make an excellent laser. Figure 6-87 shows the energy levels of interest and illustrates the similarities between the two ions.

Nephelauxetic Effect. It is known that the electronic configurations and, therefore, the energy levels of a RE^{3+} ion in a crystal or glass are perturbed by the ligand bonds to neighboring ions and that this causes the wavelength of the transitions to vary from host to host. This is called the nephelauxetic effect. Based on the wavelength range observed in neodymium systems, we felt confident that the range for Pm would include 911 and 919 nm.

In particular, the transitions energies are determined by the splittings of the $4f^n$ electronic configuration ($n = 3$ for Nd, $n = 4$ for Pm). These splittings are in turn determined by the electron-electron interaction, which becomes weaker as the ligand bonds become more covalent and the electrons spend more time apart from each other and around the ligand ion. One would therefore expect that as the bonding becomes more covalent and the splitting decreases, the transition wavelengths will shift to the red. This effect is readily observed in the $\text{Nd}^{3+} \cdot {}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ transition, which becomes longer in wavelength as the host changes from a fluoride (less covalent) to a chloride to an oxide (more covalent). Figure 6-88 shows the results of a literature search and an experimental program at LLNL for the $\text{Nd}^{3+} \cdot {}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{11/2}$ transition in a large number of glass and crystalline hosts. Also shown on this curve is the one data point for Pm^{3+} in

YVO₄ that was in the literature prior to our investigation.

Spectroscopic Measurements. The next step was to follow up our projections of emission wavelength with an experimental program:

- To validate our predictions of the spectroscopic parameters.
- To identify a host that would allow us to build an efficient solid state laser at 911 or 919 nm.
- To identify and quantify the loss mechanisms so that we can project overall laser performance.

The only Pm isotope available for doping was ¹⁴⁷Pm that had been extracted from reactor waste products. With a half-life of 2.6 yr, it produces a 225-keV electron that tends to form color centers, affecting the optical properties of the host. We were still able to make the measurements with fewer problems than anticipated. However, for an actual SLC transmitter, we would use ¹⁴⁵Pm, which has a half-life of 17.7 years and decays by electron capture, emitting a 40-keV γ ray. The electronic transitions of ¹⁴⁵Pm are essentially the same as those of ¹⁴⁷Pm, but we expect the lattice damage from the nuclear decay products will be less severe.

Table 6-10. Pm³⁺-doped crystals and glasses investigated.

Crystals	Glasses
SrF ₂	PbInPO ₃
BaF ₂	ZBLA fluoride glass (57ZrF ₄ -36BaF ₂ -3LaF ₃ -4AlF ₃)
LaF ₃	BZILT fluoride glass (30BaF ₂ -20ZnF ₂ -30InF ₃ -10LuF ₃)
YF ₃	BeCaKAl fluoride glass (45BeF ₂ -20CaF ₂ -15KF-20AlF ₃)

The samples were prepared by C. B. Finch, C. A. Culpepper, and L. A. Boatner at Oak Ridge National Laboratory.

Table 6-11. Promethium upper-state lifetimes for the various hosts and the analogous neodymium system lifetimes (see Ref. 102).

Host	Pm(ms)	Nd(ms)
LaCl ₃	0.17	0.20
CaF ₂	1.0	1.1
SrF ₂	0.90	1.2
LaF ₃	0.63	0.70
LiYF ₄	0.50	0.57
YF ₃	0.34	0.0
CaF ₂ -4YF ₃	0.54	0.0
ZBLA	0.44	0.44
BZILT	0.45	0.41
BeCaKAl	0.50	0.0
PbInPO	0.30	0.31

The next list summarizes our work (details of each measurement will follow the summary):

- We measured absorption and emission spectra for many samples. Using these measurements, we identified two fluoride glasses with usable emission features at 919 nm.
- We obtained the Judd-Ofelt parameters in PbInPO₃ glass, from which we calculated the radiative transition rates and the lifetimes. These measurements confirmed our earlier projections.
- We measured lifetimes and line shapes at low concentrations of Pm³⁺; from this we have estimated emission cross sections. By comparing these measured lifetimes with the Judd-Ofelt calculations, we estimated the radiative quantum efficiency.
- We measured 919-nm absorption by the presence of neodymium, the decay product of ¹⁴⁵Pm. Even for end-of-life concentrations ($1.8 \times 10^{20} \text{ cm}^{-3}$), this absorption coefficient is only $3.5 \times 10^{-3} \text{ cm}^{-1}$.
- We quantified 919-nm absorption due to color centers, also formed by the nuclear decay products. We identified hosts in which the color-center absorption is small.
- We looked at quenching in Pm as a function of doping and are still evaluating these results.

Table 6-10 summarizes the various glass and crystalline hosts that we studied. The fluoride crystals were grown using a Bridgman apparatus with rf heating, in vitreous carbon crucibles under an argon atmosphere. The fluoride glasses were also prepared under an argon atmosphere. All the production was done in a glove box to eliminate the dispersal of radioactive material.

We verified our predictions (based on a Judd-Ofelt analysis) that the Pm³⁺ ⁵F₁ lifetime in a given host equals the Nd³⁺ ⁴F_{3/2} lifetime in the same host by measuring the Pm³⁺ ⁵F₁ fluorescence decay signal. Samples were excited with either a flashlamp-pumped laser or a Nd:YAG-pumped pulsed dye laser. Table 6-11 shows the measured Pm lifetimes and the corresponding Nd³⁺ lifetimes. In those cases where the Nd³⁺ lifetimes

were not available in the literature, we measured samples prepared by our own crystal growth laboratory.

Emission and absorption spectra were measured by exciting the samples with light that had passed through a 0.32-m monochromator. After the fluorescence was focused into a McPherson monochromator, a mirror directed the light to the appropriate slit-detector assembly. We used a photomultiplier tube cooled with dry ice and a thermoelectrically cooled Optoelectronics OTC-22S-53TC-PbS cell as detectors. All spectra were corrected for the spectral response of the detection system.

Using measured lifetimes and emission line shapes at low concentrations of Pm^{3+} , we estimated the $^5\text{F}_1 \rightarrow ^5\text{I}_5$ emission cross sections of Pm^{3+} in various host materials using the Fuchtbauer-Lan'enburg relation

$$\sigma = \frac{\lambda^2}{8\pi n^2 \tau} g(\nu), \quad (34)$$

where τ is the lifetime and $g(\nu)$ is the normalized line-shape function. The line-shape function can be expressed as

$$g(\nu) = \frac{I(\nu)}{\int I(\nu) d\nu} = \frac{\lambda^3 I(\lambda)}{\int \lambda^3 I(\lambda) d\lambda}, \quad (35)$$

where $I(\lambda)$ is measured in experimentally relevant units of watts per wavelength interval. Table 6-12 gives the emission

cross sections for the $\text{Pm}^{3+} ^5\text{F}_1 \rightarrow ^5\text{I}_5$ transition as determined from our spectroscopic measurements.

In addition to our wavelength scaling and storage lifetime projections, we obtained detailed Judd-Ofelt parameters for Pm^{3+} from a conveniently fabricated sample of PbInPO_4 glass. From the measured absorption spectrum, we calculated the Judd-Ofelt parameters as previously described. Table 6-13 shows the results of this calculation. The measured and calculated radiative line strengths agree to within an rms error of 5.7%.

Having calculated the Judd-Ofelt intensity parameters (Ω), we calculated the radiative transition rates between the various levels of the ^5I and ^5F manifolds. These calculated rates give radiative lifetimes of $^5\text{F}_1$ and $^5\text{F}_2$ equal to 353 μs and 346 μs , respectively. These two manifolds are separated by $\sim 490 \text{ cm}^{-1}$ and will be in

Table 6-12. Stimulated emission cross sections of the $\text{Pm}^{3+} ^5\text{F}_1 \rightarrow ^5\text{I}_5$ transition as determined by our spectroscopic measurements (from Ref. 102).

Host	Average λ (nm)	Peak λ (nm)	n	$\left[\frac{\lambda_{\text{peak}}^3 I_p(\lambda)}{\lambda^3 I(\lambda) d\nu} \right]^{-1}$	σ_{peak} (10^{-20} cm^2)
LaCl_3	931.5	931.9	1.4 ^a	21.48	64.4
CaF_2	925.1	927.9	1.43	45.67	4.9
SrF_2	923.4	925.3	1.44	58.31	4.2
LaF_3	922.9	925.5	1.45 ^a	78.37	4.4
LiYF_4	926.4	931.1	1.46	52.85	8.2
YF_3	927.4	923.0	1.45 ^a	45.59	13.9
$\text{CaF}_2\text{-4YF}_3$	925.5	928.5	1.45 ^a	165.49	2.4
ZBLA	925.9	925.5	1.51	198.27	2.3
BZILT	926.8	927.5	1.5 ^a	204.93	2.2
BeCaKAl	925.4	925.5	1.5	203.09	2.0
PbInPO_4	931.5	932.5	1.76	223.14	2.3

^a Estimated.

Table 6-13. Detailed results of the Judd-Ofelt analysis of the absorption data. Measured and calculated line strengths agree within an rms error of 5.7%.

Band No.	λ_p (nm)	$\int \text{OD}(\lambda) d\lambda$ (Å)	$[\text{U}^2]^2$ ($3.79 \times 10^{-20} \text{ cm}^2$)	$[\text{U}^4]^2$ ($2.38 \times 10^{-20} \text{ cm}^2$)	$[\text{U}^6]^2$ ($2.56 \times 10^{-20} \text{ cm}^2$)	S_{meas} (10^{-20} cm^2)	S_{calc}^a (10^{-20} cm^2)
1	790	35.7	0.0018	0.3438	0.1272	1.14	1.14
2	740	39.3	0.0001	0.1068	0.4237	1.34	1.34
3	695	36.9	0.0004	0.0297	0.4697	1.34	1.27
4	630	4.76	0.0000	0.0020	0.0342	0.19	0.09
5	560	112	0.8990	0.5812	0.1190	5.1	5.1
6	495	14.3	0.0072	0.0910	0.0927	0.73	0.5
7	455	6.0	0.0147	0.0451	0.0390	0.33	0.26

^a Pm ion density = $8.0 \times 10^{19} \text{ ions/cm}^3$; $n = 1.78$

$\Omega_2 = 3.79 \times 10^{-20} \text{ cm}^2$

$\Omega_4 = 2.38 \times 10^{-20} \text{ cm}^2$

$\Omega_6 = 2.56 \times 10^{-20} \text{ cm}^2$

thermal equilibrium with an 86/14 split of the excited populations at room temperature. The calculated radiative lifetime of these coupled manifolds is 352 μ s (compared with a measured fluorescence lifetime of 285 μ s) indicating a quantum yield of 80%. We anticipated a higher quantum yield, based on the magnitude of the energy gap and the multiphonon rate present in the phosphate glasses. This discrepancy is due to OH^- impurities. It occurs for phosphate glasses from the same source, too. It is reasonable, based on the relatively large change between the first and third e-folding times, to believe that this mechanism is also responsible for this result. We used the measured line shapes and calculated lifetime and branching ratios for the 933- and 1098-nm transitions; the calculated emission cross-sections are 2.1 and $2.9 \times 10^{-20} \text{ cm}^2$, respectively.

Because Pm is radioactive, decay products can introduce losses. Absorption due to the color centers is caused by the ejected particle as it passes through the lattice. In the case of ^{147}Pm , a 225-keV electron initiates this process; for ^{145}Pm , a 40-keV photon is responsible. The other particle to consider is the daughter

ion left behind. The ^{145}Pm decays to ^{145}Nd , which could act as a source of both 919-nm absorption and quenching.

Even though we expected more radiation damage from the shorter lived ^{147}Pm , we still felt we could find a host in which the color-center absorption peak was shifted into the UV and would not absorb at 919 nm. Several promising materials (one is BZLT) that exhibit both the required wavelength emission and insensitivity to radiation-induced coloring at 919 nm have been identified among the fluoride glasses. Published work¹⁰³ on BZYT leads us to expect that BZLT will exhibit insignificant radiation-coloring-induced absorption at 919 nm. Furthermore, we see evidence that suggests that even in highly colored samples the color centers quickly bleach out under lasing conditions. In our PbInPO_3 glass, we observed strong absorption at room temperature, which could be effectively reduced through a moderate temperature rise; this promoted annealing of the radiation-induced coloring. However, when the PbInPO_4 sample was pumped and began to lase, the color centers quickly disappeared and heating was not necessary. Our conclusion, based on these observations, is that absorption due to color centers will not be a significant loss and can be managed in a number of ways.

Losses due to the presence of Nd could become important, especially near the end of life (7-yr design lifetime) when approximately 24% of the ^{145}Pm will have decayed to ^{145}Nd . There are several ways this might interfere with laser operation. First, the $\text{Nd}^{3+} {}^4\text{F}_{3/2} \rightarrow {}^4\text{I}_{9/2}$ transition, which peaks at $\sim 880 \text{ nm}$, might have some measurable magnitude at 919 nm. Figure 6-89 shows an overlay of two spectra. We measured absorption in 0.5-mol% Nd-doped BeF_2 glass. Because the absorption from these thick BeF_2 glass samples was expected to be too small to measure using ordinary spectrophotometers, we used a cavity ring-down technique. This technique relies on measuring the decay of radiation trapped in a very high-finesse optical cavity. By measuring two such decays (one with the cavity empty and one with a sample present in the cavity), it is

Fig. 6-89. Pm^{3+} and Nd^{3+} absorption. Possible absorption from the tail of Nd^{3+} ground state to the ${}^4\text{F}_{3/2}$ excited state is a potential loss mechanism in a Pm-based SLC transmitter.

possible to quantify very small sample absorption and scattering losses. The results of our Nd-absorption measurements indicate absorption losses of about $<0.001 \text{ cm}^{-1}$, giving an effective room temperature Nd³⁺-absorption cross section of $2.0 \times 10^{-23} \text{ cm}^2$ at 919 nm in the BeF₂ glass samples studied. For end-of-life concentrations of $1.7 \times 10^{20} \text{ cm}^{-3}$, the loss is 0.0036 cm^{-1} . For comparison, the typical gain will be about 0.065 cm^{-1} . For our point design, this end-of-life loss translates into a slab transmissivity of ~ 0.90 .

Another issue to be resolved is the magnitude of Pm ion-ion quenching; this scales with the doping, which may be high in the glass host. Our initial estimates indicated that when compared to Nd, Pm³⁺ would be resistant to ion-ion quenching because Pm³⁺ does not have the same accidental resonances that are present in Nd³⁺.

To verify that self-quenching is small, we measured ⁵F₁ fluorescence lifetimes in ZBLA and BeCaKAl glasses doped with 0.5-, 1-, 2-, and 5-wt% Pm. These results, shown in Table 6-14, indicate that the more heavily doped samples have shorter lifetimes.

We believe impurities cause this unexpected Pm concentration quenching. As evidence that these impurities are present, we measured absorption on a BeCaKAl sample after irradiation by an electron beam. The salient feature of this data for the present analysis is the way the wings of the absorption band saturated much quicker than did the central peak with absorbed radiation dose. This damage behavior is characteristic of impurity centers, which can act as trapping sites by undergoing a change in valence.

Although this investigation is incomplete, our initial results indicate that the concentration effects we have observed are probably caused by impurities in our host materials and not due to Pm ion-ion quenching. Experiments are being planned to verify that this is the case. At Oak Ridge National Laboratory (ORNL), a glove box for sample preparation is being readied for a fluoride atmosphere (either HF or NF₃) in an effort to further reduce sample contaminations, particularly those of incorporated OH⁻, a known quencher of rare earth ion excited

Table 6-14. Measured fluorescence lifetimes of the ⁵F₁ level in ZBLA and in BZILT as a function of concentration. τ_1 , τ_2 , and τ_3 indicate the first three e-folding times. τ_{ave} is the average of these three measurements.

wt% PmOF	τ_{ave}	ZBLA fluorescence lifetimes (μs)		
		τ_1	τ_2	τ_3
0.5	441	378	453	441
2.5	338	219	330	402
5.0	168	88	159	215
wt% PmOF	τ_{ave}	BeCaKAl fluorescence lifetimes (μs)		
		τ_1	τ_2	τ_3
0.5	499	279	505	543
1.0	438	262	433	527
3.0	249	122	232	322
5.0	188	88	172	246

states. A concentration series is also being prepared in BZILT glass to see if the same ⁵F₁ lifetime vs Pm³⁺ concentration is observed.

In summary, we validated projections of the spectroscopic parameters based on Judd-Ofelt analysis. We identified two glass hosts, BZILT and BeCaKAl, which have usable emission features at 919 nm and are resistant to radiation damage. We measured 919-nm absorption due to Nd and found that it is $<0.0036 \text{ cm}^{-1}$ at end of life.

Promethium Laser Demonstration.

Our first Pm laser was demonstrated in PbInPO₃ glass.¹⁰² This host was chosen mainly because it could be fabricated in small sizes with high optical quality (the fabrication done at ORNL). The goal of this exercise was primarily to demonstrate that we could make Pm lase. We also wanted to confirm our Judd-Ofelt-predicted transition rates and make a Findlay-Clay¹⁰⁴ measurement to get closure with the spectroscopic measurements.

Figure 6-90 shows the emission spectrum of Pm:PbInPO₃. The emissions at 820, 933, 1098 nm are due to the ⁵F₁ — ⁵I_{4,5,6} transitions; the weaker features at 790, 890, and 1042 nm are due to the ⁵F₂ — ⁵I_{4,5,6} transitions. We used the measured line shapes and the calculated lifetime and branching ratios for the 933- and 1098-nm transitions; the calculated emission cross sections are 2.1 and $2.9 \times 10^{-20} \text{ cm}^2$, respectively. We also evaluated the effect of possible excited state absorption (ESA) of the 933-nm laser transition and determined that ESA in this wavelength range was negligible.

The sample in this demonstration was cylindrical, with a diameter of 1.5 cm and

Fig. 6-90. Emission spectrum of Pm^{3+} in PbInPO_4 glass.

Table 6-15. Cavity parameters at threshold for laser action of $\text{Pm}:\text{PbInPO}_4$ and $\text{Nd}:\text{LHG-8}$ glasses. Mirror with reflectivity $R \geq 99.5\%$ was used for the high reflector.

Material	λ_{laser}	R_{out} (%)	$E_{\text{thr(pump)}}$ (mJ)
$\text{Pm}:\text{PbInPO}_4$	933	96	92
$\text{Pm}:\text{PbInPO}_4$	1098	97	44
$\text{Nd}:\text{LHG-8}$	1053	99.5	12

a length of 0.6 cm. The ends were polished flat to about $\lambda/10$ and parallel to within 45 arc seconds. To facilitate polishing, the sample was glued in the center of a cylindrical piece of Nd-doped laser glass (LHG-8, manufactured by Hoya Optics, Incorporated). This glass received the same optical quality finish as the Pm laser sample. The laser etalon was placed in the center of a nearly concentric cavity 55 cm long; the cavity was formed by two spherical mirrors with 30-cm radii of curvature. The mirrors were coated to transmit 80% of the pump radiation and were highly reflecting at 933 and 1098 nm. The laser cavity was pumped along the cavity axis with the Candela LFDL-6 dye laser. The laser could be tuned to the Pm^{3+} or the Nd^{3+} absorption maxima at 571.5 and 575 nm, respectively. The pump laser beam diameter at the etalon center was 1.6 mm, as determined by pinhole transmission measurements, for an output energy of 150 mJ. The pump waist diameter varied linearly as the pump energy was

decreased. The calculated beam-waist radius of the cavity was 0.32 mm. These parameters were chosen to facilitate production of laser action in the presence of losses caused by surface and bulk defects. The output radiation was detected with a silicon photodiode after passing through appropriate neutral density filters and a long pass filter that removed residual pump radiation. Output wavelengths were determined to the nearest nanometer using a calibrated optical multichannel analyzer.

