Exploratory Research and Development FY90

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Introduction

G. L. Struble

Since 1984, when Lawrence Livermore National Laboratory (LLNL) established the Exploratory Research and Development (ER&D) Program [formerly called the Institutional Research and Development (IR&D) Program], its goals have been to fund:

- Innovative science and technology research projects whose high risks are offset by the great potential rewards for success.
- Disciplinary research that will develop innovative solutions to problems in various scientific fields and will provide enabling technologies for the Laboratory.
- Research and development activities that may lead to new programs of national importance.

In general, the ER&D Program supports research projects considered too basic or long-range to be funded by other LLNL programs. This Program is managed for the Laboratory Director by a special assistant who chairs the LLNL's IR&D Review Committee. Membership in the Review Committee comprises senior LLNL scientists, engineers, and managers whose areas of expertise span the range of scientific disciplines pursued at the Laboratory.

The research supported by the Program falls into three categories: Exploratory Research in the Disciplines, Director's Initiatives, and Laboratory-Wide Competition. The first two, Exploratory Research and Director's Initiatives, promote pioneering work in the various scientific disciplines and programmatic areas. Laboratory departments and divisions propose and manage projects in the Exploratory Research category. The Laboratory Director, with the advice of the Review Committee, selects several larger projects to fund as Director's Initiatives. These projects, which are proposed and managed by the responsible associate director, are intended to enhance the scope of existing programs or establish new technical directions and programs for the Laboratory.

To determine the projects funded in the Laboratory-Wide Competition, the members of a select Research Committee of LLNL's scientists and engineers review the proposals submitted. The Committee evaluates projects for their scientific excellence, long-term significance, and potential for obtaining further funding.

For FY90, the total ER&D budget was \$19.509 million: \$7.138 million for Exploratory Research, \$9.119 million for Director's Initiatives, and \$3.252 million for Individual Awards. The Program funded 70 projects covering a wide range of disciplines, such as:

- Using recombinant DNA methods to express functional antibodies in bacterial hosts and, thus, allow genetic manipulation of antibody genes.
- Developing inexpensive micro-optic lenses to couple diode-laser light to solid-laser media.
- Measuring the gamma rays that result from the scattering of neutrons from protons to probe the neutron-proton interaction potential that holds nuclei together.

All FY90 projects are described in detail in this report. Other publications on ER&D projects are included in the Publications List at the back of this report.

The ER&D Program management and the Review Committee frequently review the Program and assess the work to ensure that the multiple goals of the Program are being fulfilled. With current changes in national priorities, the Laboratory finds the flexibility in the ER&D Program especially valuable for funding innovative ideas with high potential to solve problems of national importance. The Review Committee pays special attention to identifying those projects that can provide a large return on the investment for LLNL and for the Department of Energy (DOE), whether measured by the development of important enabling technologies, by new programs, or by the quality and effectiveness of the LLNL's scientific and technical staff. ■

■ Exploratory Research

Exploratory Research and Development FY90

In FY90, the Biomedical and Environmental Research Program received ER&D funding for four Exploratory Research projects and one Director's Initiative.

As part of our first project, we are examining the analytical capabilities of surface-enhanced Raman spectroscopy. The absorption of various organic species on the roughened surface of metallic electrodes can significantly increase the sensitivity of their Raman spectra. We have begun several environmental and medical applications of surface-enhanced Raman spectroscopy and published two papers on the topic.

In our second Exploratory Research project, we are exploring methods of cloning mammalian immunoglobulin genes into bacteria that can be used to produce functional antibodies. This approach provides a biotechnological source of specific antibodies and permits the straightforward modification of the antibody genes, thus allowing us to study how changes in amino-acid sequence affect the structure and function of the antibodies. We have made a good start toward producing dioxin antibodies.

Our third Exploratory Research project involves a related new genetic technology that allows us to insert foreign DNA into mouse embryos, thereby producing transgenic mice. The mice can be used to study how genes function or to produce designer animals with importantly modified genetic properties. In FY90, we began two types of gene transfer in collaboration with the transgenic facility at the University of California, Davis.

The final Exploratory Research project focuses on the partitioning of chemical species across major environmental pathways. By incorporating the property of fugacity into a five-compartment air, plant, and soil model, we are improving our ability to predict the uptake of several important pollutants in range grass and, hence, into the human food chain via foraging cattle.

The Director's Initiative is a two-part project designed to complement the Human Genome Project with new Laboratory capabilities in nonbiological disciplines. In the engineering phase of the project, we are developing robotic applications for highly repeatable production procedures and exploring new methods of capillary electrophoresis to separate and characterize DNA fragments. In the computations phase, we are using signal processing, statistical decision making, and graphics interfaces to reassemble DNA fragments and, ultimately, order a human chromosome. As a result of our efforts, we have made excellent progress in genome research and have developed important new skills.

In FY90, the Biomedical and Environmental Research Program received \$300,000 for Exploratory Research and \$700,073 for Director's Initiatives, which includes \$73,000 for capital equipment. ■

Surface-Enhanced Raman Investigations of Organic Pollutant Mixtures

S. M. Angel and M. L. Myrick

We are studying processes important in measuring and interpreting the SER spectra of mixtures and have explored a drug assay based on SERS for cocaine and methamphetamine.

We have investigated the feasibility of measuring contaminant mixtures and biological molecules using surface-enhanced Raman spectroscopy (SERS). Fleishmann et al. (1974) discovered the surface-enhanced Raman (SER) effect. For certain molecules adsorbed on roughened-metal surfaces, this technique yields Raman spectra with 10^5 to 10^6 enhancement over normal Raman spectroscopy, thus providing high sensitivity. SERS is well suited to detecting and fingerprinting environmental contaminants, such as polychlorinated biphenyls and polynuclear aromatics and biological molecules. However, SERS is a surface phenomenon, and the SER spectra of mixtures are complicated by competitive binding of compounds to the electrode surface.

Our goal was to determine how certain parameters affect the measurement of mixtures. We have investigated the SER spectra of selected contaminants and biomolecules on different SERS substrates, in situ vs ex situ roughening, and chemical and electrode potential effects. To simplify the interpretation of results, we used two-component mixtures of simple molecules. Few other SER studies use mixtures of compounds. Our work provides insight into the chemical nature of the SER effect

and a better understanding of the parameters important in analytical applications of the technique.

In FY89, we found that the electrode potential can be used to selectively enhance certain molecular vibrational bands in the SER spectra of mixtures of 3-methylpyridine and 3-chloropyridine (Angel and Archibald, 1989). In situ preparation (electrode surface roughened by an oxidation/reduction cycle in the presence of an analyte) can generate SER spectra nearly one order of magnitude more intense than ex situ preparation. We also found that the magnitude of the SER signal depends strongly on the amount of mass transfer and rate of mass deposition during an electrode oxidation/reduction cycle.

Because of the current volume of drug testing, fast, sensitive, and accurate measurement techniques are desirable. An assay using SERS would meet such criteria and could be used as a simple qualitative test to minimize the number of samples needed for more sophisticated chromatographic instruments.

Raman spectroscopy has potential as a screening test for abused drugs because the vibrational spectrum is a fingerprint of the molecule. The major problem with Raman signals, however, is their low probability of occurrence. SERS can circumvent this problem by providing greatly enhanced Raman signals. Many different substrates are suitable for surface enhancement of the Raman signal, such as colloids, electrodes, coated microspheres, and fused silica. However, we found that gold colloids are the easiest to use, yield a large SER signal, are commercially available, and can be

used with conventional sampling arrangements. We are exploring a SERS assay using a diode laser and gold colloids for rapid, qualitative chemical testing.

We obtained preliminary results for detecting cocaine and methamphetamine by SERS using gold and silver colloids as substrates (Angel et al., 1990). Dissolved cocaine does not yield large SER signals on gold colloids unless carbonate is added to the solution. The base probably leads to deprotonation of the amine and increases its adsorption to the colloids. This finding emphasizes the importance of chemical effects on SER spectra.

The assay technique we developed is simple. We mixed one part cocaine solution to one part colloid solution and added a few drops of carbonate solution to optimize the signal. The SER spectra showed mainly aromatic ring vibrations. We also demonstrated this procedure with methamphetamine. Future work will focus on optimizing chemical factors, design and construction of a simplified optical-fiber probe, SERS of other drugs, and the use of diode lasers as excitation sources. ■

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Genetically Engineered Antibodies

L. H. Stanker and R. H. Jensen

We are developing recombinant DNA methods for expressing functional antibodies in bacterial hosts. A bacterial expression system will allow genetic manipulation of the antibody genes and help us understand the relations between protein sequence and protein structure and function.

Antibodies are complex molecules formed by the association of two, identical heavy-chain polypeptides and two light-chain polypeptides. Only part of the molecule, the antibody combining site, is directly involved with recognition and binding of specific antigens. Using newly developed strategies for expression of functional antibody molecules in bacteria (Better et al., 1988), we can now substitute any amino acid in the antibody-combining site with a different amino acid. Thus, our aim is to generate a large group of modified antibody molecules to study the effects of amino acid substitutions on antibody function and protein structure. Information at this level of detail will further our understanding of antibody function and, in general terms, help define the relations between amino acid sequence and protein structure and function. Furthermore, refinement of such techniques will make it possible to customize antibodies to precisely fit the needs of specific tasks.

Recombinant monoclonal antibodies (RMAs) are generated by fusing the antibody structural genes to bacterial signal-peptide sequences and then arranging the fused sequences in a dicistronic construct under the control of typical bacterial regulators. A dicistronic construct, such as that

shown in Figure 1, is a cloning vector in which two different genes (a heavy-chain gene fragment and a light-chain gene) are arranged in tandem. Construction of a dicistronic cloning vector requires that both the antibody heavy- and light-chain genes be cloned and sequenced and specific nucleotide changes introduced into the sequences to facilitate vector assembly. In our initial studies, we are using an antidioxin monoclonal antibody (Stanker et al., 1987). We selected this antibody because it differentially binds the 75 isomers of polychlorinated dibenzodioxin and the 135 isomers of dibenzofuran. This large panel of antigens will allow us to detect changes in antibody function following amino acid substitutions in the antibody combining site. The tertiary structure of RMAs also can be studied using a variety of methods, including nuclear magnetic resonance spectroscopy and x-ray crystallography.

We have made progress on several levels. We have generated full-length complementary-DNA (cDNA) clones for both the heavy and light

chains of one of our antidioxin hybridoma. Both heavy- and light-chain clones have been sequenced, and the necessary nucleotides have been modified in both the heavy- and light-chain genes to allow construction of the dicistronic vector. We are now in the final process of assembling the dicistronic construct for expression of the antibody molecule in *Escherichia coli*.

Next, we will evaluate the function of the resulting RMAs on the large panel of antigens available for our antibody. We will then generate a set of modified RMAs, each having a single amino acid substitution in the antibody combining site. We will evaluate the modified RMAs for changes in antibody function and then for changes in antibody structure. ■

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Figure 1. Schematic of a dicistronic construct, or cloning vector, for production of a recombinant antibody. Each region of the ring represents genes that encode the functions identified around the outer surface of the structure.

Construction of Transgenic Mice

R. L. Balhorn and S. G. Grant

We are developing two independent approaches for studying gene expression in transgenic mice: investigations of protamine structure and function, and development of a mouse model for the human GPA somatic segregation assay.

Our goal is to bring to LLNL the latest technology for preparing genetic constructs (genes modified or constructed in the laboratory containing known mutations) and to establish methods for inserting these genes into animals that express them (transgenic mice). The use of transgenic animals offers a new, extremely powerful approach for studying the function of genes and the structure and function of their protein products in vivo. Our two projects provide the means for initiating transgenic studies at LLNL and for testing the feasibility

of generating transgenic mouse lines through an outside facility (University of California, Davis). The Biomedical Sciences Division and LLNL will benefit by gaining one of the most powerful new technologies in molecular biology.

One of our studies involves the production of mice that express genetically altered mouse protamine genes. Protamine is a protein that coats the entire surface of DNA, packaging it inside the nuclei of mammalian sperm. We will use these mice to determine how the proteins produced by altered protamine genes are integrated into sperm chromatin (the complex formed between DNA and proteins), affect the synthesis or metabolism of protamine synthesized by the normal gene, and alter male fertility. In another study, we will insert the human glycoporphin A (GPA) gene into the mouse genome to produce a model system in which we can evaluate the mutagenic, carcinogenic, and clastogenic potentials of various chemicals, treatments, and genetic backgrounds.

Transgenic Mice Expressing Mutant Protamine Genes

We have prepared the first mouse protamine-2 DNA construct and are ready to inject it into mouse embryos. This gene contains two amino-acid mutations within the coding sequence of the protamine-2 gene that will allow protamine molecules produced by this gene to be distinguished from those produced by the normal gene. The mutations should not alter the normal functions of the protein required for packaging DNA. The expression of this gene in mice will be used to examine how protamine molecules become integrated into sperm chromatin and

how they interact with the other protamines present in sperm. The gene will also be used as the starting sequence for constructing additional protamine-2 genes with other mutations that are expected to alter specific biochemical steps essential for chromatin organization and normal sperm maturation.

We obtained plasmids (an autonomously replicating, circular DNA molecule that can be used to move genes into and out of bacteria and other cells) containing the complete mouse protamine-2 gene from Norman Hecht (Tufts University, Medford, Massachusetts) and have synthesized the two oligonucleotides (short, single-stranded DNA molecules) required for mutating lysine 14 and lysine 21 to arginine within the coding region of the protamine-2 gene. Both mutations require only a single base change. Enzymes were used to clip out the region of DNA to be changed, and the synthetic DNA was inserted in its place. (This process of gene mutation is called oligonucleotide-mediated mutagenesis.) Protamines synthesized by the new gene can be distinguished by gel electrophoresis following digestion with the enzyme endoproteinase Lys C. Protamines produced by the normal gene will be clipped into several fragments by the enzyme, but protamines produced from the mutant gene will not.

Once we obtain mice that express this mutant protamine-2 gene, we will construct a protamine-1 gene with similar mutations. Transgenic mice that express these genetically engineered protamine-1 and protamine-2 genes will be used to determine in vivo what role the two protamines play in DNA packaging, gene inactivation, and preparation of the male genome for fertilization.

Figure 1. Expression of human GPA detected by immunofluorescence. This figure shows labeling of transfected CHO cells carrying integrated copies of the pSG5GPA^M construct with the human GPA-specific monoclonal antibody 10F7. Two smaller human erythrocytes are shown for comparison.

Mouse Model of Human GPA Somatic Segregation Assay

We have obtained a new plasmid containing a genetically engineered version of the human GPA cDNA (a DNA sequence copied from the RNA used to synthesize the protein glycophorin) from Minoru Fukuda (Cancer Research Foundation, La Jolla, California). We confirmed the presence of the GPA insert in this plasmid by amplifying the cDNA sequences via the polymerase chain reaction (PCR). Subsequent analysis using restriction enzymes allowed us to determine that the vector carried a cDNA coding for one form of the GPA protein (called the M allele) and that it was properly oriented with respect to the adjoining polylinker region. We used oligonucleotide-mediated mutagenesis to synthesize a cDNA corresponding to the other GPA protein (the N allele).

We inserted the two GPA cDNAs into the expression vectors pSV2cat and pSG5 (two special plasmids that provide additional DNA sequences required for gene expression) and introduced the resulting constructs into a hamster cell line (CHO) along with the pMAM neovector (from Clontech, Inc., Palo Alto, California), which contains a bacterial neomycin-resistance gene. Using fluorescently labeled allele-specific monoclonal antibodies, we confirmed that human GPA protein was expressed on the

surface of these transfected cells (Figure 1). The presence of integrated plasmid DNA was detected by the PCR assay (Figure 2). We excised the DNA fragment containing the human GPA cDNA and viral-regulatory sequences from the pSV2GPAM^M construct and sent it to Steven Hinrichs (University of California, Davis) for injection into one-cell mouse embryos. Several pups have now been born. The presence of integrated human DNA and expression of the human GPA mRNA will be determined by a modified PCR technique, while expression of the human GPA protein on the surface of mouse red blood cells (and perhaps other tissues) will be determined immunologically.

We are attempting to develop an assay in the mouse that is analogous to our in vivo human GPA-based somatic segregation assay (Langlois et al., 1990). If successful, this mouse model will expand our understanding of results in humans. Because the expression of our GPA construct will not be restricted to red blood cells, we will be able to isolate mutant cells from tissues that retain their nucleus and genetic material and that proliferate in culture. We can then begin to define the molecular events in human red-blood-cell precursor cells that contribute to mutant phenotypes. ■

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Figure 2. Detection of stably integrated GPA constructs by whole-cell PCR. Lanes 1 and 6 show size markers; lane 2 contains a control with no template DNA added; lane 5 is a control with DNA from an untransfected parental CHO cell line; lanes 3 and 4 contain DNA from two independent clonal CHO cell lines stably transfected with the pSV2GPAM^M and pSV2GPAN^N constructs, respectively.

Human Exposure to Organic Chemicals through Food

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Initial tests of our multi-compartmental model of air, grass, and soil systems, which predicts chemical concentrations in grass tissue that result from both air and soil contamination, show good agreement with measured concentrations of ubiquitous contaminants.

An important challenge facing DOE facilities, private industry, and public agencies is the identification and quantification of exposure pathways linked to toxic pollutants in air, water, and the ground. Such assessments are critical in determining the health hazards associated with pollutants and

in providing the scientific basis for establishing target cleanup levels. Adequate understanding of the food-chain exposure pathway is, therefore, a key element in determining an optimum strategy for and the cost of remediation. The purpose of our project is to develop and experimentally validate a model for estimating human exposures to toxic organic chemicals through the food pathway. We are focusing on organic chemicals in air and soil and on modeling their transfer through food chains. Development of the model relies on experiments that measure plant bio-transfer factors in a controlled setting and facilitate our understanding of the underlying processes that promote or deter biotransfer.

Plant interactions with air and soil have not been understood well enough to define an accurate method for predicting chemical uptake. Thus, we are developing a five-compartment air/plant/soil model (Figure 1) that splits the plant into its important compartments but remains simple enough to allow for the determination of steady-state transport rates between compartments. The prototype plant for the model is range grass because of its importance in the food chain for beef and dairy cows, which concentrate contaminants in their fat and milk. Our model is a level-III (steady-state) fugacity model (Mackay and Paterson, 1981) that uses the environmental partitioning strategy of the GEOTOX model (McKone and Layton, 1986). We are applying the model to perchloroethylene, hexachlorobenzene, and 2,3,7,8-tetrachlorodibenzo-*p*-dioxin. Test cases reveal that the air, soil, and plant are often not in chemical equilibrium and that plants can alter the distribution of atmospheric and soil contaminants. Initial tests of the model show good agreement with experimental results for ubiquitous environmental contaminants, such as perchloroethylene and trichloroethylene. ■

Figure 1. Diagram of five-compartment model used to quantify partitioning of organic chemicals from the environmental media of soil and air into plant tissues. The grass, in this case range grass, is modeled as having three compartments: cuticle, roots, and leaves. The grass interacts with the two major environmental compartments of air and soil. Inputs to the system can be through air, soil, or both.

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Chemistry and Materials Science

C. Gatrousis

As a discipline, chemistry has been called the “central” science because it deals in a fundamental way with the properties and reactions of all substances. In contrast, materials science is frequently thought of as the “enabling” science because it combines chemistry, physics, and engineering to develop practical applications for the results of our research. The Chemistry and Materials Science Department considers both disciplinary viewpoints when selecting projects for inclusion in the Department’s ER&D program. In FY90, we used ER&D funds to support one large and diverse program in high-transition-temperature superconductivity and two smaller, innovative and creative projects.

The superconductivity program is both strongly interdisciplinary, involving chemistry, physics, engineering, and materials science, and collaborative, including the participation of several universities. The program is experimental and theoretical. Its strength derives in part from the ability to produce very-high-quality single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_7$. Using these crystals, we have been studying magnetic flux pinning in directions parallel and perpendicular to the crystal basal plane. Such information is crucial for the potential use of these superconductors in high magnetic fields.

The theoretical aspect of the program emphasizes the generation of a model Hamiltonian to account for the unusual behavior of electrons in perovskite compounds: the electrons appear to be both itinerant and strongly interacting.

Our first individual project is concerned with the structure and properties of thin films. Films are two-dimensional entities of increasing importance in materials science. We are studying how the mechanical properties of a multilayer depend on the nature of the interface between adjacent layers. The magnitudes of some properties are not those expected from a linear combination of the elements that make up the multilayer. We have also observed in real time, perhaps for the first time, the amorphization of an initially crystalline layer as the element of the other layer diffuses into it.