We anticipated the need to heat the laser etalon over 100°C, in order to reduce the color-center absorption. This proved to be unnecessary. Room temperature laser action was achieved at both 953 and 1098 nm after the laser disk was exposed to several tens of laser pulses. Switching between the two laser transitions was accomplished through the choice of cavity mirrors. Table 6-15 shows the mirror reflectivities and laser action threshold energies for both the Pm^{3+} transitions and the Nd^{3+} laser glass.

Visual inspection revealed that the color-center absorption was greatly reduced in the pumped region after such irradiation. An He-Ne laser beam was passed coaxially with the pump beam to measure the transmission at 632.8 nm as a function of pump energy. Figure 6-91 shows the transmission at several pump energies as a function of time. (Note that even the probe beam has sufficient energy to bleach the color centers.) Comparison with Table 6-15 shows that laser action for the 1098-nm transition commences before the pump laser totally bleaches the color-center absorption. Laser action at 933 nm does not occur until after the color-center absorption is completely bleached. It was determined, using the data presented in Fig. 6-92, that the absorption coefficients at 571.5, 632.8, and 933 nm all anneal proportionately as the temperature is increased. We postulate that the bleaching process should show the same behavior. Comparison of the threshold energies given in Table 6-15 with the data shown in Fig. 6-91 allows us to estimate that the host transmits about 70% of the incident pump light at threshold for 1098-nm laser

action. For 933-nm laser action, the energy is sufficiently high for total bleaching of the color-center absorption. With this information, we may now predict the gain at threshold for both Pm^{3+} laser transitions to derive the value of the losses for those transitions using the equation

$$R_1 R_2 \exp(-\sigma N^* - \delta l) = 1, \quad (36)$$

where R_1 and R_2 are the mirror reflectivities, σ is the emission cross section, N^* is the excited state number density, δ is the loss per unit length, and l is the length.

At the wavelengths of both laser transitions, there is no contribution to the emission cross section from the $^3\text{F}_2$ level.¹⁰² The values obtained from this analysis yield $\delta = 0.25 \text{ cm}^{-1}$ for the 933-nm transition and $\delta = 0.15 \text{ cm}^{-1}$ for the 1098-nm transition. These values vary by at least 10% because of slight cavity mirror misalignments and the difficulties experienced in assuring that the same region was used for both experiments. Performing the same analysis on the $\text{Nd}^{3+}:\text{LHG-8}$ glass yields a value of $\delta = 0.18 \text{ cm}^{-1}$. Thus, all values of the loss are about the same, even though the LHG-8 is very low in bulk loss. This leads us to the

conclusion that poor optical finish is the reason for the high cavity loss values. The near equivalence of the loss values is also indicative that our emission cross sections are consistent, within 30%, to the observed thresholds.

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Fig. 6-91. Transmission at several pump energies as a function of the number of pulses.

Fig. 6-92. Effects of radiation coloring in the PbInPO_4 glass sample. Sharp features due to Pm^{3+} are superimposed on broad background absorption due to radiation-induced coloring.

Fig. 6-93. Schematic diagram of the laser cavity used in our Q-switched oscillator experiments.

Fig. 6-94. (a) Laser output with no etalon; (b) spectral width with the etalon in place showing single mode operation.

Oscillator Demonstration. In anticipation of constructing a Pm-doped glass oscillator capable of running single line and giving a Q-switched output pulse of 5 to 10 ns, we experimented with Nd:LHG-5 glass as a surrogate. By using this Nd sample rather than the Pm sample as the gain material, we can address the relevant design issues without the inconveniences associated with handling the radioactive promethium.

Figure 6-93 shows a schematic diagram of our oscillator design. Initial experiments used a krypton laser running on the 752-nm line as the pump source. We wanted to keep the cavity as short as

possible to make it easier to run single line, but we were limited by the physical size of the Q switch and the intracavity etalon. The shortest cavity we operated was 4 cm. For Q switching, we used a traveling-wave PbMoO₄ acousto-optic modulator running at 110 MHz, which was chosen for its small size. The response time, based on the calculated laser beam waist of 120 μm and the speed of sound in PbMoO₄ of $\sim 4 \times 10^5$ cm/s, was 60 ns. In order to meet the design requirements (5 to 10 ns), we will need a much shorter response time; however, apart from pulse width, this system did demonstrate the design requirements.

We wanted to demonstrate two important figures of merit: first, the ability to operate on a single frequency with narrow line width, and, second, the ability to tune off the gain peak to a precise frequency. This was accomplished with a single intracavity etalon. Figure 6-94(a) shows the spectral range of the output without the etalon; Fig. 6-94(b) shows the line narrowing we achieved with the etalon in place. We used a 100-μm-thick etalon with a free spectral range of ~ 1000 GHz. The faces were uncoated ($R = 0.04$). Using a scanning Fabry-Perot to measure the bandwidth, we observed a line width of less than 1 GHz, which was limited by our instrument resolution. This indicates that the oscillator is indeed running in a single longitudinal

mode. Additionally, by tilting the etalon, we were able to tune the frequency of the oscillator over a range of 500 GHz.

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Diode Pumps.

Introduction. Efficient, long-lived, and reasonably priced diode arrays emitting at 770 nm are crucial to the success of a solid state promethium SLC system. During the last several years, there have been great improvements in the output power, efficiency, and lifetime of semiconductor lasers. However, most of this effort focused on the 808-nm material commonly used to pump neodymium. Because of the shorter wavelength requirements of promethium, changes and modifications to the architecture had to be made. We had to demonstrate the following:

- Diode pumps could be made at 770 nm.
- They would be reliable and efficient enough for a promethium SLC system.
- They could be fabricated at a reasonable cost.

Accordingly, LLNL contracted with three companies to consider these issues. Contracts of approximately \$500,000 each with McDonnell Douglas Aeronautics Company (MDAC) and Radio Corporation of America/General Electric (RCA/GE) covered the period from April 1987 to April 1988; a contract with Spectra Diode Laboratories (SDL) for approximately \$166,000 began September 1987. The focus of these contracts was to optimize 770-nm material architecture for performance, lifetime, and cost.

In addition to the above requirements, the issue of diode exposure to x rays from the radioactive ^{145}Pm also needed to be addressed. The rapid failure of GaAs diodes when exposed to 1-MeV γ rays had been documented, but no data on the effects of the x-radiation (mostly 40 keV with some 70 keV) expected from ^{145}Pm existed.

Wavelength Issues. The wavelength of an AlGaAs semiconductor diode is determined to first order by the doping level of aluminum in the active layer. As the doping level increases, so does the band

gap. Unfortunately, at above approximately 0.2 mole fraction of aluminum, the material becomes indirect band gap rather than direct band gap. In principle, even before a doping of 20 mol% is reached, an increasing fraction of the transitions become indirect band gap; as a result, threshold current rises, device efficiency drops, and device lifetime falls.^{105,106} The 770-nm arrays, which require greater aluminum doping than the 808-nm arrays, are therefore expected to have reduced lifetimes when compared to the longer-wavelength material.

However, if we are using quantum well material (thin active layers of 500 Å or less), some wavelength tuning can be achieved by varying the quantum well thickness.¹⁰⁷ All the contractors use this technique to minimize aluminum-doping requirements and to fabricate long-lived, efficient devices that emit at 770 nm.

Demonstrated Performance. The three contractors have reported 770-nm diodes that are efficient and have high power. RCA/GE has already fabricated 0.5-cm² arrays. McDonnell Douglas was directed to work on the optimization of 770-nm bars, while SDL had reported diodes operational at 770 nm, even before the LLNL contractual agreement took effect.

Fig. 6-95. McDonnell Douglas study of wavelengths vs cavity length and coatings.

Spectra Diode Laboratories recently demonstrated 770-nm arrays¹⁰⁸ and reported that 100- μm aperture, 10-stripe arrays with nonabsorbing mirrors were fabricated by Si-induced disordering. These devices operated at up to 2.4 W cw without catastrophic damage. Peak powers of up to 3.5 W were obtained with 100- μs pulses before onset of damage. Although device efficiency was not reported, greater than 40% efficiency in arrays has been reported by SDL at somewhat longer wavelengths.

The staff at MDAC focused on the optimization of 770-nm bar architecture. In particular, they looked at doping level, bar length, quantum well thickness, and coatings. Before the LLNL contract was let, MDAC had reported 787-nm bars with greater than 40% wall-plug efficiency. During the contract, 770-nm material was grown for use in a coating and cleave-length matrix study for the optimization of these two parameters. A total of 250 diodes were fabricated with cavity lengths of 200, 300, 400, and 500 μm and HR/passivation- and HR/AR-coating combinations. Figure 6-95 shows the measured wavelengths of each of these devices vs cavity length for four coating types. A marked blue shift with short cavities was observed for HR/AR-coated diodes, in agreement with expectations.

We are measuring threshold current, slope efficiency, and catastrophic failure for these devices.

As of this writing, over 150 bars have been delivered by Electronics Laboratory in Syracuse to General Electric Astro-Space Laboratories. Wavelength nonuniformities in individual wafers and a factor of 2 variation in threshold current of shipped bars have been noted. Experiments are being conducted to determine whether bar length or coating reflectivity is the source of these variations. Three 2-mm-long bars have been life-tested at 5 A with 10- μs pulses (no difference should be observed at 100 μs) at 100 Hz. After 220 hr (79 million shots) at this drive level, no measurable degradation in output current has been noted.

By October 1987, RCA/GE had fabricated 15 arrays. Three of these arrays were built at the David Sarnoff Laboratories (now SRI International) and the remainder at GE Astro-Space Laboratories. Each array was within ± 5 nm of 770 nm and had a typical FWHM of 5 nm. The use of forming gas during the soldering of the back heat-sink plane to the individual bars and Thermkon spacers was a key ingredient to success. For the five arrays fabricated with forming gas, the average overall efficiency was 27% and the average power density was 1.68 kW/cm².

Fig. 6-96. Output spectra of the RCA/GE diode arrays fabricated in forming gas.

Figure 6-96 shows the spectra from each of these five arrays. Figure 6-97 is a photograph of the 35%-efficient 0.5 cm² array shown in Tables 6-16 and 6-17. Some of the subarrays (those at 0.5- to 1-kW/cm² power density) have entered life testing. At somewhat less than 10⁹ shots, these arrays had experienced a 50% degradation in power, an unacceptable value for SLC. These less efficient arrays would be expected to be shorter lived, however. Indeed, the more efficient arrays now being tested have exhibited no degradation in efficiency after 10⁸ shots.¹⁰⁹

The results on 770-nm fabrication have been very encouraging. Within the first six months of their contracts, RCA/GE and MDAC reported efficient short-wavelength material. As the optimization of quantum well thickness, doping, cavity length and coating, heat sinking, index confinement, and packing density per bar is completed, the efficiencies will continue to improve. Importantly, MDAC has recently reported¹¹⁰ longer wavelength material with slope efficiencies near 84% and overall conversion efficiencies of 57% when attention was thoroughly given to series resistance and other generic diode-efficiency degrading mechanisms. It is our position that 40% end-of-life

diode array efficiencies at 770 nm are certainly attainable and that higher values may also prove attainable. Ultimately, yield and cost will be traded off against efficiency to determine overall cost effectiveness.

Radiation Damage to Diodes. As already mentioned, an additional consideration regarding the use of diode arrays in a promethium-SLC application are the potential effects of promethium radiation

Fig. 6-97. Diode array fabricated at RCA/GE, operating at 770 nm at 35% efficiency.

Table 6-16. Diode array performance summary for the RCA/GE arrays.

Number of bars	Diode array performance summary					Power density (W/cm ²)	Comments
	Peak power W (A)	Slope efficiency (%)	Overall efficiency (%)	Wavelength (nm)			
SRI-1	6	58 (20)	56	26	771 ± 1	1500	5/15/87, 2 bars failed by 8/3/87, forming gas
A-10	6	20 (25)	16	8	774 ± 2.5	512	6/9/87, flux in air, 63LSR-215
A-11	6	14 (20)		7		358	Flux in air, 63LSR-215
Single bar SRI	1	4 (25)	14	6		370	2 thermkon bars, 15°, cleaned, 63LSR-215
Single bar SRI	1	17 (25)	60	29		1600	2 thermkon bars, not cleaned, 62LSR-215
SRI-2	6	60 (25)	40	20		1600	7/7/87, epoxy 1 BeO forming gas 63LSR-215
A-13	5	10 (25)				270	7/15/87, flux in air, 63LSR-275-T
A-14	6	22 (20)			771 ± 2	550	63LSR-275-T
A-15	6	29 (16)	31	15	775 ± 1.5	743	Flux in air, died 63LSR-275-T
A-16	6	20 (20)	18	9		51	14°, nitrogen atmosphere, 63LSR-275-T
SR-3	5	42 (20)	33	19		1100	14°, nitrogen atmosphere, 63LSR-275-T
A-17	10	174 (25)	60	35	769 ± 2.5	2000	8/24/87, 16°, forming gas, 4.75-mm bars, 63LSR-275-T
A-18	10	50 (20)	24	10	771 ± 2.5	714	Flux in air, 3.76-mm bars, died at 20 A, 16° 63LSR-275-T
SRI-4	10	67 (25)	25	7	773 ± 2.5	758	4.75-mm bars, reflowed 2 or 3 times, 63LSR-275-T
A-19	10	150 (25)	62	29	769 ± 2.5	1700	4.75-mm bars, forming gas, 16° 63LSR-275-T
A-20	6	75 (25)	55	27	769 ± 3.5	1700	4.75-mm bars, forming gas, 16° 63LSR-275-T
A-21	5	12 (30)	6	3	772 ± 2	273	High-threshold material, 4.75 mm bars, twice flowed 74LSR-215-T

on the diode lifetimes. Earlier work¹¹¹ on the effects of 1-MeV γ rays on GaAs-diode lifetimes indicated that the emission intensity of the GaAs arrays dropped by more than half at an exposure of 50 Mrad. Figure 6-98 reproduces Barnes' results on GaAs diode emission intensity vs exposure. In order to assess the extent of damage to be expected from the x rays given off in the decay of ¹⁴⁵Pm, we set up a series of measurements to quantify device performance at varying doses and as a function of x-ray wavelength.

We anticipated that the ¹⁴⁵Pm x rays (mostly 40 keV with a small component of 70 keV) would not be significant to the performance of the diode arrays. If the degradation results from the high energy radiation actually ejecting a metal atom from its site in the material, the lowest photon energy that could cause the recoil of the lightest material (aluminum) from its site in the lattice would be about

300 keV. This damage mechanism is generally referred to as the knock-on model for material defect formation.

We exposed diode arrays to narrow-band calibrated x rays from a high-energy electron tube. As the diodes were exposed, we constantly measured the light vs current characteristics, paying particular attention to diode threshold and slope efficiency. At 40 keV, we exposed the diodes to approximately 150 Mrad and observed no measurable effect (less than 3%) on any of the parameters. While this is only about 5% of the mission dose expected for diodes exposed to a heavily (~7%) doped glass promethium slab, at this γ -ray exposure, the diode would no longer be expected to function.

To further understand the radiation-induced lifetime effects, we exposed the diodes to higher energy x rays (70, 90, and 120 keV) in order to determine the threshold for damage. We observed degradation in the performance of the diodes at near 100 keV. This is in rough agreement with our expectations from very simple knock-on model calculation. The difference between model and experiment for this threshold value can be explained by recent studies, which have attributed the 100-keV damage to facet-coating damage on the devices.

While we have not yet exposed arrays to a full mission dose of x rays, we have proven that the effects of x rays are far more benign than those of γ rays. Also,

Table 6-17. Diode array performance summary for the diode arrays fabricated in forming gas.

Array number	Number of bars	Forming gas array performance summary (= 770 nm)			
		Peak power	Efficiency		Power density
		W (A)	Slope	Overall	W/cm ² (peak)
SRI-2	5	60 (25)	40	20	1500
A-17	10	174 (25)	60	35	2000
A-19	10	150 (25)	52	29	1700
A-20	5	75 (25)	55	27	1700
A-22	10	154 (30)	50	26	1750
A-24	10	119.8 (30)	36	21	1446

- SDL reports ~3.3 kW/cm² peak, catastrophic optical damage (808 nm)
- MDAC reports ~3.6 kW/cm² peak, catastrophic optical damage (808 nm)
- 56 W/cm² average, 80% duty cycle

Fig. 6-98. The γ -ray vs x-ray effects on AlGaAs emission.

the observation of a threshold is encouraging evidence that the simple knock-on model may describe of the life-limiting mechanisms. At this point, it appears promising that the ^{145}Pm x rays will have no effect on the performance of the pump diode arrays and that the insertion of x-ray filters (such as high-Z-doped glass) between the diodes and the slabs will not be necessary.

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Promethium Production.

Introduction. The isotope of choice for the Block 2 solid state transmitter is ^{145}Pm . Because of its 17.7-yr half-life, ^{145}Pm does not exist as a fission decay product in either commercial or civilian reactors. Therefore, before a Pm-based Block 2 transmitter can be judged a superior performer, we must demonstrate that the cost of producing ^{145}Pm does not significantly affect the overall costs for the laser transmitter system. After analyzing the options and estimating the costs, we selected the production cycle summarized in Fig. 6-99.

The production cycle is essentially a two-step sequence. The first involves a one-time enrichment of some tens of kilograms of samarium from a natural abundance of 3% ^{144}Sm to 20 to 50%

^{144}Sm . The second step is a series of isolated burns of enriched samarium fuel rods in a production reactor, such as the reactors at Savannah River. The one-time enrichment is needed to reduce the assay of ^{149}Sm in the feed or fuel rod and to enrich the rod in ^{144}Sm . Since the ^{149}Sm isotope has a very high neutron absorption cross section, it would reduce the special nuclear material production capabilities of these reactors excessively; at the same time, the neutron depletion caused by the ^{149}Sm degrades the rate of production of ^{145}Sm . After an enriched rod is burned, typically for a period of one to one and a half years, the rod is stored for a year so that the ^{145}Sm can decay to ^{145}Pm . A chemical separation then strips the ^{145}Pm from the unused or unburnt enriched samarium fuel rod. The unburnt samarium is recycled and recast as fuel material for subsequent burn, decay, and chemical-milking cycles. This process typically converts approximately 0.1% of the ^{144}Sm to ^{145}Sm per burn; depending upon the final host material (i.e., on the doping required to obtain optimum performance), between 2 and 20 g of ^{145}Pm are required per transmitter.

Isotope Separation. Two technologies have been identified for the first enrichment stage of the production process. The first, the plasma separation process (PSP), was developed by TRW; the second,

Fig. 6-99. Promethium production scheme.

the atomic vapor laser isotope separation (AVLIS) process, was developed at LLNL. We will discuss only the PSP for estimating the costs for enrichment in this article.

PSP uses the principle of ion cyclotron resonance to distinguish between isotopes of different masses. In a uniform magnetic field, ions can be selectively heated by controlling the frequency of the applied electric field. Thus a source of samarium ions at natural isotopic abundance can be pumped at the resonant frequency of the ^{144}Sm by an external drive coil. The orbit diameter of the ^{144}Sm ion will increase as it absorbs energy, while the other isotopes, which are not at resonance, will remain unheated. Eventually the ion flow stream reaches a "venetian blind" product plate that preferentially intercepts a larger fraction of the large gyroradius isotopes from the smaller radius (unexcited) isotopes.

Fortunately, the PSP hardware had already produced enriched U and enriched Ni at a scale relevant to the samarium mission. The sum of these tests was a considerable body of experience at running full-scale hardware on missions that were similar to that of ^{144}Sm enrichment. For example, the Ni enrichments involved taking ^{63}Ni from a few percent to 40% enrichment in a single stage at several hundred grams. This experience and the scaling relationships for enrichment developed on these tests enabled the TRW staff to predict with some confidence the operations and costs associated with the enrichment of ^{144}Sm . TRW readily proposed a program of enrichment tests and costs. The TRW staff furnished LLNL with process projections and end-to-end costs for a program that would culminate with the delivery of several tens of kilograms of enriched Sm.

In order to model PSP enrichment performance, we used codes previously calibrated against Ni-enrichment performance. These codes helped to predict important performance parameters,¹¹² such as source heat loading, samarium double ion production fraction, collector nonselective pickup, source rates, and charge exchange. By using the throughput,

assay, hardware availability, operations costs, and chemical flow sheets calculated for the process, we projected costs to be \$598/g for 40% ^{144}Sm and \$1657/g for 60% ^{144}Sm . A total of 80 kg of enriched samarium could be produced at from 60 to 167 g per week, depending upon assay.

Improvement of the process via increased throughput, increased availability, and the minimization of sputter damage to the collector plates should improve process performance by a factor of 3. Even without these improvements, TRW projected production costs of enriched samarium and initial research and development costs required to establish process chemistry and process optimization. TRW proposed a \$5.6 million, one year start-to-production research and development program and a subsequent three-and-a-half year plan that could deliver 81 kg of enriched Sm. An additional \$6 million over and above research and development costs would be required for delivery of 10 kg of material enriched to 40%.

Samarium Burn Considerations. Before modeling the reactor burn portion of the ^{145}Pm production cycle, we had to measure the relevant neutron absorption cross sections. Since the neutron absorption cross sections for the majority isotopes are well known, the drain on reactor neutrons by the introduction of a samarium rod can be reasonably well predicted. In order to properly assess the production rate of ^{145}Pm during this burn, however, measurements are necessary on the following:

- The ^{144}Sm two-group (thermal-neutron plus resonance-integral values) neutron absorption cross sections.

- The ^{145}Sm two-group cross sections, to assess its destruction rate after it is formed.

- The ^{145}Pm two-group cross sections, because significant formation of ^{146}Pm occurs during the burn.

It is necessary to know the amount of the very hot radioisotope ^{146}Pm present in the recently burned rod. If present in large enough quantity, all Pm present when the rod is first removed from the

reactor must be discarded before additional Pm is allowed to grow without the undesirable presence of ^{146}Pm .

Reference 113 and Table 6-18 report the two-group cross sections for ^{144}Sm and ^{145}Sm measured at ORNL. The relevant cross section for ^{145}Pm absorption is being measured at this same facility. Until measurements are complete, we will assume that a chemical extraction and discarding of all Pm present in the rod at the termination of burn is required.

Given these cross sections and the ability to enrich samarium isotopes, we can predict reactor neutron consumption and ^{145}Pm production. We begin by defining a depletion factor F , which corresponds roughly inversely to the depletion factor for the other isotopes during the enrichment step, namely $F = 1$ for natural Sm, $F = 10$ for 24% ^{144}Sm , and $F = 100$ for 76% ^{144}Sm .

$$F = \left[\frac{\frac{^i\chi_l}{1 - ^i\chi_l}}{\frac{^i\chi_p}{1 - ^i\chi_p}} \right] = \beta_2^{-1}, \quad (37)$$

where β_2 is the depletion factor in the enrichment process, and $^i\chi$ is the assay of samarium isotopes.

It is generally assumed that one mole of neutrons costs \$100,000. In order to reduce overall neutron absorption of the samarium fuel rods in a reactor, a reduction in assay of the strongly neutron-absorbing ^{149}Sm is desirable. An additional penalty that is paid in samples with high assays of ^{149}Sm is the longer induction time before production of ^{145}Sm . This arises from the need for the reactor to burn through the ^{149}Sm isotopes before the neutrons can be absorbed by the ^{144}Sm present in the core of the sample. Since a 10-kg fuel rod contains about 66 moles of Sm, roughly on the order of \$6.6 million worth of neutrons are required to burn to "completion," ignoring subsequent weaker absorption by the daughter products. During such a burn, approximately 0.25%

Table 6-18. Thermal neutron absorption cross sections for isotopes of samarium and assays of feed for values of $F = 1, 10$, and 100 .

	Assays ^a (%)			Thermal cross section (b)
	$F = 1$ (natural)	$F = 10$	$F = 100$	
^{144}Sm	3.1	24	76	1.64
^{147}Sm	15.1	11.9	3.8	75
^{148}Sm	11.3	8.9	2.8	2.4
^{149}Sm	13.9	10.9	3.4	41000
^{150}Sm	7.4	5.8	1.8	102
^{152}Sm	26.6	20.8	6.6	210
^{154}Sm	22.6	17.7	5.6	5.5

^a Assays for an AVLIS-type process in which the nonselective pickup portion of the product stream is largely isotope independent. For PSP, there will be a small bias toward increasing ^{147}Sm assays to first order; this bias is unimportant in this process.

of the ^{144}Sm is converted to ^{145}Sm . Thus, as a rough estimate, a natural rod or rods containing 10 kg of samarium will produce somewhat less than 1 g of ^{145}Sm , whereas at 24% enrichment about 6 g are produced and at 76% enrichment about 20 g are produced. Subsequent burns will be operating on a fuel rod that is largely ^{144}Sm , since minimal amounts of ^{144}Sm are depleted on the first burn, while the less useful isotopes are eliminated from the targets. As a result, the subsequent burns, while they will be nearly as time consuming, will represent a far smaller neutron load on the reactors.

Assuming that the isotope-enrichment process provides the reactor with 10 kg of target material at 24% enrichment, 2400 g of ^{144}Sm are available for conversion. This first burn, costing on the order of \$6 million, will produce approximately 6 g of ^{145}Sm . The production of ^{145}Pm at costs near \$2 million/g is feasible for the following reasons:

- The one-time enrichment and research and development costs amounted to \$11.6 million.

- The one-time enrichment costs can be amortized over dozens of burns of the target rod.

- Each burn will cost on the order of \$6 million (with subsequent burns being less costly than the initial burn per gram of ^{145}Sm produced).

While the data presented here do not present a complete analysis of the cost of production, they make it clear that the

eventual costs for the mission will be a very small fraction of the satellite construction and launch costs. Furthermore, the cross sections needed to model the process have all been measured. Finally, no new technologies need to be invented, and substantial amounts of new capital equipment are not required. The

principal difficulty we anticipate in this phase of the project is the administrative scheduling of the available facilities or technology.

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ICF Applications Studies

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ICF Applications Studies

Introduction

R. O. Bangerter

The focus of the ICF Applications Group at LLNL shifted dramatically this year from far future applications to the near term design of the follow-on facility to Nova—the Laboratory Microfusion Facility (LMF). The Inertial Fusion Division (IFD) in the Department of Energy (DOE) Defense Program (DP) formed a Committee to study the LMF concept. This Committee consisted of representatives from IFD and seven institutions involved in ICF research: Lawrence Livermore National Laboratory, Los Alamos National Laboratory, Sandia National Laboratories, the Naval Research Laboratory, KMS Fusion, Inc., the University of Rochester, and Lawrence Berkeley Laboratory. In the first phase of the study, the Committee established required and desired features that would be needed to meet the ICF goals for the LMF. This effort spanned all of 1987 and included four full Committee meetings and numerous subcommittee meetings.

The Subcommittee on the Experiment Area (EA) was directed by LLNL. The results of this group's efforts, a portion of the larger DOE study, is the subject of the article, "DOE Requirements for the LMF Experiment Area."

In the article "Design Issues," we discuss the design considerations that must be resolved to successfully operate an LMF. These issues include developing vaporization and condensation models, studying beam quality degradation after transport through protective but turbulent gases, and determining damage thresholds for fused silica exposed to x rays. We also examine neutronics design issues that include the performance of composites such as Kevlar (compared to Al 5083) as a first wall material and the contribution of activated impurities to the total radiation dose rate possible from aluminum (see "Neutronics Issues").

Because cost is a major concern in the design of this facility, we also evaluated the possibility of using the existing Nova target chamber as the LMF's EA to lower costs. The article, "Using the Nova Target Chamber for High-Yield Targets," introduces the use of carbon as a neutron moderator and provides new insight into the behavior of the hot gases released after a shot—the jetting effect that might actually direct the gases away from the final optics.

Near the end of 1987, the LMF Conceptual Plan and Design (CP&D) team was formed to begin a study that will culminate in a formal CP&D report. This team includes all members of the LLNL Applications Group.

General studies conducted in 1987 concerned two familiar ICF themes: safety and economics. In "Tritium Safety Characteristics of Cascade," we continue to examine the tritium safety issues associated with Cascade (an ICF reactor concept), building on previous efforts that extensively considered the safety of this design.^{1,2} In these studies, we also examine the tritium safety implications of locating the target fabrication facility on site with the ICF power plant complex. Although this configuration increases the on-site tritium inventory, we found that recent experimental results show that the conversion of HT to the much more biologically

harmful HTO occurs more slowly than previously thought. These results imply that inherent power plant safety with respect to tritium can be maintained even with this site configuration.

Another radiological concern in designing an ICF reactor is the activation of the tamper material used in heavy-ion targets. Although the large throughput (grams per day) might imply a large inventory of waste, we show that lead can be safely recycled through a fuel pellet cycle without the accumulation of large quantities of waste (see "Target Debris Safety for an ICF Reactor"). In a related article, "Contribution of Lead Isotopes to the Radiological Hazard of Natural Lead," we show that lead depleted of the isotope ^{204}Pb would be significantly less hazardous than natural lead as a tamper material.

In an update to a 1985 Laser Annual article,² we consider the economic requirements for a heavy-ion beam driver (see "Gain Required at Fixed Heavy-Ion Driver Energies to Achieve a Competitive Cost of Electricity"). As we expected, a competitive cost of electricity is possible for plants in the 1 to 2 GW_e range, using a 10-MJ driver to obtain a gain of 50.

Laboratory Microfusion Facility

DOE Requirements for the LMF Experiment Area

The primary experimental goals for the Laboratory Microfusion Facility (LMF) were established by the U.S. Department of Energy (DOE). These goals are based on the ICF Program Mission and the LMF Utility Statements and can be divided into the following five categories:

1. Obtain high gain—gains of >10 are needed to begin most applications; gains significantly ≥ 100 are ultimately desired.
2. Conduct second and third generation nuclear weapons physics experiments—thermonuclear yields >100 MJ are required; yields up to 1000 MJ are desired.
3. Conduct nuclear effects studies and vulnerability assessments—yields >100 MJ are required; yields up to 1000 MJ are desired.
4. Assess ICF's potential for electric power generation.
5. Assess ICF's potential for other military and civilian applications.

The LMF consists of three major systems: the driver, the target fabrication facility, and the experiment area (EA). In this article, we discuss the EA capabilities,

established by an LMF Committee (formed by the DOE), that are necessary to achieve the LMF objectives. These capabilities can be set up as a matrix of required and desired features (Fig. 7-1). Capabilities are only designated as "required" when they are fundamental to achieving the minimum of the stated goal. They are categorized as "desired" when they are not fundamental requirements but would enhance the flexibility of the facility, expand the range of applications experiments, or explore the feasibility of ICF facilities beyond the LMF itself. Obtaining high gain, conducting weapons physics and weapons effects experiments, and determining ICF's electric power potential are all fundamental goals for the LMF, and all have both required and desired features associated with them. The ICF applications beyond the LMF (e.g., tritium or plutonium production and space propulsion) are not considered fundamental; therefore, capabilities for these applications all fall into the desired category. We have made the facility requirements driver independent (i.e., whichever driver is eventually selected will have to satisfy the facility EA requirements set in this article).

The third axis of the matrix shown in Fig. 7-1 is time. Some of the required and some of the desired capabilities would be needed as soon as construction is

Goal Level of desirability	Obtain high gain	Weapons physics	Weapons effects and vulnerability	Electric power	Other applications
Required					
Desired					

Fig. 7-1. The EA required and desired capabilities for accomplishing each LMF goal can be considered in a matrix. The third axis of the matrix is time.

complete and the experiments begin; most of the capabilities associated with obtaining high gain fall within this category. Other capabilities that support weapons-physics and weapons-effects experiments would not be needed until high gain is obtained, and the appropriate high gain targets are developed.

The instruments and structures that comprise the EA must accomplish their functions in the severe environment created by an exploding ICF target. The functions of these systems are:

- To establish the proper preshot conditions.
- To provide a suitable interface between the driver and the target.
- To emplace and support the target before the experiment.
- To make the measurements necessary to diagnose the target's performance and to determine the experiment's outcome.
- To protect personnel, instruments, and data from the effects of the energy release.

In an LMF, the direct, prompt effects of the emitted x rays, neutrons, gamma rays, charged particles, and debris are electromagnetic pulse (EMP), shrapnel, ablation, shocks, spall, pressure pulses, and thermal stresses. Delayed effects such as those associated with hot vapors, liquid metals, induced radioactivity, unburned tritium, toxic materials, corrosion, and condensation may affect not only the experiment in progress, but also the ability to do future experiments rapidly in the same chamber. Other more detailed descriptions of this harsh environment are included in the next two articles in this subsection: "Design Issues,"

and "Neutronics Issues." Any proposal for an LMF must include an estimate of these effects for the particular design and must describe how they will be controlled.

For this discussion, we separated the EA functions into three specific areas:

1. The target support structures in the chamber.
2. The diagnostic instruments and systems.
3. The structures necessary to establish the preshot environment, to provide the interface between the driver and the target, and to protect personnel, equipment, and data from the postshot environment.

Target Support Structures.

High Gain Experiments. The target support equipment for high gain experiments is highly specialized and has three functions: to position the target for the experiment, to create the local environment to sustain the target until shot time, and to verify that the target is in the desired location and configuration at shot time. This equipment must occupy a preferred location in the target chamber that is not easily compromised, and it must be designed as an integral part of the chamber. The target support system is required for cryogenic and noncryogenic targets (with and without tritium), indirect- and direct-drive targets, and nonimplosion targets.

The most important part of the target unit in the support system is the target itself. The construction and environmental requirements of the target dictate the design and operation of the target support system. The mounting apparatus, effectively part of the target, may consist of a stalk, a film web, or thin filaments. If a levitated target were required, a new

dimension of complexity would be added to this support system.

Certain classes of targets require sophisticated alignment procedures. Because there can be as many as five or six degrees of freedom in positioning the target, it must be aligned using fixtures or other aids. Most of these aids will be precisely affixed to the target during the fabrication procedure and will interact with alignment instruments in or on the target chamber.

Many of the targets used at the LMF will have cryogenic fuel layers. These cryogenic targets will be filled with significant amounts of tritium that will produce milliwatts of heat. This heat must be controlled and removed from the

vicinity of the capsule from the time it is inserted in the target chamber until shot time. These complex and costly targets will be fragile and will require careful handling to ensure that the target is neither lost nor damaged during insertion and positioning in the chamber. Many experiments (e.g., basic research on driver energy to x-ray conversion efficiencies, on hohlraum characterization, and on dudded implosions) will use non-cryogenic targets, which do not require such careful handling.

Any target handling system proposed for an LMF must perform the following eight functions.

1. Transporting the target to the working point. The target may need to be moved several meters from the entry access port at the chamber wall to the working point (i.e., the point to which the beams are focused). During this journey, it will need to be protected from the environment of the chamber.

2. Positioning and orienting the target. The target must be precisely positioned at the working point, and it must be oriented properly in relation to the diagnostic instruments, the driver beams, and any secondary target structures. For most targets, alignment aids will be required that may consist of cross hairs, light-emitting diodes (with or without optical fiber links), and retroreflectors. (Figure 7-2 is a proposed target-positioner concept.)

3. Filling the target. The fuel (gas or liquid) must be placed into the target. This can be done before the target is inserted or at the working point.

4. Protecting cryogenic targets. Targets with cryogenic fuel must be protected and maintained between the fill time and the shot time. This may require some last-moment "uncovering" of the target, perhaps only a few tens of milliseconds before the shot. The fuel layer will be preserved by flowing helium gas around and maybe through the target, which means this cold gas must be directed to the working point and, perhaps, removed if it cannot be exhausted into the chamber.

5. Inspecting the targets. The inspection, verification, and documentation of the target's configuration prior to the

Fig. 7-2. A proposed target-positioner concept.

shot is vital in analyzing the target's performance. The position and orientation of the target must be verified, and the quality of the cryogenic fuel layer must be ascertained. This information must be recorded outside the chamber because it is used first to decide whether conditions are correct to fire the shot and later to determine if the target configuration affected the results. Most of the data will likely be optical with some of it being produced by very sophisticated diagnostic systems. Some electronic signals may be generated near the working point and must also be relayed to recording devices outside the chamber.

6. Recovering or disposing of target support equipment debris. Because spent target components must be recovered, this equipment must be positioned in the chamber in a way that facilitates expedient removal. The target and perhaps several tens of centimeters of supporting material will be vaporized during the shot; however, some of the remaining hardware (although melted, torn, or bent) will remain attached to the positioning equipment. This material must be removed and disposed of before the next target can be inserted and aligned.

7. Providing a monitoring and control system. There must be electronic communication links between the target handling apparatus and the master control. In addition to the alignment verification signals discussed earlier under function 5, there must be synchrony with the driver to remove any protective shrouds at the last instant before the shot and to provide a go/no go target-verification signal.

8. Remote handling of the target inside the chamber. Maintenance of the target handling hardware must be accomplished by remote manipulation because of the nuclear activation of the chamber and nearby structures. Because remote handling will be used for most of the interior components, it will be treated in detail elsewhere.

Weapons Physics and Weapons Effects Experiments. The EA requirements outlined for high gain experiments also apply to weapons physics and weapons effects experiments in which we study the effects of weapon output interacting

with the environment or other military systems. There will be some added requirements because the test package for these experiments will often be much larger in mass and in dimension than the target in high gain tests.

In some weapons physics experiments, the test package is distinct from the target but is in close proximity to it. Some provisions must be made to support this package and to align it with the target and diagnostic instruments.

The facility requirements for these experiments may also be affected (1) if the test package has portions of diagnostic instruments mounted on it, and (2) if the experiment requires external power, gases, or fluids.

Some weapons physics experiments will require large structures more distantly located from the target. These structures may have to be placed in the target chamber through ports other than the target insertion port. In these experiments, alignment will present a different and perhaps more difficult set of requirements.

Weapons effects simulations have some unique requirements. Although the target insertion and alignment requirements are essentially the same as those for weapons physics experiments, the test packages may be quite large and may include a large number of detectors and associated data-recording circuitry. Furthermore, in contrast to the weapons physics experiments, the device under test is valuable and should survive the experiment in working order.

Diagnostic Requirements.

High Gain Experiments. The diagnostic requirements for high gain experiments are divided into four measurement categories corresponding to the four major areas of ICF research:

1. Driver performance at the target.
2. Energy transfer from the deposition point to the ablation surface.
3. Capsule implosion.
4. Burn performance.