Our second individual project, now in its third year, explores the theory of condensed matter systems. In FY88, we developed a new formalism, based on first principles, that could calculate the density of electronic states in systems of reduced symmetry, such as a surface or an interface (as opposed to the more conventional case of a bulk solid). The next year, we devised the computer codes necessary to apply the new formalism to systems of practical interest, such as twist and tilt boundaries. This year, we studied electronic structure of the phase stability and mechanical properties of the technically interesting Al-Li alloys, notable for their combination of stiffness with light weight.

In FY90, the Chemistry and Materials Science Department received \$1.6 million in ER&D funding. ■

Thermodynamic and Mechanical Properties of Al-Li Alloys

A. Gonis

We are using improved methods to calculate the equilibrium thermodynamic properties and elastic behavior of the technologically important alloys of aluminum and lithium.

Our study is motivated by the rather strange and technologically useful thermodynamic and mechanical properties of Al-Li alloys, especially alloys with dilute lithium concentrations. Most important among these properties is weight reduction with increased stiffness that results from small additions of lithium (~4%) in aluminum. This phenomenon makes Al-Li alloys prime candidates for use in aircraft and as spacecraft structural material.

The first stage of our study consists of calculating the equilibrium thermodynamic properties of Al-Li alloys across the concentration range in both face-centered cubic (fcc) and body-centered cubic (bcc) phases. The second stage consists of

calculating elastic constants of pure aluminum, pure lithium, and the ordered Al_3Li compound as functions of lattice constant.

The thermodynamic properties of alloys can be studied on the basis of effective cluster interactions, which provide a measure of the energetics of specific clusters (i.e., pairs, triplets, and so forth) of sites in the alloy. We can use these interactions to determine ordering tendencies, stable ordered structures at temperatures of 0 K, and alloy phase diagrams. It is understood that no calculation based purely on electronic structure, thus neglecting lattice entropy, can yield accurate phase diagrams. However, our methods have been shown to provide a reliable guide to understanding alloy phase stability and phase transformations in connection with several alloys of the transition series. Our calculations represent the first time that such methods have been applied to alloys involving simple metals.

We have completed calculations of equilibrium charge densities and lattice constants, energies of mixing, and effective cluster interactions across the concentration range for both fcc- and bcc-based alloys. Calculations of elastic constants in pure aluminum and lithium elemental solids and in Al_3Li compounds as functions of volume are under way. We are also preparing calculations that incorporate the effects of charge transfer in calculating effective cluster interactions.

The calculations on Al-Li alloys revealed several interesting features, many of which are not present in transition-metal alloys. In particular, the rather large lattice mismatch between the lattice constants of pure

aluminum and lithium metals coupled with the extended nature of the electronic states in these materials leads to sizable charge-transfer effects and to cluster interactions that vary rapidly with lattice constant and Fermi energy (Figure 1).

One mechanism that has been suggested to explain precipitation of the Li_2 phase in Al-Li alloys with dilute lithium content is the strain fields that can be set up as a result of the large difference in the atomic sizes of aluminum and lithium atoms. Our calculations show that the effective pair interactions, especially between first near neighbors, are sensitive functions of lattice constant near the concentration of the δ' phase (~25% Li). Figure 1 shows the behavior of the effective pair interactions for the first, second, third, and fourth near neighbors in the fcc-based $\text{Al}_{0.75}\text{Li}_{0.25}$ alloy. Our calculations are consistent with the model according to which precipitation of the δ' phase relieves the strain caused by local fluctuations of the lattice constant throughout the alloy. At the same time, these results cannot be said to confirm the model because other effects, such as charge transfer, may cause precipitation of the δ' phase.

Our FY91 calculations will be aimed at examining the elastic behavior of pure aluminum and lithium elemental solids and of Al-Li ordered alloys as a function of lattice parameter. We will also investigate charge-transfer effects. ■

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Figure 1. Variation of first-, second-, third-, and fourth-neighbor pair interactions with lattice parameter. The vertical line corresponds to the equilibrium value of the lattice constant.

High-Transition-Temperature Superconductivity

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New synthesis methods, elemental substitutions, and theoretical formalisms are helping us to elucidate the structure-property relations of perovskite superconductors that allow these localized molecular systems to exhibit properties usually associated with metals. Control of processing conditions leads to ideal materials suitable for studies of transport phenomena.

Since its inception, the LLNL effort on high-temperature superconductivity (high- T_c) materials has focused on fundamental questions concerning the nature of their synthesis and characterization, experimental and theoretical studies of electronic structure, and the role of magnetism, phonons, plasmons, and polarization. The almost-dominant ionic character of high- T_c materials continues to intrigue us when contrasted with their important metallic behavior, which arises from the doping of these materials, yielding their now well-described high-transition-temperature superconductivity. Our strategy is to develop both a fundamental and practical understanding of the important structure-property relations that lead to superconductivity in high- T_c perovskite materials through chemical manipulation of known compounds. Along with this fundamental approach, we are emphasizing activities that will provide processing solutions for the intrinsic properties that appear to limit the technical application of such materials. Here, we describe

three of our principal FY90 research activities, each demonstrating the important link between the chemistry and physics knowledge necessary to understand and apply high- T_c materials.

Flux-Pinning Studies

The existence of twin boundaries in $R\text{Ba}_2\text{Cu}_3\text{O}_x$ (where R represents the rare-earth elements, except cerium, terbium, and praseodymium) provides a unique opportunity to study the flux-pinning mechanism in layered high-temperature superconductors. This mechanism is important to understand because the ability to sustain high currents depends on the absence of energy-dissipating processes (such as flux motion) in actual applications. The density and morphology of the retained twin boundaries in a crystal mainly depend on local conditions during synthesis, such as impurities and stress and thermal gradients during processing. Twin boundaries have a stronger pinning effect on magnetic vortices when magnetic flux lines are parallel to the a/b or basal plane (Liu et al., 1990; Welp et al., 1990).

We have grown high-quality $\text{YBa}_2\text{Cu}_3\text{O}_7$ single crystals with T_c between 93 and 94 K and transition widths between 1 and 2 K by a modified self-flux method in gold crucibles. A slow cooling procedure after melting at high temperature provides enough time for the solute ions to transfer from the melt to the growing crystals. Single crystals containing a large area of single, twin boundaries are often found in those places where growth is undertaken more freely, so less stress is

encountered by the growing crystals. We have developed a technique that is based on this observation to control nucleation so that many crystals can be grown free-standing. We then carefully select crystals with a 1-to-1 correspondence of twin structures on both faces. Figure 1 shows this morphology for a typical crystal, where a 1-to-1 correspondence of twin boundaries on both faces shows that the

Figure 1. Morphology of a $\text{YBa}_2\text{Cu}_3\text{O}_7$ crystal containing only one set of twin boundaries when viewed from each side (a) and (b). Note the 1-to-1 correspondence of the twin boundaries on both sides.

entire bulk of the crystal contains one set of twin boundaries.

We conducted magnetic measurements using a SQUID magnetometer, carefully eliminating misorientation in magnetic fields up to 5.5 T applied normal to the *c* axis. By comparing the magnetization hysteresis loops at 6 K, we discovered that when the magnetic field is parallel to the twin boundaries, the pinning force is significantly stronger than it is when the field is perpendicular. To study the temperature dependence of the vortex/twin-boundary interaction, we measured the hysteresis loops for each of the two samples at temperatures from 6 to 77 K with the magnetic field either parallel or perpendicular to the twin boundaries. We used the extended Bean model (Gyogy et al., 1989) to calculate the critical current density J_c at each temperature. Here, we confine our discussion to the situation in which the applied field is beyond the initial saturation field, which is about 0.4 T at 6 K. In the temperature range of 6 to 77 K, we observed that the magnetization critical current density J_c in the parallel case is always larger than that in the perpendicular case. Thus, earlier reported sign changes in the

vortex/twin-boundaries interaction were erroneous.

Because J_c is proportional to the pinning potential energy U_0 , the ratio $J_c^{\parallel}/J_c^{\perp}$ should be the same as $U_0^{\parallel}/U_0^{\perp}$. To verify this point, we conducted a shielding magnetization time-relaxation experiment on our best sample of $\text{YBa}_2\text{Cu}_3\text{O}_7$ at different temperatures and magnetic fields. Figure 2 shows the results obtained at 30 K with a 2-T magnetic field either parallel or perpendicular to the twin boundary. The observed faster decay of magnetization over time in the perpendicular orientation confirms that the pinning force is weaker than it is for the parallel orientation. The pinning potential U_0 can be obtained from the relation:

$$U_0 = kT(M_0 - M_{\text{eq}})d\ln(t)/dM, \quad (1)$$

where k is the Boltzmann constant; M_0 is the initial magnetization taken at $t = 400$ s, where the linear behavior of $dM/d\ln(t)$ begins; and M_{eq} is the final stable value, which can be replaced approximately by the Meissner magnetization at the same temperature and magnetic field. The pinning potentials U_0 obtained from Figure 2 for the

parallel and perpendicular cases are $U_0^{\parallel} = 0.06$ eV and $U_0^{\perp} = 0.04$ eV, respectively. The difference in pinning potential energy for these two vortex/twin-boundary interaction modes is 0.02 eV, a significant value relative to the size of U_0 .

The difference in vortex/twin-boundary interaction between parallel and perpendicular modes may be associated with the local chemistry related to the twin boundaries. Transmission electron microscopy (TEM) studies on twins (Sarikaya et al., 1988) show that the structure of the boundary region is different from the bulk orthorhombic. The value of $\Delta a/a$ (where $\Delta a = b - a$) measured by electron microdiffraction at the boundary region is much smaller than that of the bulk. This difference implies that the tetragonal structure might be a stable phase at the boundary. Recent studies using field-ion microscopy on twin structures confirm the depletion of oxygen concentration across the boundary (van Bakel et al., 1990). Hence, elastic strain exists in the region between these two structures.

Note that strain and local chemistry are believed to play a role in determining the weak-link Josephson junction and pinning properties of grain boundaries. Although second phases (e.g., BaCO_3) are often associated with grain boundary properties, the strain energy at grain boundaries, grain boundary energy, and the concomitant chemistry may be decisive in polycrystalline material properties. A search for a $\text{ReBa}_2\text{Cu}_3\text{O}_{7-\delta}$ material, where strain energy is accommodated without oxygen loss, may prove useful.

Halogen Doping of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$

Introduction of the halogens chlorine or bromine can restore >90-K superconductivity to oxygen-deficient $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. Halogen doping

Figure 2. Magnetization time-relaxation curves of a sample $\text{YBa}_2\text{Cu}_3\text{O}_7$ at 30 K and 1 T for magnetic fields parallel and perpendicular to the twin boundary.

is potentially important for applications to thin-film devices because of the low processing temperatures required relative to analogous processing with oxygen. At 260°C, the bromination process requires only 5 min to restore 90-K superconductivity with essentially a 100% volume fraction to a $\text{YBa}_2\text{Cu}_3\text{O}_6$ nonsuperconducting powder (Yu et al., 1988, 1989; Radousky et al., 1990). We previously reported that bromine is strongly bonded in the material and is not released even when heated in hydrogen to 1000°C. We have now accumulated new experimental data showing how halogenation may restore superconductivity by rearranging available oxygen in the unit cell.

We prepared powders of $\text{YBa}_2\text{Cu}_3\text{O}_7$ materials with oxygen content near 7.0 (formula weight) and 90-K transition temperatures. Then we annealed the powders in flowing nitrogen at 600°C to remove oxygen, which made the powders insulating and nonsuperconducting. Next, we exposed the powders to either bromine or chlorine in a sealed vacuum line. Flasks containing the powders were heated to the appropriate temperatures, 160°C for Cl_2 and 260°C for Br_2 , and maintained at those temperatures overnight to ensure that any moisture would be removed from the powders. Finally, the deoxygenated material was exposed to the halogen vapor.

Figure 3 shows the temperature vs time curves for 1-h halogenations using Cl_2 starting at 160°C and Br_2 at 260°C. In both cases, we observed a steep temperature rise when we introduced the halogen into the processing vessel, demonstrating that the reaction is exothermic in nature. In 3 min, the brominated sample rose from 260 to 410°C, while the chlorinated sample rose from 160 to 280°C.

For both halogens, the steep rise was followed by a temperature

falloff to a steady state, intermediate between the starting and maximum temperature. This secondary reaction is probably the source of deleterious phases, which we reported in earlier work. One intriguing aspect of the halogenated materials is the strong binding of bromine. This binding is consistent with the exothermic nature of the reaction. Although bromine is not removed at high temperature, additional oxygen can be removed by exposing the sample to a nitrogen atmosphere at 600°C. Preliminary experiments

(Figure 4) show that after removal of oxygen from the brominated powders, the material becomes insulating again. Exposure of these powders to bromine for a second time returns them to 90-K superconductivity.

Our new chemistry data confirm an earlier suggestion that bromine "harvests" apical oxygen (oxygen in the bridging site between the CuO_2 planes and CuO chains) and displaces it into the CuO chain, thus restoring superconductivity. This result implies a possible charge-transfer doping mechanism related

Figure 3. Temperature vs time histories for $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ powders exposed to bromine at 260°C or chlorine at 160°C. Note the similarity of the chlorine and bromine time dependences. The sharp decrease in temperature at 60 min corresponds to turning off the heater.

Figure 4. Magnetization data for a series of bromination studies in $\text{YBa}_2\text{Cu}_3\text{O}_{6.84}$ (square data points), after removal of ~ 0.8 moles of oxygen and a first bromine treatment (closed circles), and after removal of ~ 1.6 moles of oxygen and a second bromine treatment (open circles). The flat curve at the top (shown in color) confirms the insulating phase characteristic between bromination steps for each cycle.

to the redox properties of the CuO_2 plane and CuO chain and their relative proximity in the unit cell. Such a mechanism could be a novel chemical way of viewing such materials. Our results may also have implications regarding the controlled optimization of frequency-dependent properties, particularly so-called A.C. losses, where chain perfection appears to play an important role.

Theoretical Description of Electron Structure

What should be a simple question of electronic structure, namely, identification of the states occupied by the doped carrier holes, remains unanswered after 3 y of intense research on high- T_c cuprates by the international scientific community. The ultimate answer will discriminate between the "anyon" school of Anderson, Laughlin, Wilcek, and others who require the doped holes to occupy p_σ states; the "de-excitation" school of Weber, Cox, and others who require p_π states; and lingering theoretical suggestions that these holes may be in p_z states on the apical oxygens. Experiments to date cannot distinguish between p_s and p_p states; although most rule out p_z , this conclusion is contested by some groups.

Such simple questions of electronic structure remain unanswered because electrons or holes in the high- T_c cuprates are both itinerant, as they are in common metals, and strongly interacting, as in localized atomic and molecular systems. This challenging combination falls between existing theoretical formalisms that stress one limit at the expense of the other. Our goal is to develop a quantitative theory of electronic structure for this new class of materials because we believe

the high- T_c puzzle will not be solved until such fundamental materials questions are answered.

Our strategy in dealing with itinerant, yet strongly interacting, electrons or holes is to disregard extraneous degrees of freedom and retain, in more manageable model Hamiltonians, only those low-energy excitations that might be relevant to superconductivity. Although this strategy is shared, in part, by many groups, other models are usually based on intuition. We have modified our existing local-density functional method that generates quantitative and material-specific parameters defining such model Hamiltonians. Solving these models is now the major component of our effort. To demonstrate the quantitative abilities of this combined approach, our near-term goal has been to calculate the insulating gaps, superexchange frequencies, and doped carrier states in the isostructural compounds La_2CuO_4 , La_2NiO_4 , K_2CuF_4 , and K_2NiF_4 . Because only La_2CuO_4 has been doped to high- T_c superconductivity, our comparison may also allow us to identify features necessary for the high- T_c phenomenon.

Our earlier effort (McMahan et al., 1988) to calculate model Hamiltonian parameters was strictly valid only for the fairly localized $\text{Cu}(3d)$ states in the cuprates, although estimates were made for the more extended $\text{O}(2p)$ states, and an approximate solution of the model yielded several quantities in reasonable agreement with experiment (McMahan et al., 1990). We have now generalized this approach using numerical Wannier functions to states of arbitrary localization, providing even-handed treatment of both $\text{Cu}(3d)$ and $\text{O}(2p)$ states (Annett et al., 1989). The problem of

generating model Hamiltonians is now largely solved.

We first solved the model Hamiltonians for small clusters (e.g., 1 to 4 formula units) of the La_2CuO_4 , La_2NiO_4 , and K_2CuF_4 compounds (Grant and McMahan, 1989). Although a relative comparison of the calculated insulating gaps appeared promising, variations in the results with cluster size was disturbing and reminded us of the itinerancy of the important states, which requires bulk-like treatment.

Our approach is based on a limited configuration-interaction solution of *ab initio*-derived, multi-band Hubbard Hamiltonians. It can deal simultaneously with the somewhat conflicting attributes of the cuprate electron states. We successfully tested this method by calculating the superexchange frequency and insulating gap of the parent cuprate La_2CuO_4 and the three isostructural compounds. We are now carrying out the first realistic calculations of the quasi-particle spectra of La_2CuO_4 .

Our results for the La_2CuO_4 quasi-particles resolve previous controversies as to the relevance of $^1A_{1g}$ vs $^3B_{1g}$ symmetry states in cluster calculations, the validity of mapping to the abstracted one-band Hubbard model, and the in-plane anisotropy of the Fermi surface required to explain normal-state transport properties. We suggest that doped holes in La_2CuO_4 occupy a single, mostly $\text{O}(2p)$ quasi-particle branch for which the local $\text{O}(2p)$ - $\text{Cu}(3d)$ relation evolves from $^1A_{1g}$ -like at $k = (\pi/2, \pi/2)a$ to more $^3B_{1g}$ -like at $(\pi, 0)a$. The first holes should, indeed, form local singlets ($^1A_{1g}$ symmetry) with the pre-existing copper spins. However, at doping levels approaching $x = 0.26$, we suggest a growing admixture of local $^3B_{1g}$ symmetry, which may be

correlated with the observed loss of high-temperature superconductivity at these higher doping levels. We anticipate that angle-resolved photoemission studies at LLNL will provide critical experimental comparisons for these calculations. ■

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The Structure-Property Link in Subnanometer Materials

A. F. Jankowski

We are investigating equilibrium atomic configurations and phase stability of metallic multilayer systems, such as gold/nickel, using high-resolution electron microscopy and x-ray diffraction. The observed atomic structure is linked with measured elastic behavior using the concept of strain relaxation from coherent interfaces.

Figure 1. (a) Schematic of the beam-deflection apparatus. (b) Variation of planar stress with layer pair, arising from the curvature of gold/nickel composite microbeams, compared with applied bulge test stresses, which induced film failure.

Structure-property relationships are critically linked to interfaces in materials of subnanometer dimensions. Such materials include natural and artificial superlattices, i.e., microstructures with compositionally modulated layers composed of only a few atomic planes each. The interface structure of artificial superlattices determines the behavior of these atomically layered composites.

Superlattices have physical properties and structural characteristics that are often unlike those of the constituents, either in elemental or rule-of-mixtures form. Lattice distortions in the layered structure are commonly incorporated in modeling the electronic behavior, and they now appear useful for modeling the structural stability and elastic behavior as well. The quantitative sensitivity of each analytic technique used to measure material properties is critically different. The use of microindentation techniques, for example, may be limited to material responses on the same dimensional order as the measurement, i.e., 10 nm or more.

We synthesize the metallic multilayer structures using dc magnetron sputter deposition. The thin-film samples are composed of several hundred layer pairs with repeat periodicities ranging from 0.6 to 9 nm. This vapor deposition process is competitive with molecular beam epitaxy in that it allows for the monolayer level of control needed to produce superlattices with compositionally abrupt interfaces. The multilayer systems under investigation are gold/nickel, nickel/titanium, and gold/niobium.

We perform the mechanical property measurement of gold/nickel

multilayers using a microindentation probe, the NanoIndenter. The measurement of the in-plane Young's modulus from the deflection of a composite multilayer-cantilever beam (Figure 1a) has revealed only a 10 to 20% modulus enhancement. We are investigating the microstructure and the testing methodology to determine why the supermodulus effect, a twofold increase in the in-plane elastic modulus, is not detected. Direct measurement of the multilayer-cantilever beam curvature does prove interesting, however. If a constant modulus value is assumed, the computed internal stress of the multilayer-cantilever beam follows the composition wavelength dependence of the supermodulus effect, i.e., a maximum compressive stress at 2 nm (Figure 1b). The trend is equal in magnitude and opposite in sign to the breaking stress of similar foils examined using the bulge test. If a constant stress is assumed, however, the computed multilayer modulus follows the established supermodulus trend. These modulus measurements provide a milestone for mechanical-property evaluation of as-deposited thin-film multilayer structures.