LMF proposals should, in general terms, describe each measurement and the diagnostic equipment needed to make the measurement or should discuss why a particular measurement is not applicable or usable in the facility. Some instruments

may be used to fulfill more than one diagnostic requirement, and this should be stated in any proposal. The lists of possible measurements included in this article are not intended to be all-inclusive, but they should be considered the minimum set of measurements for diagnosing high-gain ICF targets (see Tables 7-1 through 7-4).

Many of the target performance diagnostics will be the same for any type of driver. Those diagnostics can be noted in a proposal but do not need to be described in detail. However, in some cases, the needs of a specific driver may preclude using a standard fusion diagnostic. In those cases, each diagnostic should be described briefly, and its measurements should be related to the requirements. The description should include: (1) the instrument's sensitivity and operating requirements (i.e., vacuum and x-ray or optical line-of-sight requirements and any shielding needs), (2) triggering and timing requirements, (3) data recording,

retrieval, and analysis methods, (4) mechanical support, and (5) any other special requirements.

The relationship of diagnostic operation to the target chamber environment should also be discussed in LMF proposals. This discussion should include the compatibility of target-chamber vacuum and blast containment requirements with the vacuum and line-of-sight requirements for the diagnostic. Other issues that should be addressed are the diagnostic's survivability and susceptibility to damage in the ICF environment. Each instrument should be evaluated to determine if the radioactivity produced by the target and the residual radioactivity in the system will affect diagnostic operation (e.g., sensitivity). The instrument's potential for contamination from these radioactive sources must also be addressed.

Initially, all LMF experiments will be in the first three measurement categories (see Tables 7-1 through 7-3). In these experiments, instrument sensitivity will be

Table 7-1. Driver performance diagnostics (category 1) measure the incident driver conditions so that the effects of these conditions on target performance can be evaluated.

Measurement	Definition/notes
Incident driver energy	The amount of driver energy that is available to couple to the target, and its spatial distribution focused at the target.
Driver pulse history	The temporal dependence of the driver energy delivered to the target.
Driver symmetry	The pointing, focusing, and energy distribution of the driver energy delivered to the target. The detail and accuracy of this diagnostic depends partly on the target design (i.e., direct or indirect drive) and partly on the type of driver. The diagnostic should be assessed to determine the effect of driver symmetry on target implosion symmetry.
Driver synchrony	The relative timing of the energy delivered by multiple driver beams to the targets. The required accuracy depends on driver type.
Spectral content	The spectral purity of the driver energy delivered to the target. For lasers, the spectral purity is the bandwidth of the light focusing on the target. For particle beams, the spectral purity is the relative abundance of the ion species and their velocity distribution focused at the target plane.
Spatial profile	The peak-to-average intensity across the spatial profile.

Table 7-2. Energy coupling diagnostics (category 2) evaluate the effective transfer of the incident driver energy from the point of deposition to the ablative surface of the imploding capsule. These diagnostics are more specific to the type of driver and to whether the target is directly or indirectly driven.

Measurement	Definition/notes
Transfer efficiency	The fraction of driver energy deposited in the target that is effectively transferred to the ablative surface of the imploding capsule.
Transfer symmetry	The symmetry of power intensity deposited at the ablative surface as a function of time.
X-ray drive	The intensity, spectrum, and time history of x-ray drive at the ablative surface (measured for indirect drive).
Fuel preheat	The magnitude, time history, and spectral content of any mechanism that may penetrate the dense pusher layers and heat the fuel before compression. This may include preheat from x rays, electrons, or particles.

Table 7-3. Capsule implosion dynamics diagnostics (category 3) measure the capsule's implosion performance and assess the quality of the fuel assembly. These measurements can be done without producing high fusion yields by using pure deuterium fuel or nonoptimum mixes of deuterium and tritium fuel instead of burning DT.

Measurement	Definition/notes
Pusher position vs time, ($r-t$)	The time dependence of the dense pusher shell radius during its implosion phase. This measurement technique may consist of photographs of the shell at various times or continuous time mapping of its radius. The final pusher, fuel, and spark plug radii should also be measured to determine overall target convergences.
Implosion symmetry	The symmetry of the fuel and the dense pusher as they implode and in their final position at ignition. This measurement should be correlated with the driver and target symmetry.
Instability and mix	The stability of the ablative surface, the interface between the pusher and the fuel, and any other accelerating or decelerating interface in the target design. The diagnostics should measure mixing of the layers caused by hydrodynamic instabilities and/or turbulent mixing.
Fuel ρ and ρr	The final average and peak fuel density ρ , and the fuel areal density ρr . These measurements assess the quality of the final fuel assembly.
Fuel temperature (no burn)	The fuel temperature in the spark plug resulting from the implosion (i.e., without significant heat from α -particle deposition).

Table 7-4. Fuel burn diagnostics (category 4) measure the burn performance for high gain targets. These diagnostics should be able to measure normal- and low-performing targets. The debris and radioactivity hazards that these diagnostics must withstand will be more severe because these targets may produce significantly more energy than duded-fuel experiments. Any LMF proposal should discuss how measurements on the high yield targets will be related to the target measurements from Tables 7-1 through 7-3.

Measurement	Definition/notes
Ignition time	The time of capsule ignition relative to the amount of time that the drive pulse takes to reach the target.
Target emissions	The flux, fluence, and spectra of the neutron, gamma-ray, x-ray, and debris emissions from the target. The time dependence of the fluxes should be determined so that both yield and burn rate can be inferred.
Time history	The temporal dependence of the burning fuel.
Fuel temperature	The average ion temperature of the burning fuel.
Spatial distribution	An image of the neutrons and/or gamma rays from the burning fuel region.

easier to maintain because the chamber environment will be free from any exposure to high thermonuclear yield. However, once high gain experiments (category 4, Table 7-4) are performed, the background chamber environment will change dramatically. Any LMF proposal must address the problem of experiments that are conducted after the high gain experiments begin, but still require the same instrument sensitivities achievable in a quiescent chamber. The proposals will need to show how to maintain instrument sensitivity in a nonquiescent chamber and what the instrument sensitivities will be. Table 7-5 is a list of proposed low- and high-yield diagnostics that may be included in an LMF design.

Weapons Physics Experiments. Weapons physics experiments include those that use only the driver energy and those that use the energy output from a medium- to high-gain capsule. Diagnostics for the former class of experiments

should be similar to those used to characterize target environments in which high gain is achieved. Diagnostics for the latter class must be sensitive to the higher temperatures and pressures that are characteristic of high-yield capsules.

Mix and Hydrodynamics Experiments.

Experiments to study mix, hydrodynamic instability growth, projectile acceleration, and equations of state will require diagnostic techniques similar to those used in characterizing capsule implosion dynamics and drive conditions (e.g., energy coupling diagnostics). The positions of interfaces and tracer layers must be tracked in one and two dimensions as a function of time. Spectroscopy, radiography, and other diagnostic methods should be able to measure the densities and temperatures of the interfaces or tracer layers as a function of time. Any LMF proposal that includes x-ray backlighting as a diagnostic technique must describe how to achieve the necessary

Table 7-5. Possible LMF target-chamber diagnostics to measure sub-ignition and ignition experiments.

Diagnostic	No. required	Required solid angle (msr)
Sub-ignition diagnostics		
Energy balance	~50	2.0
Optical spectrometers	2	2.0
Optical/UV imager	1	28.5
X-ray diode array	1	28.5
High-energy x-ray array	1	4.6
Sub-keV x-ray imager	1	4.6
Soft x-ray time-resolved spectrometer	1	2.0
X-ray crystal spectrographs	2	2.0
X-ray imaging and spectrometer manipulators	2	3.2
High-resolution x-ray imager	3	12.7
Neutron yield detectors		
Silver	2	2.0
Copper	2	2.0
Indium	1	2.0
Neutron time-of-flight detectors		
Low yield	1	2.0
High yield	2	2.0
Ignition time and burn width detectors	2	2.0
Neutron activation detector	1	12.7
Neutron imagers	2	4.6
Ignition diagnostics		
Neutron yield detectors	2	2.0
Neutron and gamma-ray imagers	2	2.0
Ignition time and burn width detector	1	12.7
X-ray imagers	2	12.7
Neutron spectrometers	2	2.0
X-ray spectra and imaging access	2	28.5
Low-resolution x-ray imagers	2	12.7

backlight x-ray energy and flux, and the spectral, temporal, and spatial resolution dictated by the experiment. Multiple diagnostic lines of sight (or lines of sight different from those used to measure energy transfer from the driver energy deposition point to the ablation surface and to observe capsule implosion) may be necessary when backlighting is used.

Experiments that are driven by capsule yield will require measurements of drive environments. The diagnostics for these experiments must have capabilities appropriate to the higher energy density occurring in this environment. The capsule burn diagnostics described in Table 7-4 will also be required for these experiments. The drive characteristics (magnitude, spectrum, time history, and symmetry) and preheat mechanisms must be accounted for when conducting experiments in this regime. Finally, the effects of prompt capsule yield on the ability to diagnose weapons-physics-package parameters should be assessed.

Atomic Physics, Radiation Flow, and Opacity Experiments. Experiments to study atomic physics, radiation flow, and opacity will require the diagnosis of drive environments as do the mix and hydrodynamics experiments. In addition, these experiments will need extensive time-resolved spectroscopy and imaging (at keV and sub-keV energies), possibly along multiple lines of sight or different lines of sight than the diagnostics used to measure driver energy transfer to the ablator and capsule implosion. The state of the material being measured must be characterized. Typical experimental measurements should include: the spectrum as a function of time, both incident on and transmitted through a characterized sample; and the radiation and density distributions through the sample as a function of time.

Measurements for experiments driven by capsule yield should take into account the effects of the yield on diagnostic systems and the diagnostic requirements

necessary to measure different regimes of x-ray energy or flux.

Weapon Vulnerability, Lethality, and Effects Experiments. Diagnostics used for these experiments must characterize the target output, the effective radiation environment at the object under test, and environmental effects.

Target Output Diagnostics. These diagnostics are essentially the target emission diagnostics of fuel burn performance. They measure the yield and spectra of neutron, gamma-ray, and x-ray output from the burning capsule.

Test-Package Radiation Dose and its Distribution. These diagnostics measure the effective radiation environment at the test package. They include the following measurements:

- The neutron flux and spectrum as a function of time.
- The x-ray (<10 keV) and the capture gamma-ray spectra and flux as a function of time (different time and spectral resolution may be necessary to measure prompt and delayed effects).
- The spectrally integrated flux of x rays (<10 keV) as a function of time.
- The directional or angular distribution of the x-ray, neutron, and gamma-ray environment if the test package is located near a shield or scattering materials.

Target-Chamber-Environment Effects. Because EMP from an ICF target and target-chamber-generated EMP may affect system survivability and performance, they must be assessed at the test package location. Bandwidths and frequency ranges of importance will depend on the specific test package and on the type of material positioned between the working point and the test package. Debris from target support hardware, shields, and filters can have damaging effects on both the test package and diagnostics. Approaches to mitigate this damage will have to be developed for each proposed LMF configuration.

Environmental and Protective Structures and Systems.

High Gain Experiments. The environmental and protective structures and systems must first establish the preshot

vacuum, the temperatures, and the background noise levels required for beam transport, target data collection, and cryogenic target support. To perform these functions, specific preshot conditions (depending on the specific driver and diagnostics considered) must be established to allow the driver energy to reach the target area and also to allow adequate target-output radiation to reach each specific diagnostic where it can be measured. For instance, the preshot chamber pressure selected must allow beam propagation without unacceptable reduction in beam quality caused by air breakdown. Additionally, the x-ray portion of the target output has to reach the various x-ray diagnostic platforms (i.e., the mount, hardware, and actual diagnostic) at the required fluence. The chamber pressure will affect first-wall ablation and may provide some optics protection; therefore, the acceptable solutions to these inherent dilemmas may involve adding structures that differentially pump various parts of the EA, or moving diagnostic or final optical elements to a radius that eliminates the possibility of x-ray and neutron damage to these systems. The pumping system to provide an acceptable preshot chamber would then be included as an environmental and protective system under this requirement.

The pressure of the quarter-ton yield in high gain experiments must be contained in accordance with the intent of the ASME code. Meeting ASME code is not intended to preclude later facility modifications to accommodate larger yields or applications experiments. Containment here also means managing the shrapnel hazard to the first wall and other structures to support tritium confinement in the EA.

These environmental and protective systems must satisfy all health and safety standards for prompt dose, residual dose rate, waste disposal, routine releases, accidental releases, and toxic hazards. The radiological consequences of a design-basis accident must not exceed 25 rem to the whole body, 300 rem to the thyroid, 300 rem to the bone surface, or 75 rem to the lung.³ The dose to members of the

public from all routine operations cannot exceed 500 mrem/yr for occasional exposures and 100 mrem/yr for prolonged periods of exposure (>5 years). Radioactivity discharged into the atmosphere from routine operations cannot result in a dose >25 mrem/yr to the whole body or 75 mrem/yr to any organ of any member of the public.⁴ Exposure to an employee is limited to 5 rem/yr. The LLNL design guidelines for radiation exposure (0.5 rem/yr) will apply to the LMF.⁵ Radiation detection and monitoring equipment to support the radiation safety program must be identified and included on the platforms in the EA.

The environmental and protective systems must also protect diagnostics against loss of data and allow high-cost diagnostics to survive. Certain diagnostic measurements must be made to support high gain development (as discussed earlier in this article). The preshot environment established by the environmental and protective systems must reach conditions where these measurements can be made by the specific instruments chosen or specially designed for the LMF. If a diagnostic is valuable, then it must be protected by the platform supporting it. If the diagnostic cannot or will not be protected, then it is expendable and must be inexpensive. This trade-off, determining which diagnostics should be expendable and which should be protected, must be resolved through consideration of the experimental goals vs the operational costs for the facility.

Collected data may be in the form of exposed film or electronic signals that will be sent by means of cables to a central collection or processing point. (The actual data-handling system should be specified in the LMF proposal.) The film, cables, or other components of this data handling system may need to be protected from decay gamma radiation, activated EA components, debris, waste, loss of an instrument's interior vacuum (which could occur during EA radiation cooldown), and prompt gamma rays and neutrons that may produce EMP or transient radiation effects on electronics (TREE).

These same environmental and protective systems must also allow any EA equipment to be serviced and repaired starting at most one month after an unplanned outage. The EA will contain both nuclear materials and those systems and structures designed to contain and remove such material. An unplanned equipment outage could disrupt these systems and the experimental sequence. The goal for an LMF facility is to ensure that a comprehensive and aggressive service and repair operation can begin within one month after any conceivable outage and under the worst possible conditions. This implies limiting the biological hazard present after the accident and planning for immediate access to equipment that may be needed for the cleanup.

The required number of LMF shots for achieving high gain is:

- ≥ 100 experiments at 100 to 1000 MJ,
- ≥ 300 experiments at 10 to 100 MJ,
- ≥ 1000 experiments at ≤ 10 MJ.

The required annual rate is ≥ 300 target experiments (≥ 600 is desired). The required maximum-yield shot rate is ≥ 1 shot per 10 days (1 shot per week is desired). The LMF Committee constructed a specific series of target design experiments that will lead to high gain. The Committee working group on the EA reviewed these experimental objectives and reached a consensus on the above number of experiments at specified yields over a given time period. The consensus evolved by anticipating various contingencies and by the desire to obtain high gain within 5 yr.

Issues such as fatigue, radiation damage, activation, and waste can only be resolved when we can accurately calculate the total integrated fluence that the structures will be exposed to over the facility's lifetime. At least 1400 experiments with the relative yields listed above will be conducted in order to achieve reliable high gain. The amount of time required to obtain high gain can be determined using the maximum-yield shot rate and total number of shots set by an LMF designer.

After high gain is achieved, military applications experiments (e.g., weapons

effects and physics experiments) may be conducted at this same facility (perhaps using multiple target chambers in which experiments for several applications can take place concurrently). Potentially, even the high gain development goals may be met using more than one chamber. The total time required to complete all experiments in the high-gain series is about 4.6 yr (1400 experiments at 300 experiments per year).

Any LMF design must allow personnel access to the diagnostic platforms, located outside the neutron shield, within 24 hr after each shot. Although the diagnostic sensors will be inside the vacuum chamber and neutron shield, much of the data recording and processing instrumentation will be outside the chamber. Rapid EA reentry may be necessary to preserve data and to expedite shot turnaround.

It is desirable for LMF structures to meet shallow burial waste standards at the end of the facility lifetime. Another desirable feature is that all waste produced from the facility should meet shallow burial requirements during and at the end of facility operation as outlined in *10 CFR 61* (Ref. 6) or the applicable fusion industry standard. (Many relevant nuclides are not currently listed in this *CFR*.) Shallow burial waste is generally less biologically hazardous than deep burial waste; therefore, meeting the shallow burial standards will significantly reduce the cost of processing the waste and will result in a safer facility in case of a severe accident.

It is also desired for EA systems to handle small amounts of fissionable material. Structures for this task would include those that remove the material after the experiment, separate the material, store the material, and finally transport and dispose of the material. The shallow burial goal is still desired even if EA systems are handling fissionable material.

Another desired feature is for the EA systems to allow rapid (within 24 hr) reentry to the interior of the chamber while meeting all safety requirements (protective suits and breathing apparatus

may need to be used to ensure personnel safety). Attainment of this reentry goal may have a significant impact on LMF operations, especially on sensor setup, data collection, target alignment, and experimental setup. If rapid reentry is not achievable, then robotic systems must be used for all functions inside the chamber, and any LMF proposal must describe these systems.

Weapons Physics Experiments. The LMF Committee established the specific weapons-physics capabilities that should be demonstrated by the LMF. They evaluated these capabilities and arrived at a consensus as to the weapons-physics experiment rate and total number of experiments at specific yields. This experiment rate and total number of experiments required for both high-gain and weapons physics experiments are:

≥500 target experiments per year

(≥800 experiments per year are desired),

≥1 per week at 1000 MJ (1 experiment every 3 days is desired),

≥1000 experiments at 100 to 1000 MJ,

≥3000 experiments at 10 to 100 MJ, and

≥2000 experiments at <10 MJ.

Weapons physics experiments in an LMF will require some target and technique development (e.g., the capability to vary the radiation output of the target). Fortunately, some variations in output can probably be adapted from the weapons effects program if it occurs concurrently. Each experiment in the weapons physics series must be reviewed to determine the shrapnel hazard, the complexity of the structures needed to support the experimental platform (i.e., the working point combined with the physics experiment), and the implications for waste and cleanup. Any LMF should be designed to meet the experimental requirements of the most severe postshot environment planned for the facility. The weapons physics review process may establish that more than one chamber is required for physics experiments; this is an acceptable option if it is shown to be the optimal solution.

The various LMF physics experiments will require a range of yields, but the low

end of this range will not be $\leq 5\%$ MJ (the driver alone would suffice for yields below this value). The rate at which high yield shots are made will be increased over that for the high gain development series (from one every ten days to one every seven days, which is the desired rate for the high gain series). The desired rate for physics experiments significantly improves on the required rate by allowing one 1000-MJ shot every three days. Proposals to modify the chamber (or chambers) used for the high gain experiments to accommodate these physics experiments are acceptable.

All required and desired features applicable to high gain experiments also apply to physics experiments because they also require high gain. Other desired features for physics experiments include beam pointing and focusing so that more than one object can be irradiated at nearly the same time. In addition, some weapons physics experiments call for preheating certain materials at or slightly before capsule ignition. Although the designer will decide on how to meet these preheat requirements, using a portion of the driver may be the most cost effective and least complex option.

The EA waste systems must be able to handle fissile material in order to support the complete set of weapons physics experiments. Using the LMF for weapons physics experiments will introduce significantly more transuranic and fission product waste to the other radiological wastes. This actinide/fission product waste may need to be separated and perhaps temporarily stored until it can be contained and transported away for disposal. Decontamination measures to allow reentry into the chamber after 24 hr are desired, but, if this cannot be achieved, in-chamber functions must be performed with robots; these robotic techniques should be fully explained in any proposal.

In weapons physics experiments, the close proximity of certain experiments to the working point will exacerbate the shrapnel hazard; in addition, the first wall and all structures will have to withstand a generally greater impact load

than in the high gain experiments. In addition, platforms for these experiments must accommodate setup and alignment of all required weapons physics diagnostics in order to conduct successful experiments.

Vulnerability and Weapons Effects Experiments. The vulnerability and weapons effects experiments might not be started until high gain is reliably achieved, until the proper target output characteristics are obtained and characterized, and until the desired exposure configurations are designed and tested. The characterization process will be complicated by the amount of hardware that will be required to provide features such as neutron shields, conversion schemes, and blast or shrapnel screens. Another complication is the fact that the incident x-ray fluence and spectrum will be modified from those emitted from the capsule. The required number of shots and shot rates at specific yields for these experiments are:

- ≥ 10 target-output-development experiments during the weapons-effects experimental series,

- ≥ 6 exposures per year at 100 to 1000 MJ,

- ≥ 100 lifetime exposures at 100 to 1000 MJ,

- ≥ 10 exposures per year and 200 lifetime exposures at 10 to 100 MJ.

Source development experiments will be used to characterize the output of target assemblies designed especially for use in weapons effects experiments. A desirable feature would be for the chamber to be able to sustain a repeat of the initial weapons effects experiments with minimal repair or upgrade.

All EA systems must provide large solid angles and an area ($\sim 6 \times 3 \times 3$ m) in which to expose an object. A single object should achieve 50% uniformity at a fluence of ~ 1 cal/cm². The overall effects of all possible materials (e.g., blast and debris shields) placed between the working point and the exposed item must be considered in determining the incident fluence. Neutron doses and dose rates at the test item are a major concern because they could spoil the fidelity of

an x-ray effects experiment. This reduced fidelity is caused by neutrons that affect the test item and mask, counter, or contribute to the effects expected when the test item is exposed to x rays; consequently, the measurement of the specific x-ray effect could become very difficult or impossible. To prevent this from occurring, the neutron dose and dose rate must be reduced to $<1\%$ of the values that can cause the transient radiation effects on electronics that are of interest in the particular experiment.

The EA systems must allow for supporting large masses with high dynamic loading. For instance, large assemblies for tailoring target output may surround the capsule. The EA systems must include the means to install, rigidly support, and clean up such assemblies without creating a shrapnel hazard to the test item or unsafe conditions for workers or the public. The test items must be supported rigidly because of the high dynamic loads that will develop in such items.

The flexibility that is needed in simulation environments requires altering the spectra, energy partitions, and pulse lengths of the target output. Proposals to accomplish each of these modifications must discuss how each is to be accomplished and must show that all EA assemblies are adequately supported, reliable, and safe, and that each provides the desired net output. The entire net output must be characterized in any LMF proposal.

The EA systems must protect exposed items from collateral effects. Schemes must be devised and in place to protect test items from the following: shrapnel, excess neutrons, prompt or decay gamma radiation that could affect test results, and target blast. The test items must also be protected from being contaminated by activated or toxic material that would render the article hazardous. The item itself cannot become so activated that it cannot be easily handled 24 hr after a shot. The test items must be removed from the chamber within 24 hr so that they can undergo postshot examination quickly and so that the chamber can be cleaned and set up for the next shot.

Multiple target chambers are a desired feature for an LMF because they may allow work on the other applications (e.g., weapons physics) to proceed concurrently with the vulnerability and weapons effects experiments. The anticipated long setup and refurbishment times as well as the unusual required configurations make it desirable to use a separate target chamber for this application.

All proposals must show how each of the systems within the EA performs its function under nominal conditions, and also what will happen to each system in the event of a failure or partial failure (e.g., unexpectedly low or high target yield). The proposals must also examine the effects of the subsystem designs on each other, the experiment rate, the postshot environment, robotics requirements (either routine or emergency), the experiment execution sequence, operating costs, waste disposal, and accident potential.

Commercial Power Applications. The primary long-term goal included in the ICF Mission Statement is to determine the potential of ICF as a source of electrical energy. If gains of ~ 100 are achieved in the LMF, it would be clear that an electric power plant could be built that would have a power output about a factor of 3 greater than the power required to run a reactor. This is sufficient for economic feasibility at some power plant size. Unfortunately, if the high gain is achievable only at a high drive energy, then the plant size at which ICF is economically competitive would be very large, perhaps so large that no one could afford to build such a plant. To compete, ICF will have to establish not only that high gain can be achieved but that it can be achieved at a low drive energy. However, the nature of target physics makes it much easier to obtain high gain at high drive energy rather than at low drive energy. It is expected, therefore, that the initial attempts to achieve high gain will first occur at the highest drive energy obtainable in the LMF. Once this high gain is achieved, further target physics experiments designed to obtain the highest gain at the lowest drive energy

possible will be required to determine the feasibility of the energy application. This would involve optimizing the target's utilization of drive energy and exploring completely different target types. Options such as direct drive targets, double shell targets, and polarized fuel targets are some of the possibilities that would be studied.

Another energy-related LMF study involves quantifying various system options that could be used as system trade offs to reduce the cost of a commercial facility without affecting its electric-power-producing capabilities. For example, the cost of a reactor is minimized if all the driver beams enter the reaction chamber from a small number of beam directions, preferably all located on the same horizontal plane. However, our calculations indicate that there is a drive energy penalty for doing this (i.e., more drive energy, which adds cost, is required for this configuration than for one in which the beam directions are more uniformly spread over the surface of a sphere). In addition, costs of a power plant will be reduced and the practicality of ICF will be increased if some of the high technology requirements placed on a research facility such as an LMF can be relaxed for a power plant (e.g., relaxing the precision of the pulse-shaping requirement). The initial high gain experiments will create the target conditions to produce high gain; therefore, it will be necessary to conduct experiments that reveal the drive energy penalties incurred when other parameters are varied in ways that would both save cost and increase reliability in other parts of the plant.

Finally, if the above studies indicate that ICF is indeed a promising energy source, then the way must be cleared for the design of an engineering test reactor, a materials test reactor, and an economical demonstration power reactor. Although designing such facilities is beyond the scope of the LMF charter, the LMF could be used to perform experiments that would provide the fundamental data necessary to design such a reactor, and an LMF is the only facility from which we could gather such data.

For example, we could examine the feasibility of various proposed first-wall materials with an LMF. We could also use an LMF to test target injection and tracking systems developed off line and demonstrate (before another large driver is built for the pulsed reactors) that a ballistically delivered target can be ignited.

Conducting a set of energy application experiments would have some impact on the LMF design requirements because these experiments could include:

- Building and handling a wider variety of target types.
- Varying the pulse shaping, beam quality, focusing characteristics, and other such variables.
- Changing the above variables quickly and inexpensively.
- Handling hot liquid metals, ceramic granules, and other material being considered as first wall candidates.
- Developing diagnostics that address the first-wall vaporization and condensation issues.

Other LMF Applications. There are many applications for ICF technology other than those discussed in this article. Basic scientific work studying the conditions of matter at extremes of temperature and pressure would interest many research communities. The laboratory x-ray lasers that could be pumped with a driver or an ICF capsule will be the brightest x-ray sources on earth, inspiring many biomedical and lithographic applications. ICF reactors could inexpensively and effectively produce plutonium and tritium for weapons or fissile fuel such as uranium-233 for reactors. For interplanetary missions, ICF propulsion engines and power systems for use in space would have great advantages over nuclear-electric propulsion or other advanced propulsion schemes in terms of achievable velocities and payload sizes.

Although the LMF could not be used to fully explore all of these applications, it would be the only facility in the world where high-yield ICF pulses could be observed in a laboratory setting. We anticipate that such a facility would be in great demand for conducting at least rudimentary experiments to allow a basic

assessment of ICF's potential in these other areas. Data from such endeavors would allow the proponents of these other applications to decide if and when they should build their own dedicated test facilities to more thoroughly evaluate the application. These test facilities might even consist of simply adding a separate experimental area to the LMF, using the same driver as the other experiments. Even though none of the required or desired features for the LMF are specifically oriented toward these long term applications, we feel that the future of fusion research in all areas will be better served if the LMF is designed so that it can be modified to address these applications at a later time.

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Design Issues

In an ICF high-gain (~ 100) facility, we will be able to conduct experiments with up to 1000 MJ of thermonuclear yield. The experimental area of such a laboratory facility will include many systems and structures that must operate successfully in the hostile environment created by these sudden large energy releases. In this article, we discuss many of the nuclear effects that will occur in this environment and the implied design issues, and we suggest possible solutions so that a useful experimental facility can be built.

The evidence is now good that, if an ICF driver capable of delivering 10 MJ of energy in suitable form is built, fusion energy gains of up to about 100 can be obtained, resulting in yields of up to 1000 MJ (equal to about one-quarter ton

of high explosive). Our studies have shown that such gains and yields are desirable for both military and civilian applications. To perform their tasks in support of these applications, the structures and systems in the next ICF research facility must be designed to account for the extreme environment created by this large thermonuclear yield.

Structures and systems in the experimental area can be divided into four groups:

1. Target support structures that are in contact with or very near to the target. These structures create the proper environment for various types of targets (including cryogenic targets), provide target positioning and alignment features, and verify proper target configuration before an experiment.

2. Diagnostic systems that measure target performance during four phases of operation: deposition of driver energy, transfer of energy from the deposition surface to the capsule ablation surface, implosion of the fuel capsule, and burn of the thermonuclear fuel. Some of these systems will require a hard vacuum (10^{-5} Torr) between the target and the sensor. Other systems will need to be placed relatively close to the target where they must be protected from exposure to postburn effects. Some systems that make preburn measurements may not survive long enough in some postburn environments to transmit their data in a useful form. In these cases, the preburn measurements will have to be made in "dudged" fuel experiments; these systems will be removed for burn experiments.

3. Protective systems that include systems designed to protect people, equipment, structures, or data from the effects of the thermonuclear burn.

4. Beam handling systems that provide transport and accurate pointing and focusing for the driver beams and that establish the environment necessary for the beams to reach the target. Near the target, vacuum may have to be below 10^{-1} Torr to prevent air breakdown caused by the intense laser beams as they reach the target area.

These last two systems can be located at the most cost effective distance from the target, but all of the systems must be designed to permit us to conduct high-yield experiments about once a week with many low- or no-yield experiments in between.

The prompt effects of the target explosion include x-ray ablation, neutron and gamma-ray doses, electromagnetic pulse, shrapnel, shocks, spall, pressure pulses, and thermal stresses. The delayed effects include those produced by induced radioactivity, unburned tritium, hot vapors, liquid metals, toxic materials, corrosion, and condensation. The structures and systems must be carefully designed to perform their function while these effects are occurring. Because these systems and many effects are interactive, the designer will have to avoid making one effect worse while attempting to mitigate another effect.

Containing the Thermonuclear Yield.

When a 1000-MJ thermonuclear capsule explodes, about two-thirds of the energy

is emitted in the form of high-energy neutrons. Most of the remaining energy is emitted as varying fractions of x-ray and debris energy. The neutrons have a long range, whereas the x rays and debris have a very short range, which (combined with the short duration of the pulse) results in large specific energies deposited in materials.

Figure 7-3 shows the specific energy as a function of depth in aluminum for the x rays from a 1000-MJ target at a distance of 10 m. This figure also shows the cohesive energy E_c (the energy necessary to raise Al to its boiling point plus the heat of fusion and vaporization) and the boiling point energy E_b . This deposition profile is established so quickly that heat transport within the Al does not play a significant role. It is clear that all the material above the cohesive energy will vaporize and will have a temperature significantly above the boiling point. It is also clear that some of the material between E_c and E_b will vaporize. As can be seen, between 0.5 and 2.0 μm of Al will vaporize; these thicknesses increase as the Al is moved closer to the target. At 1 m from the target, tens to hundreds of microns of most materials will vaporize; at 10 cm, a few centimeters of material will vaporize.

As the vaporized material expands, it imparts momentum to the unvaporized material behind it and may also create a shock. If the material in the first wall can be crushed, the shock will die out, but the momentum imparted is conserved.

Figure 7-4 shows the momentum imparted per unit area for lead, beryllium, iron, and aluminum as a function of the x-ray fluence on the object. For 300 MJ of x-ray and debris energy, the horizontal scale covers the distances from 2.5 cm to 5 m. (Note that, to determine the largest momentum that may be transferred, debris is included with the x-ray output, but we do recognize that debris will play a very small role or no role at all in transferring momentum to chamber materials.)

The momenta imparted to the metals cover four orders of magnitude; we used a simple one-dimensional approximation to calculate these curves, so they really represent upper limits and should not be applied to small objects where two- or

Fig. 7-3. The specific energy in Al at 10 m from a 1000-MJ target.

Fig. 7-4. Momentum imparted to unvaporized material by the x-ray-induced ablation of the first-wall surface.

three-dimensional calculations should be done. Nevertheless, we can make the following observations:

- Each curve is S-shaped because all output spectra from ICF targets have a hard x-ray "bump." As each type of material is moved closer to the target, even the deposition by very hard x rays eventually causes ablation.
- The inflection point is different for each material. This implies that as an object is moved closer to the target, it is better to fabricate it from lower Z materials. (Of course the use of low- Z material is not important if the object is completely vaporized.)

• Shrapnel is created if sufficient momentum is imparted to an unvaporized object. For example, an unvaporized, 1-cm-thick piece of Al at a distance of 10 cm could be given a velocity of 7.5 km/s, thereby becoming a hyper-velocity bullet. On the other hand, that same piece of Al at 1 m from the target would achieve a velocity of only 40 m/s. The curves in Fig. 7-4 show that, at a distance of less than 5 cm, any of these metals can be used because the shrapnel hazard is independent of material. However, between 5 and 30 cm, Be is the best choice. Beyond 30 cm, Al is good, and beyond 75 cm even Fe is satisfactory from the standpoint of shrapnel production.

Objects that must be close to the target (such as components of the target support or diagnostics systems) usually subtend only a small portion of the solid angle. Most of the solid angle is taken up by the first wall. Two possible methods to avoid ablation of the first wall are to protect it by introducing a background gas into the target chamber or to move the wall far enough away that it suffers no ablation. Figure 7-5 shows the minimum radius we calculate for no vaporization of the first wall for a variety of wall materials and gases.

Two factors determine the vacuum required in the target chamber: (1) The maximum pressure under which the laser beams can still propagate to the target is 0.01 to 0.1 Torr, depending on the gas used. (2) The vacuum required by some diagnostics to perform their tasks, particularly those measuring the soft x-ray spectra, is 10^{-4} to 10^{-5} Torr. (Note that

Fig. 7-5. Minimum wall radius to prevent ablation vs pressure of nitrogen or xenon chamber gas.

some of these diagnostics are also very sensitive to the atomic number of the gas in the chamber.) Because of the above points and the fact that vacuum chambers larger than 10 m in radius are very expensive, it appears that avoiding first wall ablation altogether is impractical.

Accepting first-wall erosion as probable led us to consider placing the first wall as close as possible to the target and fabricating it out of an erodible, crushable, and easily replaced material. We are examining a carbon/carbon fiber material that may be able to operate 1 to 2 m from the target. About 1 kg of carbon would vaporize from such a wall during each high-yield experiment. The major issue with this material is determining where the vaporized material will recondense. Condensation back onto the first wall itself is more probable with very porous and granular materials.

In the next article, "Neutronics Issues," we discuss the details of issues related to the high-energy neutrons emitted by the target. It is clearly important to moderate the neutrons before they strike load-bearing structures made of materials such as Al because the dose rate from Al in the time scales of interest (a few hours after the experiment) are governed by threshold reactions that can be eliminated by moderating the neutrons. Also, these structures should be moved as close to the target as possible because the cost of the neutron shield decreases as the radius shrinks and because the cost of the pressure vessel is independent of radius until the internal shield displaces a significant fraction of the volume.

Containing the pressure generated in each high-yield experiment is important but perhaps not as difficult as first imagined. One thousand megajoules of thermonuclear energy is the same as that contained in one-quarter ton of high explosive, but, of course, the time scale and form are very different. In ICF, the neutrons, gamma rays, and hard x rays do not contribute to the pressure on walls further than 1 m from the target. In fact, only about one-fifth (~ 50 -kg high-explosive equivalent) of the total yield contributes to the pressure; this energy is associated with low mass, high temperature, and very short time scales, which would imply that a structure far smaller than that required to contain 50 kg of high explosive may suffice. The High Explosive Applications Facility (HEAF) now under construction at LLNL is designed to contain up to 10 kg of conventional high explosives. The pressure vessel needed for the ICF experiments should be only a few times more massive than HEAF.

Using the facts discussed above, we have identified the elements of a self-consistent experimental chamber. Figure 7-6 shows the major elements of this 3- to 5-m-radius structure.

The Laser/Target Chamber Interface.

On their path to the target, the laser beams pass through target chamber windows and are directed toward the target. The final optic may be either a window/

lens or a mirror. These windows or mirrors (if the the windows are located on the sides of the beam tubes) are directly exposed to the output of the target. The solid angle subtended by the beam lines at the target is based on the following considerations: the area of glass needed to allow the laser beams to pass or reflect without damaging the optical elements, and the distance that must be left between the glass and the target to avoid optical damage from neutron, soft x-ray, and debris energy. The design of the final transport geometry must also account for the maximum beam intensity allowed at the target.

Sol-gel antireflective coatings were invented to increase the damage threshold of transmitting optical elements on Nova. We have expanded our work on high-damage-threshold coatings and have now made a highly reflective coating for mirrors with damage thresholds of more than 20 J/cm^2 at the pulse lengths of interest. This highly reflective coating compares favorably with the damage thresholds for the antireflective coatings. Thus, a 10-MJ laser will require the same amount ($\sim 50 \text{ m}^2$) of glass for the final optics regardless of the type of coating.

We have conducted tests at the Rotating Target Neutron Source (RTNS) facility at LLNL to measure the neutron damage threshold for various optical materials. We found that, although the damage threshold for transmission in most glasses is $\sim 5 \times 10^{12} \text{ n/cm}^2$, the thresholds for fused silica and potassium dihydrogen phosphate (KDP) are $> 1.7 \times 10^{15} \text{ n/cm}^2$. This suggests that a lens at 10 m would survive more than 60 high-yield experiments. We also found that the neutron effects will anneal the glass at 400°C , allowing the lens to be reused.

The optical surface will not survive unimpeded x rays and debris at 10 m (recall that a few microns of material vaporizes at this distance). However, 50 m^2 of glass at 10 m subtends only about 4% of the solid angle. This small area could be protected by gas, which would have to be injected near the lens (or mirror) just before the experiment. The intense laser beams would then be sent to the target before the gas could

Fig. 7-6. The elements of a self-consistent target chamber design.

travel close enough to the target to cause breakdown in the gas. Our calculations indicate that only a few Torr·meters of gas are needed. Because the pressure vessel wall may be at a radius of 3 to 5 m and the lens (or mirror) may be at 10 m, the beam pipes will stick out from the surface like porcupine quills, leaving more than enough space (an additional 5 m) for a gas puff protection system.