The microstructure of the gold/nickel multilayers is characterized by measuring x-ray diffraction (XRD), modeling, and using high-resolution electron microscopy (HREM). Measurements of interplanar spacings from high-resolution gold/nickel micrographs (Figure 2) provide a direct test of the "strain relaxation from coherent interfaces" concept as proposed in the coherency strain model (CSM). From the HREM measurements and new modeling analysis techniques, the composition

profile through the interfaces of the multilayer may be analytically and uniquely determined. Preliminary kinematic XRD results are supportive of the CSM.

The gold/niobium system is one in which a near-zero misfit exists between (111) Au and (110) Nb. Yet, when deposited as a multilayer, a strained layered superlattice results. In fact, the average lattice spacing d_{Bragg} in the gold/niobium modulation direction varies with the repeat period beyond a rule-of-mixtures average (Figure 3), as also found for gold/nickel.

Multilayer structures are useful for studying alloy-stability phenomena and phase transformations. Consider the case of solid-state amorphization as induced through the mechanical alloying of powders through ball milling or the irradiation of crystalline solids. The kinetics and driving forces of bulk processing do not readily clarify the physics of amorphization. Thin-film studies, without an amorphous parent phase, remove the bulk-processing obstacles. We used a completely crystalline multilayer to study the solid-state amorphization of nickel/titanium (55 at.% nickel). An in situ transmission electron microscope (TEM) isothermal anneal was used to amorphize the crystalline binary structure via interdiffusion of nickel preferentially into the titanium. This is perhaps the first time that the phenomenon has been observed in real time. Determination of the structural origin may now be more rigorously pursued, including the question of whether the amorphous phase nucleates from an incoherent or coherent interface. Additional samples with progressively shorter repeat periods may lead to nickel/titanium interface structures that pass through an incoherent-coherent transition and, therefore, help clarify the

structural mechanism of solid-state amorphization.

In summary, we verified atomic configurations (corresponding to relaxation from strained interfaces) proposed to model the elastic

behavior of metallic superlattices through the use of HREM and XRD. We are developing experimental techniques such as microdeflection/indentation to measure thin-film elastic behavior.

(a) Projection [110]

Figure 2. (a) Digitized image of a gold/nickel multilayer with a 4.4-nm period, as viewed in cross section along the [110] pole projection. The atomic planes within the nickel layers are coherent on the gold layers, whereas the gold "twins" on the nickel during film formation. (b) The (111) spacing measured in the growth direction of the figure shown in (a). Relaxation from each strained interface to bulk values is seen within each layer.

Figure 3. The x-ray diffraction measurement of $1/d_{\text{Bragg}}$ variation with the repeat period for $d_{\text{Au/Nb}}$ of gold/niobium multilayers reveals a strained superlattice despite a near-zero bulk misfit between the components.

The progress generated from the use of metallic multilayers to study structure-property phenomena led to the following FY91 goals:

- Examine the coherent interface role on stability in the silver/nickel system (by comparing the results of the embedded atom calculations with the interface structure and the formation of

layering) and in the nickel/titanium system (for effects on solid-state amorphization).

- Conduct indentation experiments at shallower depths (<10 nm) for a dimensional probe on the same scale as the multilayer composition modulation.
- Conduct bulge-test measurements to correlate beam-deflection data with the technique used in the discovery of the supermodulus effect.

- Conduct dynamic XRD modeling of interface composition and the large lattice distortions in metallic superlattices.
- Uniquely determine the interface composition of sputter-deposited metallic multilayers through the coupling of high-resolution electron microscopy and XRD lattice-distortion measurements.
- Perform novel transport-property measurements and correlate the results with available theory for multilayers with small layer-pair spacings. ■

Computation

R. R. Borchers

The Computation Directorate conducts multidisciplinary research to provide scientists with the most effective means for using computers to solve their problems. In FY90, the Directorate received ER&D funding for three Exploratory Research projects: one in fluid dynamics modeling and two in massively parallel computing.

Our fluid dynamics project is part of a significant collaboration involving LLNL, New York University, Los Alamos National Laboratory, the University of California (UC), Berkeley, and UC, Davis. We are developing new and efficient computational algorithms using high-resolution discretization techniques, local adaptive mesh refinement, and front tracking to solve complex, three-dimensional compressible-flow problems. In FY90, we improved the basic methodology and completed a three-dimensional adaptive mesh-refinement algorithm for tensor-product grids. Computational experiments for a shock wave hitting an ellipsoidal bubble of Freon 22 accurately model the comparable physical experiments and require an order of magnitude less computation time than the uniform grid computation of comparable resolution.

The goal of our Sisal 2.0 massively parallel computing project is to demonstrate the potential of functional languages for parallel scientific computing. Using Sisal, programmers can more easily express concurrency and, at the same time, avoid all possibility of creating time-dependent errors. These features should greatly reduce the cost of producing efficient, correct parallel codes. In FY90, we completed the definition of Sisal 2.0, adding several new features based on six years of experience with the previous version. We also began moving our existing software to the BBN TC 2000 because it enables us to increase speed without reworking the system. However, to produce outstanding results (comparable to those we achieved for shared-memory systems), we will need substantial research on the memory-mapping and partitioning issues.

As part of our project to model plasma phenomena on massively parallel computers, we are studying methods for solving Fokker-Planck equations and comparing our ability to exploit different types of multiprocessors. This research is essential to understand the ways in which the Laboratory will be able to use parallel machines. This work is of particular interest in the fusion community, specifically in the area of tokamak modeling. We expect to learn how the complexities and trade-offs of different multiprocessor systems affect the ways in which solutions to problems are represented and computed. In FY90, we compared the Cray-2 and two massively parallel systems—the Connection Machine 2 (CM2) and the BBN TC 2000. Our initial results show which problems each system solves best, even though the systems vary substantially in terms of programming time and machine cost. Our analysis will provide valuable data on the selection and effective use of future generations of computers for LLNL.

In FY90, the Computation Directorate received \$475,000 in ER&D funding. ■

Adaptive Numerical Methods for Fluid Dynamics in Three Dimensions

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We are developing adaptive numerical methods for modeling time-dependent fluid flows in three dimensions by combining high-resolution discretization techniques with local adaptive mesh refinement and front tracking. The net result is a dramatic improvement in computational efficiency.

Computational fluid dynamics play a central role in numerical simulations in such diverse areas as weapons design, weapons effects fusion, and energy production. Typically, these computations have been based on assumptions of physical symmetry to reduce the dimensionality of the computation. However, there is an increasing need to perform calculations in three space dimensions. A common feature of three-dimensional (3D) flows is the appearance of small-scale, localized phenomena; one routinely encounters features such as jets, vortex rings, and mixing layers whose characteristic length scales are small relative to the size of the problem domain, but whose dynamics are important in the overall physics.

Adequate resolution of these phenomena using conventional spatial discretizations would require computational effort exceeding the most optimistic projections for

near-term growth in computational capabilities. Our goal is to develop adaptive methods for modeling 3D, inviscid compressible flow that will concentrate computational effort where it is needed to maintain the accuracy of the numerical solution.

The choice of a discretization scheme for the flow equations is central to developing effective adaptive methods. Our techniques are based on a higher-order Godunov method (Bell et al., 1989), which is a high-resolution, conservative finite-difference scheme that incorporates nonlinear wave-propagation properties of the flow to provide both high accuracy and robustness for strong shock waves. In addition, the method centers all of the variables for each grid cell at the same locations on the grid, which simplifies its coupling to adaptive methodologies.

We are developing two types of adaptive techniques. The first is adaptive mesh refinement (Berger and Colella, 1987) in which the finite-difference mesh dynamically changes as a function of space, time, and data to maintain a fixed level of accuracy in the calculation. In particular, the total number of mesh points in the calculation may change as a function of time, yielding a method with a high degree of flexibility in concentrating computational effort in regions of greatest interest. The second technique is front tracking in which we treat selected discontinuities as free boundaries moving through the finite-difference mesh.

During FY90, we developed a 3D, adaptive-mesh-refinement algorithm for tensor-product Cartesian grids. The algorithm is based on a hierarchical grid structure composed of grids of varying resolution. The grid hierarchy is constructed from an initial coarse grid on which we use an error estimation procedure to identify cells having unacceptable errors. These cells are then clustered into logically rectangular grids that are subdivided to form finer cells where more resolution is required. We apply this procedure recursively until the desired accuracy is achieved or a maximum level of grid nesting is reached.

We have also incorporated several improvements in the basic methodology. First, we developed a new algorithm based on signature analysis techniques from image processing to organize grid cells that require refinement into grid patches. The new algorithm provides a more efficient cell grouping than the original bisection algorithm and improves performance. We also developed a new 3D integration scheme for modeling a general fluid equation of state that reduces the computational cost from 45 μ s/zone to about 25 μ s/zone on a Cray XMP. To test the improved algorithm, we modeled the interaction of a shock wave with an ellipsoidal bubble of Freon 22 taken from a study of Haas and Sturtevant (1987). The use of adaptive mesh refinement reduces the computational cost by more than an order of

magnitude compared with the cost of a uniform grid computation of comparable resolution. Figure 1 shows a time history of the Freon bubble deformation.

In FY91, we will incorporate front tracking into the adaptive-mesh-refinement algorithm. This incorporation will allow explicit modeling of the propagation of selected discontinuities, such as a shock wave or a material interface, through the computational grid. We have incorporated a standard slip-line in-cell (SLIC) algorithm

for tracking a material interface and are developing a material-interface algorithm that uses a higher-order representation of the interface. This algorithm gives an accurate representation of the frontal geometry even on the coarser-level grids. We are also developing a volume-of-fluid algorithm for tracking shock waves. The combination of tracking and adaptive mesh refinement will provide the necessary tool to investigate 3D phenomena in fluid dynamics. ■

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Figure 1. Time history of a shock-induced collapse of a Freon bubble computed using adaptive mesh refinement. Images show the location of the cloud material. The use of adaptive refinement reduced the computational cost by an order of magnitude compared with the cost of a uniform grid computation of comparable resolution.

Modeling Plasma Phenomena on Massively Parallel Computers

M. G. McCoy and A. A. Mirin

We are investigating the suitability of massively parallel computers for modeling plasma phenomena. We find that, for finite-difference Fokker-Planck equations, the Connection Machine outperforms the Cray-2 for some simulations and that, for a Monte Carlo Fokker-Planck model, the BBN TC2000 is competitive with the Cray-2.

Historically, enhancements in computer power have been driven by improvements in hardware. However, technical limitations have shifted the focus to computer architecture, which has led to shared-memory multiprocessors such as the Cray-2. Researchers have recognized that a more cost-effective approach might be to link numerous slower, cheaper processors, each with its own local memory. Such machines are known as massively parallel computers.