In 1987, we also considered the implications of the light intensity desired at the target. Our data indicate that conversion of light into x rays can be efficient if the incident light intensity is less than $\sim 2 \times 10^{14}$ W/cm², but that the conversion efficiency drops off quickly above that value. (We will explore ways to raise the conversion efficiency in 1988.) If the final lens has a 1-m² aperture and if all the light is focused on the target, intensities greater than 2×10^{15} W/cm² result, which are far too high. Thus, we want to focus only about 0.1 m² per beamlet and allow some overlap. This implies a final focusing element similar to the eye cluster of a bee. Because of the space needed between elements, the total solid angle subtended by the beam lines may increase to about 12%.

Conclusions. From the general considerations discussed here, we know that designing an experimental area to handle up to 1000 MJ of yield will be challenging, but not impossible. In fact, we have identified several alternative approaches to solve each issue. For several of these issues, it is clear that further data are needed to determine the best design. For example, the vaporization/condensation model needs to be thoroughly tested; the performance of alternative first-wall materials must be examined; any beam quality degradation upon propagating through turbulent gases needs to be measured; and the threshold for damage to fused silica by x rays needs to be determined. The results from these studies should make possible the design of a useful experimental area.

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Neutronics Issues

In this article, we discuss radiation hazard issues related to the safe operation of a Laboratory Microfusion Facility (LMF). The sources of these hazards are many, and the implications of their containment are important in terms of cost and facility capabilities. We also suggest how the performance goals for the facility can be analyzed, in light of established federal safety guidelines, to develop shielding and other requirements. We examine the containment of the neutron yield by both moderating and absorbing the neutrons, and we describe which materials at what locations might best accomplish this containment. We also analyze the implications of variations in chamber radius, and we consider interior radiation hazards from target debris, tritium, and from target-support and diagnostic materials. Finally, we analyze a specific LMF design to further illustrate neutronics issues such as the effects of prompt gamma and neutron doses on electronics, the need to compensate for direct-shine neutrons through beam ports, and the need to use low-Z materials to shield against decay gamma rays.

Radiation Safety Standards. The U.S. Department of Energy radiation design guidelines limit an employee's dose to 500 mrem/yr, but allow up to 5-rem/yr total dose from both internal and external hazards.⁷ To meet these standards, a facility must be designed to give workers not more than one-tenth of their annual dose limit. The *International Commission on Radiological Protection Publication #30* (Ref. 8) is the guide for determining the dose limits for people ingesting or inhaling hazardous nuclides. Internal hazards consist of airborne or waterborne nuclides that are ingested or inhaled by workers or the public. External hazards consist of decay gamma radiation emitted from activated materials in the proximity of employees working in an LMF. No member of the public can receive more than 25 mrem/yr during the routine operations of an LMF or more than 25-rem total lifetime dose from a design-base accident.⁷ Disposal of wastes must be done in accordance with 10 CFR 61 (Ref. 9). Shallow

land disposal [which is possible when the specific activity (Ci/m^3) of the waste is less than the value tabulated in the *CFR* for each nuclide] is desirable for economic and political reasons.

Implications of LMF Operations. The experimental plan for an LMF should include a projection of the shot rate over the lifetime of the facility, taking into consideration the increase in the average yield of each year's shots. This is critical information for the design of shielding that will adequately moderate or absorb neutrons. This projection should be optimistic but not so inflated that it requires purchasing large quantities of shielding that will never actually be needed over the facility's lifetime. Likewise, significantly underestimating the number of design-yield shots might result in unacceptably long delays for "cool down," which would reduce the number of experiments that could be conducted and, therefore, would hamper experimental progress.

Table 7-6. Hypothetical 10-yr LMF experimental schedule at five different energy levels. The total energy released is 1305 GJ, and the total number of neutrons released is 4.6×10^{23} .

Year	Number of shots at five energy levels					Total shots
	1000 MJ	500 MJ	100 MJ	10 MJ	<10 MJ	
1	0	0	0	0	300	300
2	5	20	25	50	200	300
3	10	25	40	75	150	300
4	35	50	65	75	75	300
5	100	75	50	25	50	300
6	125	100	15	10	50	300
7	150	100	15	10	25	300
8	175	75	15	10	25	300
9	200	50	15	10	25	300
10	200	50	15	10	25	300
	1000	545	255	275	925	3000

Fig. 7-7. Comparison of exterior dose rates from two 2.2-m-radius Al chambers: one irradiated over 15 min and the other over 5.5 yr with 1000 evenly spaced 1000-MJ shots.

To meet the performance goals of the facility (the number of experiments at a particular yield over a given time period), we must contain or moderate induced radiation in the EA to allow timely personnel access in accordance with the facility's operational plan without exceeding radiation exposure limits. Functions such as chamber cleanup, postshot data collection, and diagnostic and target preparation for the next shot must be allotted enough time for the task to be completed without contributing significantly to each employee's annual allowable dose. This implies that certain trade-offs exist among thickness, type, and location of shields, length of cool-down periods, and the number of personnel exposed to ionizing radiation.

Table 7-6 represents a hypothetical LMF shot schedule for a 10-year facility lifetime. (This is an ad hoc schedule, and any discrepancies between it and the DOE Subcommittee report in the preceding article will be resolved prior to facility design.) This schedule takes into account radiological safety standards and implies a possible reentry time and dose rate. The following simple assumptions led us to a maximum "acceptable" dose-rate estimate of 2 mrem/hr:

- The time between shots for setup and takedown is 4 hr.
- Four 4-person teams are used (one team at a time).
- The plant is operated for 200 days per year.
- A 12-hr cool-down period is allowed after each shot.

This maximum dose rate (the condition for reentry of unshielded workers into the facility) is 200 times greater than the background radiation, but is less than the allowable dose rate for a 40-hr work week in the nuclear power industry (2.5 mrem/hr).

The operation of an LMF will differ from that of a reactor or accelerator in the lifetime fluence, the neutron-radiation spectrum, and the resulting activation. Figure 7-7 compares the exterior dose rates from two 2.2-m-radius chambers fabricated from aluminum (a likely material of choice for an LMF). Each was exposed to 1000 high-yield shots of 1000-MJ each, evenly spaced over 15 min

in one case and over 5.5 yr in the other case. For both cases, the chamber wall was 12 cm thick, and no shielding was in place. The shielding requirements for the first case will far exceed those for the second case because radioactive nuclides build up in the Al. These calculations were performed (as are all activation calculations in this article) with the code TART¹⁰ to determine the neutron spectrum and fluence in a material, and with the code FORIG¹¹ to determine the activation products and the dose rates. The code ORLIB¹² was used to compute average reaction cross sections over the fluences and spectra determined by TART.

LMF Shielding Issues. It is clear that the short (<1 hr) and long (>100 yr) half-life nuclides will not pose difficulties that are as great as those from the medium half-life nuclides. Short half-life nuclides will decay rapidly between experiments and, more importantly, will not be a source of secondary activation products (i.e., successive neutron capture in the first activation product) that might pose an additional disposal problem or contribute significantly to the dose rate as their inventory increases. Long half-life nuclides probably will not be produced in large enough quantities to pose a disposal problem, and, furthermore, they do not decay rapidly enough to make a significant contribution to the external dose rate. Medium half-life nuclides decay fast enough to dominate the external dose rate but slowly enough to begin to accumulate over time when the time between shots is short (<1 day). In the cases shown in Fig. 7-7, 900 times more activated ²⁴Na was produced at the end of the shorter irradiation period than at the end of the longer period. (The ²⁴Na has a half-life of 15 hr and is the principal source of the activated-Al dose.)

The containment of the neutron yield and the prevention of activation occurs through moderation and absorption in the shielding. Moderating neutrons to energies below the threshold of the medium half-life reactions (if they have a threshold) suppresses neutron production. The remaining neutrons must be absorbed somewhere in the facility. Materials that perform this function close to the source more effectively deter the

Table 7-7. Shield thicknesses and costs for a 10-m-radius Al chamber that is 10 cm thick that achieves a dose rate of 2 mrem/hr, 12 hr after a 1000-MJ shot.¹³

Material	Thickness (cm)	Cost (M\$) ^a
Boron carbide	85	400
Lithium hydride	125	60
Borated paraffin	127	3
Borated water	130	400

^a M\$ is millions of dollars.

activation of large support structures external to the EA, where simple moderation cannot prevent activation.

Aluminum has predominately short-lived activation products. Examples of the thicknesses of various shielding materials required to keep Al activation low enough to allow a 2 mrem/hr dose rate, 12 hr after a 1000-MJ shot, are listed in Table 7-7. The costs indicate that water may be a good choice even though similar thicknesses of lithium hydride, borated paraffin, and borated water are required (about one-third less thickness is required for boron carbide). However, other considerations concerning the use of water are: leaks and the resultant shield loss and cleanup, shock wave transmission to the Al, and oxygen and hydrogen gas production as the water disassociates under irradiation.

The shield costs, listed in Table 7-7, are for high purity materials and do not reflect the container or restraint used for each material. The relative purity necessary for all materials used to contain the neutron yield will depend on the total fluence of neutrons and on the shot interval times each material will see over the facility's lifetime. For material receiving large lifetime fluences, we found that the utility of low-Z material for inhibiting activation is compromised if the amount and type of activated impurities (usually higher-Z material) can significantly add to the dose or other radiological hazards. Figure 7-8 illustrates this point by comparing the dose rates from a 2.2-m-radius, 12-cm-thick Al 5083 chamber (with impurities included) after one 1000-MJ shot and after 1000 such shots spaced every two days.

The activated impurities in Al 5083 contribute only 1% of the total dose 2 days after the one-thousandth shot but

Fig. 7-8. After 1000 high-yield shots (7 days apart), activated impurities in Al 5083 accumulate to such an extent that the external dose rate is dominated by their decay, rather than by the Al activation—despite the much larger mass of the Al.

all of the dose 12 days after this shot. This evaluation of Al 5083 indicates that it is probably pure enough to be used in an LMF with the shot schedule shown in Table 7-6, which allows access after 12 hr. However, we must also evaluate the potential impurities of any materials that might be used as LMF shielding (especially those that might be used in large quantities) to determine acceptable impurities for each. Even if relatively impure shielding materials can be used, the values in Table 7-7 will not vary by more than a factor of 4 or 5, and water will still retain its economic advantage.

Radius-Dependent Effects. The material quantities in Table 7-7 represent a large capital cost for the facility. Efforts to decrease the radius of the chamber (thereby reducing cost) affect the facility's shielding capability. Specifically, since the amount of a particular nuclide produced is invariant with chamber radius, the exterior dose rate is increased by $1/r^2$ as the radius is reduced. Therefore, the required thickness of shielding must be increased to maintain the same exterior dose rate. This is further exacerbated when mechanical considerations require a thicker chamber wall as the chamber radius is decreased.

Absorber Placement. The least expensive neutron absorber is boron-10. The absorber should be located as close to the source as feasible because the large quantities of inexpensive, but probably highly activating, structural material used in the spaceframe might be a source of unacceptably high dose rates, which might prevent EA reentry when desired. Although relatively inexpensive, boron is

still more expensive than water or carbon, which are our moderator-material choices. Therefore, it is important to determine where in a chamber/shield design a fixed quantity of natural boron is best utilized to reduce activation in the Al wall and in the supporting (primarily iron) spaceframe.

The base case is an Al chamber with a 2-m-radius wall (10 cm thick). A 20-cm carbon shield is located inside the chamber and a 5-m-radius, 10-cm-thick Fe wall is placed outside the chamber. In these calculations, we placed a fixed quantity of natural boron in five locations: in front of the shield, evenly seeded throughout the shield, behind the shield, behind the aluminum, and in front of the iron. The boron-10 location that resulted in the most absorption and in the least activation in Al and Fe was just in front of the Al chamber wall. Certain LMF designs might require less boron if the boron were placed in a distinct layer before the Al wall (rather than evenly seeded throughout the bulk shield material). This decreased quantity of boron might more than offset the increased cost of installing that layer.

In-Chamber Hazards and Diagnostics.

The target containment area will be an extremely severe environment during an experiment. X rays and debris will inevitably ablate kilograms of material, will stress final optics, and will destroy diagnostics close to the capsule.¹⁴ Postshot cleanup will be hindered by the radiological hazards inside the chamber. One source of ionizing radiation will be any high-Z material that becomes activated during a shot. High-Z material might be used for local shielding of diagnostics against prompt gamma rays and might be included in the design of the diagnostics. High-Z material might also be used to provide alloying strength to in-chamber structural materials. The probability that the chamber's structural material will include trace high-Z impurities that could affect its activation must be explored. We must also consider the effects of this high-Z material if it condenses on the first wall or is evacuated from the chamber in a gas or aerosol form.

Among the candidates for high-Z materials are tantalum, lead, gold, and tungsten. We examined the relative hazard

each of these may present as an internal or an external hazard. Figure 7-9 compares the relative inhalation hazards of these materials to a reasonable estimate of the hazards of unburned tritium. These values are based on the assumption that the entire inventory of each material is available for inhalation. The absolute value of the volume of air needed to reduce the concentration of the material to nonhazardous levels is unimportant, but it is important that one material, Pb, would not contribute significantly to the overall hazard present from tritium after a shot. (The hazards shown in Fig. 7-9 range over 4 orders of magnitude.)

The first wall will contribute to the external dose if activated high-Z material condenses on it. For an inner radius of 1.6 m and at 12 hr after the shot, the first wall contribution to the dose could be $<1 \mu\text{rem/hr}$ for gold, $10 \mu\text{rem/hr}$ for lead, or nearly 2 mrem/hr for tungsten. The dose rate inside the chamber from the activated Al wall (also 12 hr after the shot) is 1 mrem/hr ; therefore, the high-Z material radiation hazard may be insignificant, depending on the choice of material and on the allowable dose rate in the chamber at a specific time after a shot.

Certainly diagnostics located close to the capsule must be built inexpensively since they will be sacrificed on each shot. They should be constructed from pure nonactivating materials, such as plastics, rather than from stainless steel.

Structural Wall Materials. Our discussion of walls thus far has been limited to Al. Other feasible candidates for this function include Kevlar and carbon-carbon fiber composites. Although the construction of a 2.2-m chamber from such materials would be quite expensive, the tremendous reduction in induced activation in comparison to Al might prove worth the cost because the shielding requirements would be reduced and the EA could be entered earlier, resulting in more experiments in less time. Figure 7-10 compares the dose rates from Al 5083 and Kevlar after one 1000-MJ shot at a 2.2-m-radius, 12-cm-thick chamber wall.

Carbon composite, which is more expensive than either Al or Kevlar, would activate to an even lesser degree. For the operational plan in Table 7-6, it would appear both carbon and Kevlar (depending

Fig. 7-9. Comparison of the respective volumes of air required to render activated high-Z materials or unburned tritium inert (all sources of activation are in aerosol form). Lead does not appear to contribute significantly to the existing hazard.

Fig. 7-10. Comparison of dose rates from Kevlar and Al first walls after one 1000-MJ shot.

on their purity) could be used without shielding. However, the increased fluence and average neutron energy incident on the support structures because of the shield's absence would have to be compensated for in ways that might reduce some of the composite's advantages (e.g., placing neutron shielding in front of the structural supports; at a large radius, this shielding would be quite massive). In addition, Al would likely retain its strength after the integrated lifetime fluence, but Kevlar might not, and carbon might be prone to unacceptable swelling.

Use of the Nova Target Chamber. In the next article ("Using the Nova Target Chamber for High-Yield Targets"), we examine the option of using the existing Nova target chamber for an LMF. Our investigation of the shielding requirements for this design revealed a characteristic

that will be true of any small-radius LMF chamber made of metal—insufficient space inside the chamber because of the mechanical limitations that result from providing enough shielding thickness to restrict the Al activation to an acceptable level. Therefore, a gamma shield must be added outside the Al wall to attenuate the decay gamma rays and allow access to the target chamber in the operationally required time. In this design, 60 cm of carbon must be placed in front of the Al wall inside the chamber, and another 70 cm is required outside the chamber to perform the attenuation function. Even though high-*Z* materials are usually employed to stop gamma rays, in this situation, such materials would also be activated by neutrons leaking through the shield and by backscattered neutrons that shine through the beam ports. Therefore, although only 15 cm of lead would be needed to provide the same attenuation as carbon, we chose to use carbon because the lead would add more mass and would also present another activation concern.

The effect of having 20 holes for beam lines in an EA chamber can be substantial. With an open solid angle of 20%, 14-MeV neutrons escape outside the EA. The final optic assemblies and/or vacuum vessel will scatter these neutrons and permit them to activate the first wall and support structures. Our selection of carbon as an exterior gamma-shield to the Al chamber mitigates this problem. The neutrons incident on the back side of the chamber (the fluence is ~ 15 times lower here than in front) have lower average energy (because of at least one scatter) and encounter an effective moderator thickness greater than that in front of the Al (> 70 cm). Therefore, in this case, the sandwiching of the Al solves the direct-shine neutron problem for first-wall activation.

Extensive electronics may be installed just outside this last carbon shield to support diagnostic systems. Neutrons and prompt gammas may provide a combined dose in this region of 1000 rad (Si), 1% neutron and 99% gamma. The electronic equipment might require hardening to

this environment to provide high confidence in our ability to collect data.

Conclusions. The first-wall shielding in an LMF should be composed of either water (because of its low cost and good performance as a moderator and as an absorber) or of carbon (because of its good resistance to x rays and good performance as a moderator). The first wall itself could be fabricated from Al and could still be shielded well enough to allow a cost-effective use of an LMF; however, the use of Kevlar or carbon composite for the first wall should also be considered.

The placement of the neutron absorber is critical to ensure its efficient performance (most neutrons absorbed per unit mass) and to minimize overall activation. Small-radius metal chambers will need exterior shielding, which may also compensate for direct-shine neutrons that leak from the beam ports.

The radiological hazards of the interior chamber will be dominated by the dose rate from the metal wall, not by the activation of materials (such as diagnostics or high-*Z* materials) inside the chamber. The LMF designers must consider radiation effects on electronics and mechanical degradation of materials after irradiation, both of which will be significant with the experimental realization of frequent high-yield shots over an extensive period of time.

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Using the Nova Target Chamber for High-Yield Targets

The next ICF facility beyond Nova will illuminate targets with 10 to 12 MJ of energy and will produce yields up to 1000 MJ. In the interest of economy when building this facility, we considered modifying the existing Nova target chamber (which has a 2.2 m inside radius and is fabricated from aluminum) by adding boron-seeded carbon shields inside and outside the chamber (Fig. 7-11). The carbon lining inside the vessel will reduce activation of the chamber wall by

moderating and absorbing the 14-MeV neutrons emanating from the fusion reactions. The carbon coating outside the vessel will protect personnel from Al activation and will allow them to return to the target chamber within a few hours after an experiment.

The target chamber is a convenient means of supporting the carbon shields because the carbon can be placed at a smaller radius with a containing structure. A smaller radius reduces the volume and, therefore, the expense of the carbon. Optical components, diagnostics, and the target positioner will be supported from structures outside the chamber, so they will not impose additional constraints on the target chamber as they do in Nova. Another contrast with Nova is that the laser-beam and diagnostic holes in the modified target chamber will be left open (i.e., the holes will not be covered with glass or any other material).