The purpose of our project has been to investigate the suitability of massively parallel technology for modeling plasma phenomena. In FY89, we studied two related areas: kinetics, using the finite-difference Fokker-Planck approach, and turbulent transport. In FY90, the turbulent transport project spun off into a new project funded by the DOE, and we continued the Fokker-Planck research under ER&D auspices. In addition, we expanded the scope of

the ER&D effort to include Monte Carlo Fokker-Planck calculations.

The Fokker-Planck equation can be used to model collisional relaxation in an ionized gas. In its full generality, the Fokker-Planck operator models time-dependent phenomena in six-dimensional phase space. However, symmetry and ordering assumptions can often be made, and if they are coupled with the assumption that the magnetic field is uniform and that radial transport times are slow relative to velocity space diffusion, the dimensionality can be reduced to two velocity space variables.

A widely used code that invokes such assumptions is FPPAC (Mirin et al., 1988), which solves the multi-species Fokker-Planck operator using finite differences. Over the past 2 y, FPPAC has been converted to run on the Connection Machine 2 (CM2), a massively parallel device consisting of up to 65 536 single-bit processors, each with up to 32 kbytes of random access memory. The CM2 is a single-instruction multiple-data device; that is, all active processors must simultaneously execute identical instructions on their data sets. A Fortran compiler with parallel extensions is available. In converting FPPAC to the CM2, we replaced Cray-optimized algorithms with ones that minimize the number of parallel steps (Mirin, 1990).

Calculations have been carried out on CM2s at the National Aeronautics and Space Administration (NASA) facility at Ames, Iowa, and at Los Alamos National Laboratory. As the

problem size increases, the CM2 becomes more advantageous. For a relatively fine mesh (65 536 mesh points), the time-integration procedure, which involves solving parallel tridiagonal systems, executes at the same speed on the CM2 (using 64-bit arithmetic) as on the Cray-2. Using a 64-mode Legendre expansion to compute the collisional coefficients, which involves matrix multiplication and moment computation, the CM2 executes at six times the speed of the Cray-2. Cases that do not require a long Legendre expansion do not fare as well on the CM2 because the degree of parallelism is smaller and the matrix multiplication on the CM2 is tuned for very large matrices.

The primary goal of the Monte Carlo code is to quantify certain radial diffusion coefficients. In our application, the trajectories of an arbitrary number of charged particles are tracked using the Littlejohn (1983) orbit equations. The collisional effects are achieved by imparting random kicks, assuming that the background with which the particles collide is Maxwellian and constant in time. This assumption is easily relaxed. In the absence of excitation, the particles are confined to unchanging orbits and thus have some fixed-average radial coordinate. However, excitation alters a particle's adiabatic invariants, resulting in a change in the radial coordinate. This diffusion is the focus of our investigation.

The fixed orbits are of three types: passing, co-passing, and trapped. The trajectories of the first two have the aspect of closed curves

when projected onto one toroidal slice of the tokamak and are distinguished only by the direction that they rotate about the device, whereas the trapped particles execute a "banana" orbit (Figure 1). This code is written for the LLNL BBN TC-2000 (or Butterfly) and also runs on the Cray-2 so we can compare efficiency.

The BBN Butterfly is a multiple instruction, multiple data machine; that is, the various processors perform independent tasks simultaneously. We use a shared-memory programming paradigm. Using 64-bit arithmetic, the code executes at about the same rate using 48 processors on the BBN as on a full four-processor Cray-2 operating with largely scalar arithmetic. Vectorization of the Cray version, which would involve indirect indexing, could result in a factor of 3 improvement.

Of interest is whether the technique of computing the coefficients using statistics gathered from particle excursions is practical on this or a follow-up machine. Coefficients are required for 24 000 mesh points describing a crude (v, θ, r) mesh. It currently costs 0.4 s to compute a single diffusion coefficient. Substitution of a low-order, ordinary differential-equation solver, relaxation of error tolerances, and single-precision arithmetic will result in a

probable factor of 6 improvement to 0.07 s on a 48-processor machine. This improvement translates to about 28 min for the entire array, which is reasonable. Optimism is enhanced by the linear scalability of speed with the number of processors.

These projects deal with relatively simple models to achieve a proof of principle in a timely manner. Our plan for FY91, however, is to show that a production Fokker-Planck code with its mathematical complexities, size, and modularity can be used to good effect. We intend to convert CQL3D, a bounce-averaged, finite-difference Fokker-Planck code (McCoy and Kerbel, 1986), to the BBN TC2000 computer at LLNL. Over the long term, we hope to generalize CQL3D to include radial transport, among other things. The Monte Carlo module would calculate the radial diffusion coefficients self-consistently. ■

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Figure 1. Particle orbits projected onto one up-down slice of the toroidal device. An initially passing particle (A) becomes trapped (B), then co-passing (C), then trapped (D), and finally passing (E). Magnetic geometry and collision frequencies correspond to the DIII-D experiment at General Atomics in San Diego.

Sisal 2.0 for Distributed-Memory, Massively Parallel Machines

D. C. Cann and J. T. Feo

Our goals are to define, implement, and evaluate Sisal 2.0, a general-purpose functional language, on distributed-memory computer systems and to help define the role of such machines at LLNL.

Although we have purchased several parallel computer systems over the past decade, the number of parallel production codes at LLNL remains virtually zero. Most of the Laboratory's multiprocessor computer systems are run as collections of uniprocessors, greatly reducing the potential of these machines. The primary reason for the lack of parallel programs at LLNL is that current programming languages, such as Fortran, make the development of correct parallel programs arduous and error-prone. These languages, which were originally developed for sequential processing, require

users to correctly encode program decomposition, task synchronization, task communication, mutual exclusion, and determinacy.

Sisal, which is a general-purpose functional language, provides an easy-to-use, parallel programming model that facilitates algorithm development and simplifies compilation. The semantics of Sisal promote the construction of correct, parallel programs by isolating the user from the complexities of parallel programming. All parallelism is implicit; a user does not explicitly manage communication and synchronization primitives. Moreover, programs give the same results without modification regardless of how, when, or where they are executed.

In FY90, we defined Sisal 2.0, an extension of Sisal 1.2 (McGraw et al., 1985). The two most important new features are higher-order functions and a mixed-language interface. Higher-order functions provide a means to customize program codes. For example, consider a program that applies a set of reductions to a data set. Instead of writing a separate function for each reduction, we can write a single higher-order function that takes a reduction and a data set as input and applies the reduction to the set. Thus, the program is customized dynamically. Next, consider the higher-order integration routine, Simpson(f, a, b, δ), where f is the function to be integrated using the

Simpson Method. If the function is known at compile time, the compiler can expand the function and optimize the integration routine. The resultant code will be as efficient as if the user had written an integration routine specifically for f .

The mixed-language interface provides for the opportunity of mixing imperative and functional program modules in the same application. It is unrealistic to expect LLNL, or any scientific group, to rewrite its production codes from scratch in Sisal. However, a module-by-module evolution from sequential Fortran to Sisal will be cost effective and will allow the immediate exploitation of parallel machines. The enormous cost of rewriting large mathematical Fortran libraries will also be avoided.

In the spring of FY90, we began investigating possible implementations of Sisal on the BBN TC2000. Critical to this study is an understanding of the cost of local vs remote-memory accesses. The current Sisal implementation assumes uniform memory-access times. Although this model achieves execution performance equivalent to Fortran and exhibits significant speedups on shared-memory machines in both scalar and vector modes (Cann and Feo, 1990; Cann et al., 1990; Feo et al., 1990; Cann, in press), it fails to effectively exploit the BBN architecture. Table 1 shows the execution times of three Sisal applications run on the TC2000. On

Table 1. Execution times of three scientific applications on the BBN TC2000 assuming uniform memory-access times.

Name of application	One processor (s)	Five processors (s)
Simple	326.0	155.3
Ricard	32.3	12.9
BMK11A	15.0	23.8

one processor, the applications ran in the local memory of the single processor, whereas on five processors, they ran in shared memory. Both the Simple and Ricard scientific applications showed speedups but considerably less than that on shared-memory machines. The BMK11A, an access-intensive application, showed a decrease in performance. All three codes demonstrate the need to exploit local memory.

Our intention is to develop a Sisal run-time system that schedules the producers and consumers

of data objects on the same processor, thereby localizing array accesses. If our goals are met, we will help establish the programming medium for the foreseeable future, much as the first Fortran compilers did in the 1950s. ■

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Earth Sciences

L. W. Younker

The Earth Sciences Department supports the Laboratory's energy, defense, environmental, and basic research programs in various disciplines, including geology, seismology, physics, geophysics, geochemistry, geohydrology, chemical engineering, geotechnology, and mechanical engineering. Comprising approximately 100 professional scientists and engineers, the Department is one of the largest geoscience research groups in the nation.

Current support is divided between two areas: defense-related work, which includes research in containment of radioactive debris from underground nuclear tests, treaty verification, and shock physics; and energy and environmental work, which includes research in nuclear waste disposal, fossil energy, environmental restoration, and basic energy sciences. The Department uses ER&D funds to develop new capabilities aimed at possible future program directions, to prepare for possible changes in program priorities and funding, to encourage high-leverage basic research outside the scope of current program interests, and to attract high-quality staff members.

The ER&D program funded projects in four major areas: in situ environmental remediation, global climate change, computational rock mechanics, and high-pressure experimental geophysics.

In the aqueous organic geochemistry project, we are developing a modeling capability to complement our inorganic modeling EQ36 codes. This calculational ability will enable us to evaluate geochemical problems of interest to DOE, including the fate of organic contaminants in the subsurface, the role of aqueous organics in the formation of petroleum deposits and enhanced oil recovery, and the potential role of organic species in the mobilization of trace-metal pollutants.

In another project, we are evaluating indicators of global climate change. Our goal is to produce a data base for studying current and past climatic conditions.

As part of the computational rock mechanics project, we are developing and applying new methods that will allow us to better estimate the effects of nuclear and conventional explosives in rock. These methods are based on discrete-element analysis, which is currently the only viable tool for simulating the dynamic response of discontinuous media such as rock masses penetrated by numerous faults and joint sets.

The objective of the experimental geophysics project is to design, fabricate, and develop a multi-anvil, solid-media pressure apparatus capable of achieving pressures to 25 GPa and temperatures to 2400°C. The facility will allow us to research the evolution of planetary interiors and differentiate primordial planetary processes. It also will extend LLNL's material synthesis capabilities.

In FY90, the Earth Sciences Department received \$200,000 in ER&D funding. ■

Large-Volume, High-Pressure Apparatus for Geophysical and Geochemical Research

F. J. Ryerson

We have built a multi-anvil solid media pressure apparatus capable of achieving pressures to 25 GPa and temperatures to 2400°C for studying the evolution of planetary interiors.

The study of basalt petrogenesis has greatly benefited from high-pressure phase equilibria. The surfaces of the terrestrial planets, meteorites, and presumably asteroids are composed of large fractions of basaltic material. Basaltic melts are produced by partial melting of the more mafic materials (peridotites) that comprise the interiors of the terrestrial planets or, in meteorites and asteroids, the interiors of their parent bodies. For example, the mid-ocean-ridge basalts that cover two-thirds of the Earth's surface are produced by partial melting of peridotites at depths less than 100 km. The compositions of basaltic magmas allow geologists to make inferences about the nature of planetary interiors.

Recent models theorize that the waning stages of Earth's accretion were characterized by the impact of relatively large bodies—on the order of the mass of Mars. The energy dissipated by these impacts coupled with that liberated during the formation of Earth's metallic core melted the entire planet. Upon cooling, the crystallizing, terrestrial, magma ocean experienced chemical fractionation due to the density-driven segregation of crystals from residual melt.

To test this hypothesis, we must compare the composition of the present-day Earth's mantle with the model's suggested mantle by determining the partitioning of major and trace elements between all of the potential crystallizing phases and residual melts at the appropriate pressures and temperatures. The depths at which density-driven fractionation would have occurred far exceed those of present-day basalt formation as well as the capabilities of standard experimental apparatuses. To address this problem, we designed, constructed, and calibrated a multi-anvil press with several furnace/sample assemblies capable of achieving pressures to 25 GPa and temperatures to 2400°C; details of the design are given in previous reports (Ryerson et al., 1988; Ryerson and Dibley, 1989, 1990).

Using the press, we have begun experiments relevant to the fractionation of the terrestrial magma ocean. Figure 1 is a photomicrograph of a sample produced in such an experiment at 1800°C and 4.5 GPa, showing the coexistence of three crystalline phases—garnet, olivine, and clinopyroxene—with a melt of komatiitic composition. Komatiites are high-MgO (~30 wt%) magmas thought to represent the composition of magmas following fractionation of the magma ocean. Additional runs are being conducted at higher pressures to synthesize assemblages containing minerals such as majorite, perovskite, and magnesiowüstite. ■

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Figure 1. Backscattered electron image of a komatiite composition equilibrated at 1800°C and 4.5 GPa. The minerals garnet (Gt), clinopyroxene (Cpx), and olivine (Ol) coexist with a "melt" phase (M) that is present as a mass of quench crystals.

Aqueous Organic Geochemistry

K. J. Jackson and N. M. Rose

Irreversible mass transfer calculations demonstrate how organic species may respond to changes in solution composition and participate in the phase relations of complex geochemical systems

The capabilities of the EQ3/6 geochemical modeling codes have been enhanced to include reactions that account for aqueous solutions containing both inorganic and organic species. This addition improves the ability of EQ3/6 to model geochemical processes because reactions involving organic aqueous components can impact the irreversible reaction path followed during the geochemical evolution of a system, influence fluid flow and mass transport, and affect the geochemical fate of constituents of a natural aqueous solution.

An important aspect of chemical calculations involving aqueous organic species is the ability to model one or more states of partial equilibrium among aqueous species in the system. It also may be necessary to define the metastable persistence of organic species with respect to aqueous carbonate and methane or the precipitation of graphite (Shock, 1988). For example, Shock argued that aqueous carboxylic acid species apparently persist over geologic periods in oil-field brines even though they are metastable with respect to CO_2 and CH_4 . With its new capability and data from Shock and Helgeson (1990), EQ3/6 can compute aqueous organic speciation, determine the regions of predominance of organic species with respect

to various controlling parameters of the system (e.g., temperature, pressure, or redox state), and calculate how changes in these parameters are reflected in the geochemical properties of the aqueous phase.

The aqueous geochemistry of ocean-floor hydrothermal systems and the role these systems play in ore deposition and regulation of seawater chemistry have been widely studied (e.g., Janecky and Seyfried, 1984; Bowers et al., 1985; Von Damm et al., 1985). Thus, we selected one of these systems to demonstrate our new modeling capability. Although many studies have used aqueous speciation and mass-transfer models to understand the evolution of end-member hydrothermal solutions associated with cooling, mixing, or irreversible reaction with ocean-floor basalt or sediment, the contributions of organic species in regulating pH, redox conditions, and mineral saturation states usually have been ignored.

To quantify the effects of aqueous organic compounds on the system, we used the modified EQ3/6 codes to model the chemical evolution of Guaymas Basin hydrothermal effluents. Reaction path models that follow the conductive cooling of hydrothermal fluids starting at conditions approximating those achieved in the Guaymas Basin system generate aqueous carboxylic acid concentrations on the order of a few millimolal. Experimental investigations of organic-rich sediment reacting with seawater indicate that much of the organic material in solution is in the form of carboxylic acids, their acid anions, and aqueous carboxylic acid complexes (Thornton and Seyfried, 1987; Seewald et al., 1990).

Model results for acetic acid and propanoic acid concentrations were consistent with the millimolal concentrations of these species measured by Martens (1990) in the pores in the Guaymas Basin sediment. The ratios of computed acetic to propanoic acid components of the system are also consistent with the Martens measurements. Although our results are preliminary, the apparent agreement between calculated and measured concentrations is encouraging. ■

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FY90: \$75,000

Dynamics of Discontinuous Media

F. E. Heuze, D. M. Maddix, O. R. Walton, R. J. Shaffer, and T. R. Butkovich

We are creating new methods of analysis to simulate the response of discontinuous media, such as jointed rocks, to nuclear and conventional explosions.

We are developing new algorithms based on the discrete element method. With funding from other sources, such as the DOE Treaty Verification Program and the Defense Nuclear Agency, we are then applying the new analytic capabilities to specific problems, such as near-source explosion phenomena and vulnerability of underground structures.

We have pursued a dual-track approach to enhance our understanding of explosion effects in rocks. First, we have implemented new developments in our Discrete Block System Model (DIBS). They include:

- New silent boundaries to minimize wave reflections from the ends of the computational domain.
- Rounded corners to the discrete blocks to introduce variable porosity in the medium and prevent unwanted interlocking of blocks.
- Initial modification in data structure to promote standardization of software and portability to a Unix environment.

We also have installed DIBS on the LLNL Open Computing Facility (OCF).

The synergy of ER&D support with that of other sources can be illustrated by two applications. For the Treaty Verification Program, we have started to examine the problem of spall. A remarkable result for the simulation of an event similar to Shoal was the demonstration of a

return acceleration much greater than gravity (-1.46 g) of the spall layer at surface ground zero. Such data had been obtained on U.S. granite shots but without a plausible model to date. We also reproduced the collapse of a drift near the Piledriver Event in a fashion similar to the behavior of the prototype. We have made a video of that simulation and have recently documented our work in a publication (Heuze et al., 1990) ■

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Lake Response to Global Climate Change

W. E. Glassley

We have examined lake sediments as a basis for evaluating how changes in climate influence geochemical processes and how those processes can be monitored using satellite systems.

We have used the data of Street-Perrott et al. (1989) to obtain a global record of lake levels over the last 18 000 y, the most recent glacial period, from which we evaluated global variations in lake level as a function of latitude and time. We used this information to select those lakes most sensitive to climate change for further geochemical study.

The data document that low- and mid-latitude lakes preserve the most extreme records of changes in lake levels and show that a dramatic high level occurred approximately 8000 y

ago (which corresponds to the most recent period of glacial maximum) on all continents in these latitudes. Current lake levels are significantly low relative to the 8000-y levels.

On the basis of these data, we decided to study Lake Malawi, Lake Tanganyika, and Lake Turkana, all in central Africa. Our preliminary study of these lakes indicates that they experienced dramatic changes in water chemistry and lake levels, although many of the chemical changes do not directly correlate with the period of glacial maximum. More detailed work is under way to determine the extent of the chemical changes and their absolute ages.

As a means of testing models of past changes in lake systems, we are also conducting studies to evaluate how present-day systems may respond to climate change. After consulting with researchers at the Jet Propulsion Laboratory in Pasadena, California, we selected lakes in eastern California for detailed infrared

and near-infrared spectral studies using very high-resolution airborne spectrometers. These lakes are seasonal and preserve a pattern of evaporite minerals that may, on the basis of geochemical models, provide insight into the present-day response of weathering processes to climate change.

Initial matching of spectral properties with evaporite mineralogy suggests that such remote-sensing techniques may provide more detailed information concerning the distribution of mineral phases than has been possible in the past. They also may provide a sensitive indicator of changes in the chemical features of weathering that may result from present-day changes in climate. ■

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- Street-Perrott, F. A., D. S. Marchand, N. Roberts, and S. P. Harrison (1989), *Global Lake-Level Variations from 18,000 to 0 Years Ago: A Paleoclimatic Analysis*, Department of Energy Technical Report TR046 (DE90-000998), Washington, DC.

The Energy Program, which draws its personnel from various disciplines, oversees all nonfusion energy research at LLNL. Although the Program's emphasis has changed over the years, its mission has remained the same: to conduct research in a broad range of technical areas to help ensure that the nation will have adequate energy supplies at competitive cost. Our effort is focused on those promising but as yet unproven technologies that have been avoided by the private sector because of the long lead time from the research effort to the entrance of the product or technology into the marketplace.

Environmental issues increasingly influence the directions taken by the Energy Program because they ultimately affect the cost of the product energy. In FY90, we continued our research in fossil, nuclear, and renewable energy and began a concerted effort in advanced research for hazardous waste cleanup, minimization, and treatment. The ER&D Program funded two key projects in these areas.

In the first project, we began to develop an economic model of the U.S. energy system. The Lawrence Laboratories Energy Model (LLEM) model, developed in collaboration with Lawrence Berkeley Laboratory, can project changes in the types of resources and technologies used in the energy system as well as the resulting environmental impact of energy production and use.

The LLEM is a single-region model of the U.S.: each technology and end use represent that technology or end use for the entire country. The LLEM uses 85 energy conversion technologies, 27 end uses, and 13 energy resources. We also can add other technologies as needed. Ultimately, we want to use the LLEM to evaluate the long-term impact of government policies for reducing greenhouse gas emissions.