A vacuum of 10^{-2} Torr will be maintained both inside and outside the high-yield target chamber. Sooy has proposed a separate target chamber room to be used as a vacuum barrier to the atmosphere.¹⁵ The vacuum in both the target chamber and the target-chamber room will allow high-pressure gases produced inside the target chamber to jet out through the open laser-beam and diagnostic ports into the target-chamber room. These jets of gas will spread dramatically once outside the target chamber in a manner similar to the vapor from a rocket jetting into the vacuum of space. This dramatic spreading (initially, an $\sim 85^\circ$ half angle) reduces the amount of material striking the optics and diagnostics, which are located in a direct line-of-sight from the target.

Carbon Shielding. We propose adding three layers of carbon shielding to the existing target chamber: the outer layer will coat the target chamber, and the middle and inner layers will line the inside of the target chamber.

The inner shield has an inner radius of 1.6 m, and it is made of boron-seeded carbon-carbon fiber composite blocks to withstand the high-temperature,

Fig. 7-11. High-gain test facility concept using the Nova target chamber modified by the addition of carbon shields.

high-thermal-shock environment. The blocks are shaped to form a self-supporting bridge or roof, similar to a Roman stone bridge or an Eskimo igloo. The blocks have an essentially zero coefficient of expansion in the circumferential direction. This lack of expansion reduces macroscopic thermal stress and thermal shock problems; however, some microscopic thermal stress will still exist between the carbon matrix and the carbon fibers.

The middle shield is made of boron-seeded carbon foam and/or granular carbon material, which dissipate shock waves resulting from the x-ray and debris energy deposited at the inner surface of the shield. With this shield, none of these shock waves will reach the target chamber wall.

The outer shield is made of inexpensive bulk carbon material, again seeded with boron, because it experiences near-zero stress and does not require much strength. All the shield material will have conical holes matching those in the target chamber so that the laser beams can illuminate the target.

The advantages of using boron-seeded carbon for each shield are that it moderates and absorbs 14-MeV neutrons, has good high-temperature properties (which

are necessary to avoid excessive ablation at the inner surface of the shields), and does not activate significantly itself. Tobin in "Neutronics Issues," earlier in this section and in Ref. 16, calculated the activation for shields having a variety of dimensions and levels of boron seeding (wt%). Even with shielding, activation of the Al target-chamber wall may result in excessive delays before personnel can reenter the high-yield facility. If these delays make the use of aluminum unacceptable, the methodology outlined here can be applied to other target chamber designs fabricated from low-activation, carbon-carbon fiber composites, rather than from aluminum.

Area Available for Laser Beam Penetration. The existing Nova target chamber has 20 entrance holes specifically manufactured for laser beam penetration, each hole is 0.91 m diam. The projected area of these 20 holes at 10 m radius is 270 m². In addition, two 1-m-diam holes are located on the horizontal centerline. The combined projected area at a 10 m radius is 310 m². For a total laser-beam energy of 12 MJ and a maximum fluence of 12 J/cm² with 0.35- μ m laser light, the required area (the quotient) is 100 m² (Refs. 15 and 17). Hence, if the optics are located at 10 m radius, we have at least 2.7 to 3.1 times the required area for laser beam penetration.

Target Chamber Wall Stresses. The pressure produced inside the target chamber and the momentum imparted to the inner shield cause stress in the target-chamber wall. The highest stress is caused by the internal pressure.

Pressure Stresses. Although specific hydrodynamic calculations are required for an exact solution, we used Glenn's analysis¹⁸ to estimate the pressure produced by a nuclear explosion inside a spherical chamber with surface holes. For a 1000-MJ-yield shot, Orth found that ~300 MJ of x-ray and debris energy reside in the ablated carbon.¹⁹ We estimate the ablated mass m as

$$m = \frac{W}{h_s} = \frac{300 \text{ MJ}}{60 \text{ MJ/kg}} = 5 \text{ kg}, \quad (1)$$

where W is the x-ray and debris energy in the ablated mass, and h_s is the

sublimation energy for monatomic carbon.²⁰ (X rays and debris arrive at different times at surfaces some distance from the target. The debris probably does not make a major contribution to ablation; however, the x-ray and debris energy are considered together for our first-order "worst-case" analysis.) Next, we calculated the period²¹ of the Nova Al target chamber T as

$$T = 2\pi R_c \left[\frac{2E}{\rho(1-\nu)} \right]^{-0.5}, \quad (2)$$

where R_c is the radius of the target chamber, E is Young's modulus, ρ is density, and ν is Poisson's ratio [for Al, $E = 69 \text{ GPa}$, $\rho = 2700 \text{ kg/m}^3$, and $\nu = 0.33$] (Ref. 22). Nova has an R_c of 2.2 m, which results in a period T of 1.6 ms.

We then used results for a chamber initially at vacuum (Glenn's Fig. 1 in Ref. 18) for various values of the chamber's radius R_c and the shield's inner radius R_0 . Glenn's results for a hole fraction of the chamber surface area were zero and one-half. We interpolated between these results because Nova has a hole fraction of about one-quarter. Next, we averaged the pressure P over a time equal to the chamber period T . Finally, we calculated the wall stress caused by the pressure σ_p as

$$\sigma_p = \frac{PR_c}{t}, \quad (3)$$

where t is the thickness of the target chamber wall (0.12 m for Nova). Note that this equation gives a value of σ_p that is twice as high as that for a statically loaded spherical vessel because the pressure loading is applied over a short time and the peak dynamic stress is double that of static conditions.²³

We used an allowable strength for Al of 72 MPa and calculated the required wall thickness for the chamber.²⁴ Note that fatigue is not important because the number of high yield targets expected to be tested in the next laser facility is only about 1000—less by at least an order of magnitude from conditions where fatigue life becomes a limiting factor. Figure 7-12 shows variations in wall thickness as a function of R_c and R_0 that satisfy the ASME Pressure Vessel Code requirements.²⁴

For a given value of R_c , the wall thickness decreases as R_0 increases (and the thickness of the inner plus middle shields $R_c - R_0$, decreases) because there is more volume into which the ablated material can expand. The selection of R_0 is based on the required shielding thickness and the required wall thickness. For the Nova target chamber ($R_c = 2.2$ and $t = 0.12$ m), the minimum value of R_0 is 1.6 m.

Tobin calculated variations in the radiation dose with different carbon shield thicknesses.¹⁶ He found that, for carbon seeded with 10-wt% boron and with $R_0 = 1.6$ m, the dose 1 hr after a 1000-MJ shot is 60 mSv/hr if the exterior of the target chamber is uncoated, and 25 μ Sv/hr if the exterior of the chamber is coated with 700 mm of carbon.

In Fig. 7-13, we plot the combined weight of the inner and middle carbon shields and also the weight of the Al target chamber for various values of R_c and for various values of the combined thickness of the inner and middle shields. Note that the target chamber weight and, hence, the cost increases as R_c decreases, but that the combined weight of the inner and middle carbon shields decreases more rapidly. (Our carbon material is also more expensive than Al.) Consequently, for a given thickness of shielding, the cost of the complete target chamber including shielding decreases as R_c is reduced. As a result, it is desirable to use a target chamber smaller than Nova's; however, as the chamber becomes smaller, stresses in the inner carbon shield increase. Before proposing a smaller target chamber or even using the Nova target chamber, specific hydrodynamic calculations and experiments are needed to determine a minimum size at which the carbon will not comminute.

Ablation Generated Stresses. Momentum generated when carbon is ablated from the inner surface of the shield is transmitted to the target chamber and produces stress in the wall σ_m equal to

$$\sigma_m = \frac{M}{t} \left[\frac{E}{2\rho(1-\nu)} \right]^{0.5}, \quad (4)$$

where M is the momentum generated per unit area (Refs. 25 and 26). Orth

▲ Fig. 7-12. Relation between the inner radius of the carbon shield, the radius of the target chamber, and wall thickness. The ASME allowable stress for Al is 72 MPa.

Fig. 7-13. Variation of inner plus middle shield and target chamber weights with target chamber radius and shield thickness.

calculated the momentum generated if a 1000-MJ fusion target exploded inside target chambers of various radii when the inside surface was carbon, aluminum, and solid nitrogen.¹⁹ He found that the momentum per unit area reaching the Nova target chamber was $300 \text{ kg}(\text{m} \cdot \text{s})^{-1}$,

which resulted in a value of σ_m equal to 11 MPa, or $\sim 15\%$ of the magnitude of 73 MPa for σ_p . Therefore, we conclude that pressure-generated stresses dominate our target chamber design.

The momentum is generated mainly by ablation of the inner shield and not from the explosion of a high-yield target at the center of the target chamber. Hence, application of Glenn's momentum results (Fig. 4 in Ref. 18) is not appropriate for our conditions and in fact would give a magnitude that was too high for the amount of momentum generated.

Vapor Spreading. Using Glenn's Fig. 1 from Ref. 18, we calculated that the material vaporized from a shield with an inside radius of 1.6 m produces a pressure P inside the Nova target chamber of ~ 4 MPa. This vapor jets out the target chamber holes at supersonic speed. Because the exterior of the target chamber is also in vacuum, dramatic spreading of the vapor occurs. We calculated the initial spreading by assuming isentropic expansion of a perfect gas with Prandtl-Meyer flow.²⁷ The initial exterior pressure P_∞ is 1.3 Pa. The pressure ratio is related to the external Mach number M_∞ by

$$\frac{P}{P_\infty} = \left[\frac{1 + (\gamma - 1)M_\infty^2}{2} \right]^{\gamma/(\gamma - 1)}, \quad (5)$$

where γ is the ratio of specific heat (5/3 for our monotonic gas). The Prandtl-Meyer expansion half angle ν for $\gamma = 5/3$ is

$$\nu = 2 \tan^{-1} \left(\frac{M_\infty^2 - 1}{4} \right)^{1/2} - \tan^{-1} (M_\infty^2 - 1)^{1/2}. \quad (6)$$

In our case, $M_\infty = 34$ and $\nu = 85^\circ$. The boundary of the vapor would curve as shown schematically in Fig. 7-14, but we must still accurately calculate the specific profile. It is clear, though, that extensive spreading of the jets will occur. Optical components placed at 10 m radius would still need to be protected from debris and from condensation of vapors by either a gas puff and/or a blast shield, but the amount of material directed toward the optics would be reduced substantially because of the jet spreading.

Remaining Issues. Two issues must still be resolved before we decide on the final design of the high-yield target chamber. First, we need to determine the minimum radius for an inner carbon shield that could survive the environment following a 1000-MJ-yield target shot, which is the equivalent of detonating 50 kg of high explosive at the center of the Nova target chamber (see "Design Issues," earlier in this section for more details). We are concerned that, if the inner radius of the shield is too small, thermal-shock loading will cause the carbon material to comminute. In addition, we do not know how carbon composite materials behave under conditions of rapid, nonuniform energy deposition; therefore, hydrodynamic calculations and experiments are needed to ensure that the inner shield can survive the loading conditions. We propose obtaining some carbon-carbon fiber material and conducting laboratory tests that include lining a scaled tank to simulate the Nova target chamber. We could explode a few kilograms of high explosive inside this tank; with data from this test, we could compare experimental results to

Fig. 7-14. Spreading jet pattern of vapor and debris leaving the Nova target chamber after a shot.

hydrodynamic calculations, could determine the equation of state for our material, and could evaluate how well the carbon shield survived. In other experiments, we could illuminate carbon material with lasers, electron beams, or ion beams and could observe how it ablates and transmits shock waves. We could also conduct split Hopkinson-bar tests to determine some of the shock propagation properties of carbon.

Second, although the Nova target chamber already exists, estimates of the cost of the carbon shields are from \$1 million to \$20 million. One way to reduce the amount of carbon shielding is to build a new and possibly smaller target chamber made of low activating material. Fabricating this new chamber may be less expensive than modifying the Nova target chamber by adding carbon shields to protect the aluminum.

Conclusions. By adding carbon shields to the Nova target chamber, we could potentially reduce the cost of the next ICF facility and yet satisfy most if not all of the target chamber objectives. Even if the Nova target chamber is not used, the carbon shield concept is of value because it can be applied to target chambers in general. This concept also has the advantage of protecting the target chamber without producing excessive amounts of activated material itself. Before the final target chamber is designed, we advocate a program of hydrodynamic calculations coupled with experiments to determine the lifetime of the inner shield during high-yield experiments.

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General Studies

Tritium Safety Characteristics of Cascade

We will likely have to demonstrate that future fusion power plants will be inherently safe. In this article, we analyze Cascade, a potential ICF reactor that breeds

tritium but is inherently safe. We underscore this safety of the Cascade design by comparing it with a Chernobyl-like fission reactor in terms of possible dose rates to the public from an accidental release.

In an earlier tritium safety analysis of Cascade, GA Technologies determined doses to the public for several accident scenarios. In detailed and extensive studies,^{28,29} they analyzed various tritium release rates for a fixed quantity of tritium (200 g). We used those results as a basis for this expanded analysis in which:

- We use HOTSPOT,³⁰ an LLNL Hazards Control Group computer code, to determine downwind doses to the public after a tritium release.
- We include more tritium on site by assuming that the power plant and the target-fabrication/fuel-fill facilities are at the same site—this may be the most cost-effective way to build these facilities.
- We examine tritium doses to the public by varying the fraction of T₂ converting to HTO in light of experimental data that suggest this value is far less than 100% (the current assumption in safety analyses) at the plant boundary or at the nearest low-population zone.
- We compare our Cascade results with another HOTSPOT calculation (with an identical release height and atmospheric conditions) of a Chernobyl-like tritium release from a pressurized water reactor (PWR).

Tritium Decay. Tritium decays by β^- emission (a neutron transforming into a proton, electron, and neutrino), which results in ³He. Energy released by this decay is deposited locally, that is, human skin would easily stop it. (This energy is 0.0186 MeV or 7×10^{-19} cal.) Therefore, unlike dental x-rays or airplane flights, tritium does not present an external radiation hazard to humans. Tritium is a hazard if it is ingested or inhaled because, once inside the body, the decay energy is deposited in internal biological tissue. Tritium has a 10-day biological half-life inside the body. The number of tritium atoms decaying per second (the rate the energy is deposited) depends on the characteristic half-life of tritium (the time it takes for half of the tritium to convert to ³He—12.3 yr) and the amount of

tritium present. Activity is the rate of decay and is usually expressed in curies, or 3.7×10^{10} decays per second. Tritium has a specific activity of 9690 Ci/g.

Maximum Permissible Concentration.

Extensive calculations indicate that the maximum permissible concentration (MPC) of HTO per cubic meter of air is 2×10^{-7} Ci. This MPC is set at this amount to prevent someone who continually breathes air containing tritium from receiving 500 mrem in a year. By comparison, radon gas, a radioactive decay product of naturally occurring uranium, is 100 000 times more hazardous to humans than tritium (it has an MPC of 2×10^{-12} Ci/m³) (Ref. 31). Radon currently poses a health hazard because it has seeped up out of the ground and has collected in basements of homes. It is estimated that, in the U.S., up to 13 000 deaths per year from lung cancer can be attributed directly to radon.

Tritium Permeation. We know that hydrogen can permeate the container that holds it. Some hydrogen diffuses through the material and some goes "into solution" in the material. Diffused tritium is a concern because of the biological hazard. Soluble hydrogen, deuterium, and tritium are concerns because they can degrade the properties of some materials (e.g., the embrittlement of steel caused by hydrogen when it is dissolved at >100 parts per million). The permeation of tritium into a material depends on the type, thickness, pressure, and temperature of the material. Tritium permeability values can range from 5.8×10^{-5} mol/m·s·Pa^{1/2} for aluminum to 5.8×10^{-14} mol/m·s·Pa^{1/2} for beryllium. Temperature dependence of these values (for constant pressure) is indicated by an $\exp(-14\,800/T)$ multiplier for Al and $\exp(-2220/T)$ for Be. Glasses have both a linear and exponential dependence on temperature. The increase in permeability between 300 and 750 K (for constant pressure) is almost 10^{12} for Al but is only 10^2 for Be (Ref. 32). Therefore, under severe accident conditions (i.e., fire), the release of tritium can be a serious concern.

Tritium Release Analysis for Cascade. Cascade is a rotating, 1500-MW, ICF reactor that uses a ceramic-granule blanket to breed tritium, to protect the first

wall, and to remove heat. This design eliminates the fire hazard liquid lithium presented in previous reactor designs. Cascade has a 2.5-MCi (260-g) tritium inventory in the blanket at steady state. However, studies show only 1.4 MCi (145 g) would be released in the 10 hr after an accident in which the blanket (LiAlO₂) reached about 1300 K. The remaining tritium would be released over the following 10 yr (Ref. 28). To the 145-g of tritium released over 10 hr, we have added: 90 g of tritium released from the vacuum system (or the equivalent of 4 hr of fuel consumption), and 80 g of tritium released from the fuel storage area that feeds the target fabrication/fill system (or 10% of a single day's fuel needs). We assume that a day's worth of fuel comprises the total on-site fuel storage requirement and a 10% release. Because tritium is stored in so many compartments, we assume that a complete compromise of all tritium is not realistic.^{33,34} The total quantity of tritium released is then 315 g over 10 hr, neglecting the very small amount present in the tritium purification system's distillation columns.

In this analysis, we assume an effective release height of 20 m, a wind speed of 1 m/s, and an atmospheric stability class of Pasquill F. These conditions are intended to represent a worst-case scenario.

A major issue in determining the hazard of an accidental release of a large amount of tritium is how much of it is converted to HTO (from T₂ or HT) and in what time period. This is because T₂ is nearly 25 800 times less hazardous biologically than HTO (Ref. 30). Information from experiments intended to confirm the conversion fractions in environmental conditions indicate that the results are ambiguous. For instance, the Bardolle experiment in France in 1980 (Ref. 35) indicated a conversion of 24% at 1 km. Another French experiment (Ref. 35) conducted in October 1986 seemed to indicate a 0.1% conversion fraction at 1 km. A Canadian experiment, at a 1- to 2-m release height, implied a 0.5% conversion at 400 m (Ref. 35). Reference 36 states that: "Exposure chamber measurements indicate that the dominant mechanism for conversion of a release of HT to atmospheric HTO is oxidation in surface

soils due to microbial action followed by resuspension [to the atmosphere]." Therefore, wind conditions, precipitation, humidity, soil conditions, ground covers, and release height all affect the conversion of HT to HTO.

For a first-order safety analysis of an accidental tritium release from Cascade, we assume a range of values for the conversion fraction. Clearly, it may no longer be acceptable to assume that 100% is converted. The range considered here is 0.01 to 100%. This covers the range of experimental values, but the greatest confidence is at 1% (i.e., most experiments would substantiate that the conversion is not greater than this value). Table 7-8 lists the HOTSPOT calculations for the HTO conversion vs the maximum dose to the population, using the Gaussian plume model for dispersion and equivalent whole-body effective 50-yr doses at 1 km (the location of the peak dose).

Table 7-8 shows that an essentially linear relationship between the conversion percentage and dose exists. The 10 CFR 100 guideline³⁷ for plant siting specifies that the nearest population center (for planning purposes) cannot receive more than 25 rem in a 30-day period subsequent to any credible accident scenario. Even at a 100% conversion, the Cascade reactor is within 10 CFR 100 guidelines, given the previously stated release conditions and assumptions. Furthermore, even if 100% of the stored fuel were released (all 800 g instead of 80 g), a conversion fraction of 33% still appears to meet 10 CFR 100 criteria. However, a value of 1% for HT conversion to HTO may be acceptable (only the "maverick" Bardolle experiment exceeds it). Using this value, Cascade could have an immediately releasable tritium inventory of 3.3×10^8 Ci or 34 kg.

Comparison with a Fission Reactor. We can compare the fuel hazards for an ICF and a fission power plant in similar accident scenarios. Using the same conditions as for the accidental tritium release, we determined the dose for someone 1 km from a Chernobyl-scale accident. Table 7-9 lists the nuclides and inventories that were released from the Chernobyl accident and the quantity of the same nuclide available at discharge

Table 7-8. Total dose (50-yr whole body at 1 km, for a release of 310-g HT at a 20-m height, 1 ms wind speed, and Pasquill F atmospheric conditions.

Conversion to HTO (%)	Maximum dose (rem)
0.01	0.0032
0.1	0.024
0.5	0.115
1	0.229
10	2.28
25	5.7
50	11.4
100	22.8

Table 7-9. Estimated radioactivity released from Chernobyl compared with the inventory in a 1000-MW_e uranium-oxide-fueled reactor and total dose in rem (50-yr whole body) at 1 km.

Nuclide	Radioactivity released ^a (MCI)	PWR ^b (MCI)	PWR inventory (%)	Total dose	
				(rem)	(%)
¹³⁷ Cs	2.4	2.94	82.0	6.3×10^3	4.9
¹³⁶ Cs	0.4	1.66	28.0	2.5×10^2	0.2
¹³⁴ Cs	1.3	6.7	19.0	5.3×10^3	4.1
¹³¹ I	36.0	23.5	150.0	9.5×10^4	74.0
¹³³ I	9.1	—	—	4.6×10^3	3.7
¹⁴¹ Ce	0.23	37.9	0.6	—	—
¹⁴⁴ Ce	0.14	30.2	0.5	4.3×10^3	3.4
¹⁴⁰ Ba	1.0	39.5	2.5	1.3×10^2	0.1
¹⁴⁰ La	1.0	40.9	2.4	3.8×10^2	0.3
⁹⁵ Zr	0.23	37.3	0.6	3.8×10^2	0.3
⁹⁵ Nb	0.23	37.6	0.6	1.3×10^2	0.1
¹³² Te ^c	5.3	—	—	4.1×10^3	3.2
¹⁰³ Ru	0.76	33.2	2.3	6.3×10^2	0.5
¹⁰⁶ Ru	0.16	14.8	1.1	6.5×10^2	5.1
¹³³ Xe	120.0	43.9	273.0	1.3×10^2	0.1
				1.22×10^5	100.0

^a See Ref. 38.

^b See Ref. 39.

^c These nuclides generally are not considered in fuel-handling issues because they have short half-lives and decay away before waste disposal is accomplished.

(3-yr irradiation) for a 1000 MW_e uranium-oxide-fueled reactor.

The released quantities were indirectly determined from environmental measurements in Europe shortly after the initial detection of radioactive material on the clothing of workers entering a nuclear power plant in Sweden. This same potential hazard exists for virtually every nuclear-fission power plant in the world. Fortunately, nearly all of these plants have more stringent safeguards than were in place at Chernobyl; therefore, a similar accident elsewhere may or may not be credible.

In an uranium-oxide-fueled-reactor accident at 1 km (the location of the peak

dose), individuals would receive a 50-yr whole-body dose of 1.22×10^5 rem, if they remained there for the duration of the plume (about 3 days). If the people were to remain at this location only for 10 hr (as in the tritium accident scenario), they would still receive a lethal dose of radiation (about 16 550 rem, assuming a linear release rate).

Conclusion. Recent HT to HTO conversion fractions seem to confirm that tritium accidentally released from the ICF reactor Cascade would not pose a public hazard—that is, for tritium, Cascade may be inherently safe. The hazard, from an accidental release of radioactive material, posed by a fission reactor in a Chernobyl-like accident is nearly four orders of magnitude greater than for Cascade. More research to substantiate an unambiguous set of values for HT to HTO conversion is important and may allow us to develop a publicly acceptable ICF reactor safety analysis.

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Fig. 7-15. A fuel-pellet material cycle.

Target Debris Safety for an ICF Reactor

If an ICF reactor includes a heavy-ion beam driver and uses high-Z direct-drive targets, then large amounts of high-Z material will be injected into the reaction chamber over the lifetime of the reactor. Because high-Z material activates to a greater extent than low-Z material, we were concerned that high-Z material activation from exposure to this high-neutron-fluence environment might be a major issue for ICF reactors. Although not all targets are designed with high-Z materials, we decided to assume that they are in order to evaluate a worst-case scenario. For this analysis, we

- Selected a potential high-Z target material (lead) and calculated its activation products over the lifetime of the reactor.
- Estimated the dose to the local population in a major accident scenario in which some of this material is released.
- Calculated the waste heat from radioactive decay while the material is in storage.

- Calculated the material's shallow burial index at the end of the reactor's life.

From the results of these calculations, we conclude that the waste generated by activated target material does not pose a significant radiological hazard to the public.

Fuel-Pellet Material Cycle. To conserve material, a fuel-pellet material cycle should be used in ICF reactor operations (Fig. 7-15). If we assume that each capsule weighs 0.065 g and is 35 μ m thick with a 3.5 mm radius (Ref. 40), then 0.065 g times a repetition rate of 5 Hz equals

0.325 g/s,
20 g/min,
1200 g/hr,
28.8 kg/day,
201.6 kg/week,
864 kg/month,
7358 kg/yr,
220.8 metric ton/30-yr lifetime.

We also assume a capsule yield of 300 MJ or 1.065×10^{20} neutrons—the same yield as for the Cascade ICF reactor concept.

Although activation products do build up in the tamper material as it is cycled through the target chamber (>1000 times over the facility's life), it does not appear that there is enough accumulated activation to pose a significant risk either to the public or to workers. To protect workers, processes in the cycle (e.g., pellet forming techniques such as casting and vapor deposition) will undoubtedly be completely automated, and remote handling will be employed.

Pellet Material Management. The vacuum system of an ICF reactor may need to remove materials in their atomic state; therefore, the possibility of contaminating reactions occurring between the tamper material, unburned fuel, and ablated blanket material is great in the vacuum system and in the target chamber. Some means to continually purify the recycled lead will be needed. If the melting point of the tamper material is lower than that of the ablated carbon and blanket granules, the tamper material can be removed by simple techniques, and then it can be chemically reduced to the pure original material. When the vapor pressure reaches the ambient pressure, the tamper material will be deposited on available surfaces (e.g., vacuum system walls). This material could be removed from these surfaces and from the blanket materials through an interaction with a strong getter, such as hydrogen sulfide. When hydrogen gas is evolved, a solid tamper-material sulfide results (e.g., lead sulfide). Hydrogen sulfide is poisonous. Non-toxic handling techniques should be developed.

Sometimes a certain small isotopic constituent of the natural tamper material contributes substantially to the overall radiological hazard of the activated material (e.g., ^{204}Pb in lead) (see "Contribution of Lead Isotopes to the Radiological Hazard of Natural Lead," later in this section). Isotope separation through the Atomic Vapor Laser Isotope Separation (AVLIS) process may be available when this ICF reactor is built; this process can be used to remove these small constituents and, thus, alleviate this radiological hazard.

Determining the Hazard of Lead Recycling. We have calculated the relative safety of recycling the lead from which the ICF pellet may be constructed. As Fig. 7-15 shows, we assumed that a certain amount of lead is allowed to "cool" after a shot before being fabricated into a new pellet. Instead of providing extensive shielding throughout the entire fuel pellet cycle, we included this cooling period to allow the short-half-life nuclides to decay behind shielding while they are in storage. The primary hazard to the public during this lead recycling process would arise if a fire were to compromise the "hot" material while it was in storage.

To quantify the hazard to the public from recycling this lead, we calculated the nuclide inventory accumulated over time in the storage area. For this calculation, we assumed that

- The lead from the irradiated fuel pellet would be recycled once each week for 1110 consecutive weeks. (We included a 70% capacity factor over a 30-yr reactor lifetime in this calculation.)

- Each storage-area container would hold a week's worth of pellet-activated lead at any given time. [We calculated the total inventory of nuclides in the container for three separate times: the end of the first week of operation, the end of the first year (37 weeks), and the end of the plant lifetime (30 yr).]

If we know the amount of a given nuclide produced in a shot and the number of shots per second, we can determine the inventory in the tank from

$$N = \frac{P}{\lambda} [1 - \exp(-\lambda t)], \quad (7)$$

where N is the total activity produced (Ci), P is the activity (Ci) produced in the first second, λ is the decay constant for the nuclide, and t is the total production time (Ref. 41). We used the code TART to determine the appropriate neutron energies in the tamper material.¹⁰ We also used two other codes in these calculations: ORLIB (Ref. 12), which we used to produce energy-averaged cross sections for all possible neutron

reactions in the tamper material, and FORIG (Ref. 11), which we used to actually calculate nuclide production and decay.

After determining the activity produced by each nuclide, we calculated the dose to the local population during an accidental release to the environment. We used the code HOTSPOT (Ref. 42) to make a first-order prediction for these local doses. We assumed that all of the nuclides in a gaseous form and at room temperature were released to the environment. In this case, tritium was the only nuclide that fell into this category. We assumed that the other nuclides were released to the environment as an airborne particulate or aerosol at 0.1% of their mass.⁴³ Because each nuclide has a unique residence time in the body, the dose rate is an integrated value over the remaining life of the recipient, which is assumed to be 50 years. During the nuclide residence time, the dose to the body is from the decaying gamma rays, beta particles, or alpha particles.

U.S. Department of Energy standards limit the total 50-year dose to the population to 25 rem for a maximum credible accident (10 CFR 20, Ref. 44). We found that the dose to the public from the accidental release of the fuel pellet material at the end of an ICF reactor's lifetime would be less than 45 mrem, a negligible quantity. This dose was almost entirely from ²⁰³Pb.

The decay heat from the activation products is also of interest. For the nuclide inventory calculated here, we determined that the decay heat was less than 600 W after 30 yr (again solely from ²⁰³Pb), a manageable quantity. The shallow burial index was 5 at the end of life. The fact that this index was entirely from tritium (n, T) reactions indicates that

- The waste could be buried as class A or low-level waste after 28 years.
- The waste could be diluted to qualify for shallow burial by adding about a factor of 5 more inert material.
- The tritium could be separated using distillation, electrolysis, or other methods.

Conclusions. The low values for population dose, waste heat, and burial index indicate that each pellet could be

irradiated more frequently than once a week. This would result in less required volume in the pellet material holding tank, although the same amount of nuclides would be produced. Since the reason for the holding tank is to allow the short-lived nuclides with their intense decay gamma rays to dissipate, the degree to which the time between cycles can be shortened must be influenced by the allowable external dose rate from the unshielded irradiated tamper material. For lead, the dose rate 1 week after irradiation, even after 30 yr of operation, would be less than 10 mrem/hr. At this low value, only minimal shielding would be needed for the target-fabrication, fuel-loading, and pellet-injection processes. Future work will include a specific design of a temporary storage facility with shielding for active nuclides.

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Gain Required at Fixed Heavy-Ion Driver Energies to Achieve a Competitive Cost of Electricity

A simple economic analysis, introduced by Hogan and Meier,⁴⁵ can be used to estimate the fusion gain required for a particular driver energy in order to achieve a pre-established, or goal, cost of electricity. We modified this technique to consider heavy-ion drivers rather than solid state lasers. Specifically, we assumed that the cost of the driver scales as

$$\text{Driver cost} = 0.2 \text{ or } 0.4 E_d^{0.4}, \quad (8)$$

where driver cost is in billions of dollars and the driver energy E_d is in MJ. The value 0.4 for the driver cost coefficient is taken from Ref. 46. The value 0.2 accounts for technological advances that could reduce the cost.

We considered four cases: cases 1 and 2 use 0.2, and cases 3 and 4 use 0.4. We set the driver efficiency at 30%, rather than the 10 or 15% assumed for the solid

Fig. 7-16. Fusion gain vs heavy-ion driver energy to achieve a pre-established cost of electricity ($\text{¢/kW}_e \cdot \text{hr}$). Each case represents the results of varying net power and the driver cost coefficient.

state laser. We set the cost of electricity at 3¢ , 4.5¢ and $6\text{¢/kW}_e \cdot \text{hr}$ and calculated three curves for each of the four cases using these costs (Fig. 7-16). Cases 1 and 3 constrain net power to 1 GW_e , whereas cases 2 and 4 cap net power at 2 GW_e . We used conventional plant parameters for the power-unit cost coefficient (0.35), the power-unit scaling coefficient (0.8), and the multiplier to convert direct to total costs (1.65). (We define "conventional" as using a non-nuclear standard for plant construction.)

At low driver energies, the reactor costs dominate and the cost of electricity is insensitive to variations in driver energy. Conversely, at high driver energies, the driver costs are so much greater than the reactor costs that even infinite gain cannot bring the cost down to the pre-established value.

Table 7-10. Characteristic power plant values for a driver of 10 MJ and a gain of 50.

	Case			
	1	2	3	4
Driver efficiency	0.3	0.3	0.3	0.3
Reactor efficiency (Cascade)	0.54	0.54	0.54	0.54
Blanket multiplication factor	1.11	1.11	1.11	1.11
Fixed fraction of gross power for auxiliary power	0.017	0.017	0.017	0.017
Thermal power (MW)	2124	4250	2124	4250
Driver power (MW)	128	255	128	255
Auxiliary power (MW)	20	39	20	39
Average repetition rate	3.83	7.65	3.83	7.65
Recirculating power fraction (%)	14.7	14.7	14.7	14.7
Direct capital cost of the reactor (M\$)	647	1126	647	1126
Direct capital cost of the driver (M\$)	502	502	1004	1004
Direct capital cost of the target factory (M\$)	100	100	100	100
Tot. 1 direct capital cost (M\$)	1249	1728	1751	2230
Total capital cost (M\$)	2061	2851	2889	3680
Cost of electricity ($\text{¢/kW}_e \cdot \text{hr}$)	3.80	2.63	5.32	3.39

In case 3, we plot the driver energy from 0.1 to 100 MJ [Fig. 7-16(c)]; in the other three cases, we plot the driver energy from 1 to 1000 MJ [Fig. 7-16(a), (b), and (d)]. These graphs show the fusion gain required to achieve the desired cost of electricity for a particular driver energy. The heavy-ion gain estimates must be reviewed to determine the feasibility of a required gain for a particular driver energy.

The cost of electricity used here is derived from a constant-dollar analysis in

which the dollar retains the same purchasing power throughout the period of interest. This cost does not include transmission, storage, or any other costs incurred by a utility in supplying customers with power. Therefore, this electricity cost is not directly comparable to a consumer electric bill. Rather, it is a figure of merit that can be compared with equivalent values derived from similar analyses of future fission and coal plant costs of electricity. The figures of merit for these plants are 3.5¢ and 4.4¢/kW·hr, respectively.^{4,5}

Table 7-10 lists characteristic values for each power plant option, assuming a particular driver energy and gain within the regions of interest. Our results indicate that all but case 3 would give a competitive cost of electricity for a driver of 10 MJ and a gain of 50.

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Fig. 7-17. Contribution of each natural-lead isotope to the radiological inhalation hazard after an irradiation of 3.03×10^{19} n/cm². Plot over time of the cubic meters of air needed to render an aerosol of activated lead inert.

Fig. 7-18. The radiological inhalation hazard for safe inhalation allows a comparison among natural lead and its isotopes for an irradiation of 3.03×10^{19} n/cm².

Contribution of Lead Isotopes to the Radiological Hazard of Natural Lead

Certain high-Z materials may be chosen for use in ICF research or reactor chambers. The presence of high-Z material may require local gamma-ray shielding of diagnostics, may provide alloying strength to in-chamber structural materials, may include trace impurities, and may affect diagnostic design. When an isotope separation process, such as AVLIS, becomes available, isotopic tailoring of high-Z materials may be desirable to minimize radiological hazards after a shot or over a facility's lifetime.

Natural lead, a relatively inexpensive material, may be an attractive high-Z candidate. Some of the isotopes occurring in natural lead contribute more to the overall hazard of irradiated lead than others. In this article, we examine this aspect in detail by examining the specific radiological inhalation hazard of each

Table 7-12. Relative hazards of lead isotopes compared with natural lead. The shallow burial index is 0.003 for activated natural lead, and the radiological inhalation hazard is the same as the chemical inhalation hazard about 24 to 30 hr after a shot.

	^{204}Pb	^{206}Pb	^{207}Pb	^{208}Pb	Natural Pb
Quantity (g)	0.0069	0.1201	0.1107	0.2637	0.5014
Total discharge (Ci/g)	95	6	4.6	0.15	3.85
Biological hazard potential					
At discharge					
(Ci/m ³ air)	1.1×10^7	2.4×10^7	1.7×10^7	2.0×10^5	5.2×10^7
(%)	21	46	33	<1	100
1 hr after discharge					
(Ci/m ³ air)	9.6×10^6	6.4×10^3	2.8×10^3	1.6×10^5	9.8×10^6
(%)	98	<1	<1	2	100
1 day after discharge					
(Ci/m ³ air)	4.5×10^5	5.2×10^3	31	1.3×10^3	4.6×10^5
(%)	99	<1	<1	<1	100
1 week after discharge					
(Ci/m ³ air)	170	4.7×10^3	30	0	4.9×10^3
(%)	4	95	<1	0	100
1 month after discharge					
(Ci/m ³ air)	170	3.4×10^3	20	0	3.6×10^3
(%)	4	95	<1	0	100
1 year after discharge					
(Ci/m ³ air)	140	43	0.15	0	180
(%)	76	23	<1	0	100

isotope of lead, and then we compare the hazard from each isotope with the overall hazard for natural lead. This radiological inhalation hazard is quantified by the amount of air required to render the particular nuclide inert enough to cause less than a 500-mrem dose per year to someone who continually breathes such air.

Natural lead is composed of four isotopes with abundances as shown in Table 7-11. It is useful to determine if lead, which in its natural state is already less hazardous than many high-Z materials, can be made significantly less hazardous by removing one or more minor isotopes. We made five calculations to determine the contribution of each isotope to the total radiological inhalation hazard of natural lead. We used the computer code TART¹⁰ to simulate a lead sample irradiated with a fluence of 3.03×10^{19} n/cm² (which corresponds to 1000 MJ of yield for this example). We made the activation calculations with the codes ORLIB¹² and FORIG.¹¹ The results of these calculations are shown in Table 7-11 and Figs. 7-17 and 7-18.

Figure 7-17 shows the contribution each isotope makes to the radiological inhalation hazard of natural lead. This is essentially a plot of the data in Table 7-12. Figure 7-18 shows the comparative

Fig. 7-19. Radiological hazard potential of natural lead and of lead with the isotope ^{204}Pb removed.

isotope hazards for equal masses of each isotope and for natural lead. This figure illustrates the significantly higher hazard of ^{204}Pb . Figure 7-19 shows the hazard of lead if the isotope ^{204}Pb were removed.

The time of reentry into an ICF research facility chamber (such as the Laboratory Microfusion Facility) will probably be between 1 and 24 hr if necessary at all. These inhalation-hazard results indicate that nearly all of the hazard at that time is due exclusively to the radioactive products of ^{204}Pb , which makes up only 1.4% of natural lead. The specific activity of each isotope after discharge provides further evidence of the much greater hazard of ^{204}Pb

compared with other lead isotopes. This particular isotope has 25 times more specific activity (Ci/g) than natural lead. Not only is it the major short term hazard, but its activation products also appear to dominate a year or more after discharge. (Note, however, that 1 to 7 days after discharge ^{206}Pb activation products dominate.)

Our results indicate that isotopic tailoring of the high-Z material lead could result in a much lower radiation hazard in future ICF facilities if lead is the high-Z material of choice.

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