In the second project, we are studying the structure and function of natural enzymes associated with selected bacteria that can convert methane to methanol. Methane is an attractive alternative energy resource with large proven reserves. At present, no economically viable technologies are available to process methane directly to liquid fuels. Our research is aimed at developing a catalytic partial oxidation reaction of methane. By coupling inorganic catalysis research with biotechnology, we can study the enzyme proteins that certain bacteria use to biocatalyze the formation of methanol from methane and oxygen. In FY91, we will continue to develop this strategically and economically important technology.

In FY90, the Energy Program received \$1.2 million in ER&D funding. ■

Microbial Methane Conversion Enzymes

M. W. Droege and R. T. Taylor

The direct conversion of natural gas to methanol requires research aimed at developing a catalytic partial oxidation reaction of methane. We are coupling inorganic catalysis research with biotechnology by studying in detail the enzyme proteins that certain bacteria use to biocatalyze the formation of methanol from methane and oxygen.

Liquid transportation fuels are a critical energy commodity that affect nearly every sector of U.S. economy and account for 27% of the total U.S. energy consumption. The transportation sector alone uses more oil than the nation produces. Consequently, it is strategically and economically important to develop new technologies that produce liquid transportation fuels from fossil-energy resources other than crude oil. An attractive alternative energy resource with large proven reserves (both in the U.S. and in non-Middle

Eastern countries worldwide) is natural gas, or methane. However, no economically viable technologies are available to process natural gas directly to liquid fuels.

The future for methane conversion and natural gas chemical processing will depend on the development of catalyzed routes that directly convert methane to liquid fuels. Noncatalyzed, simple thermal reactions of methane and oxygen require very high operating temperatures (500 to 800°C) and result in unacceptably low product yields as a result of the uncontrolled overoxidation of the methane to carbon oxides. The key to natural gas conversion is the controlled catalytic activation of methane, which prevents overoxidation.

A select group of aerobic soil and water bacteria called methanotrophs can efficiently and selectively use methane at ambient temperatures as the sole source of their energy and carbon for cellular growth (Figure 1). The first reaction in this metabolic pathway is catalyzed by enzymes, called methane monooxygenases (MMOs), that form methanol.

Methanol is a technologically important product from this partial oxidation of methane because it can be converted easily to common liquid hydrocarbon transportation fuels (such as gasoline), used directly as a liquid fuel or fuel additive, mixed with jet or diesel fuels, or used as a feedstock for producing chemicals, such as the octane-boosting additive methyl tertiary-butyl ether. Naturally occurring biocatalyst MMOs accomplish a technologically important transformation (methane directly to methanol) for which there is presently no analogous chemical (nonbiological) process.

The emphasis of this research is to develop new catalyst materials that facilitate the overall transformation of methane to more valuable liquid products. We are using the above biocatalytic (enzymatic) process as a reference point in developing discrete chemical catalysts (biomimetic catalysts) that can convert methane to more desired end products. The advantage of this approach is that information on catalyst structure and function needed to rationally and systematically develop new catalysts for methane conversion is obtained from a biocatalytic system that is already performing a desirable transformation of methane.

Our research is divided into two closely integrated efforts. The first is an investigation of two distinct, but related, methanotroph enzymatic systems that appear to use differing metal cofactors for the partial oxidation of methane to methanol. This work is designed to elucidate a sufficiently detailed biochemical description of the structure and function of

Figure 1. Pathway of methane catabolism in methanotrophs. The numbered reactions are carried out by the following enzymes: (1) methane monooxygenase; (2) methanol dehydrogenase; (3) formaldehyde dehydrogenase; and (4) formate dehydrogenase.

the catalytically active site or sites to serve as a guide in the development of biomimetic materials. The second effort consists of synthesizing and subsequently determining the chemical reactivities of discrete complexes that structurally mimic the methane-active sites of the MMO enzymes. These materials will then be optimized for use in a process for converting natural gas to methanol.

Current work on the first research effort is focused on studying the MMOs, which are biosynthesized by the methanotroph *Methylosinus trichosporium* OB3b (Figure 2). This microorganism is being cultured in sterile, minimal-salts media using methane gas as the sole carbon substrate. Small volumes of single-colony subcultures have been frozen at -80°C and serve as seed inocula for future experiments. We generated shake-flask (250-mL) growth kinetic curves for this microorganism to optimize its growth rate and to maximize the amount of intracellular MMO enzyme activity in preparation for the subsequent increased mass productions of cells required for enzyme isolations. We have also studied the influence of a variety of cell-culture parameters on the intracellular location, form, amount, and broken-cell extract stabilities of the MMO catalytic activity. This work has led directly to larger-scale production of the bacteria using 5-L fermentation bioreactors for culturing gram quantities of cellular biomass in both batch and continuous modes. Recent pilot-scale work has led to a streamlined purification of the soluble MMO enzyme-hydroxylase-protein A from small quantities of broken cell extracts using a two-step procedure that consists of molecular size filtration and anion-ion exchange chromatography.

M. trichosporium OB3b can produce two forms of MMO, depending on the culturing conditions. Ordinarily, it forms varying amounts of both forms. For detailed comparative biochemical and spectroscopic studies of each form's active site, we needed the capability to generate consistently large amounts of cells that contain only one form of the MMO biocatalyst. Copper-ion and substrate gas (CH_4 and O_2) concentrations in the culture medium are key determinants of the MMO form and amounts produced by this methanotroph. Thus, in FY90, we devised minus-copper-ion (limited CuSO_4 added to culture medium) bioreactor growth conditions to achieve *M. trichosporium* OB3b cell densities of ~ 5.4 g dry cell weight/L (equivalent to ~ 20 g wet cell weight/L or $\sim 1 \times 10^{13}$ cells/L). We maintained high levels of the exclusively soluble form of MMO activity, even after approximately 150 to 200 h of culturing. We found that nitrate-ion is the limiting component of the culture medium that causes the cells to shift from a fast to a slow bioreactor growth rate (Figure 3); ionic-iron appears to be the limiting component that determines the maximum final cell density in a batch-mode bioreactor. The growth conditions we developed permit the rapid accumulation of fresh cells with soluble MMO for protein isolation and whole-cell nonaqueous catalysis experiments.

We have almost completed work to obtain high bioreactor cell densities of plus-copper-ion-grown *M. trichosporium* OB3b (excess CuSO_4 added to culture medium) in which all of the MMO is in a membrane-attached particulate form, with perhaps a copper-ion alone or copper-plus-iron ion at its active site. This

has required careful monitoring of the level of the CuSO_4 culture medium during the exponential bioreactor growth phase in a batch mode, followed by a shift to growth

Figure 2. A typical phase-contrast microscope view of the methanotrophic bacterium *M. trichosporium* OB3b taken at 1200 $\times$ magnification (bacterium is about 0.5×1 to $2 \mu\text{m}$).

Figure 3. Plot of batch-mode bioreactor cell growth (black line; measured by turbidity/light absorbance) and culture-medium nitrate-ion consumption (orange line; measured with an ion-specific electrode) for *M. trichosporium* OB3b grown in a minus-copper-ion culture medium.

in a continuous-culture mode to control the ratio of input-flow CuSO_4 -containing culture medium to output-flow cellular biomass. In the course of this work, we found that chloroform is a useful substrate for distinguishing between the soluble and particulate forms of MMO, i.e., in washed, intact cell suspensions. It is an active substrate for *M. trichosporium* OB3b cells carrying the soluble form—but not the particulate form—of this biocatalyst. We developed a $[^{14}\text{C}]\text{CHCl}_3$ -based radiotracer assay to measure this substrate activity quantitatively. The assay showed that $^{14}\text{CO}_2$ was formed stoichiometrically as the final oxidation product.

We have placed an equal emphasis on parallel studies of the membrane-bound particulate MMO and the iron-containing soluble MMO as

a result of the following important findings made during FY90:

(1) CuSO_4 not only regulates the biosynthesis of particulate MMO, but it also directly stimulates particulate MMO activity during assays of methane (or propene) oxidation. This strongly suggests that although copper is definitely not involved in the catalysis of soluble MMO, it plays some critical cofactor role at the particulate MMO active site. (2) In addition to chloroform, isobutylene and toluene are also very active substrates for the soluble form of *M. trichosporium* OB3b MMO but not for its particulate MMO. Moreover, current work with deuterium-labeled as opposed to hydrogen-containing methane, isobutane, and toluene is showing some remarkable kinetic primary isotope effects that demonstrate

clearly that C-H bond breaking is the rate-limiting step in MMO catalysis, except when multiple substrate-binding orientations result in more than one alcohol product.

Collectively, our observations strongly indicate that some important fundamental structural and perhaps functional mechanistic differences exist between the active sites of the two forms of MMO that can be synthesized in a single methanotroph. A dual comparative investigation of these differences is therefore critical for the MMO chemical-mimic modeling effort.

The objective of the second research effort is to develop biomimetic (nonbiological) chemical catalysts for methane oxidation. The biocatalyst systems relevant to methane oxidation are large proteins that can be viewed as highly elaborate coordination complexes in which the metal-containing "active site" is the center of electron transfer, substrate binding, and catalysis. The soluble form of the MMO enzyme consists of three protein components: A, B, and C. Protein A is the hydroxylase component that reacts with methane and oxygen, and it contains iron ions at its active site. Although some of the details vary, the general description of the iron site in protein A is a binuclear cluster containing some type of μ -oxo ligand between the iron atoms. The remaining ligands (derived from adjacent amino acid residues) contain either nitrogen or oxygen, and the iron-iron distance is between 0.30 and 0.35 nm. Our approach is to synthesize inorganic and organic chemical models of the biocatalyst (MMO) active site.

In FY90, we focused on the synthesis of a family of binuclear chelating ligands that possess a potentially bridging alkoxo group as well as aliphatic and benzimidazole nitrogen-coordination groups. Molecular

Figure 4. Synthesis pathway leading to the binuclear ligand $(\text{BzIm-CH}_2)_2\text{-N-CH}_2\text{-CH(OH)-CH}_2\text{-N(CH}_3\text{)-BzIm}$ (BzIm = benzimidazole).

models and energy minimization calculations confirm that this ligand can adopt a coordination geometry that incorporates two metal ions. The first member of this family that has been synthesized is $(\text{BzIm-CH}_2)_2\text{-N-CH}_2\text{-CH(OH)-CH}_2\text{-N(Cl i}_3\text{)-BzIm}$, where BzIm is benzimidazole. It is synthesized in five steps with acceptable yield (Figure 4). We have fully characterized this ligand and deduced its structure using proton NMR studies. We have synthesized several new coordination complexes containing copper ions and isolated them as crystalline solids using the above ligand system. These complexes are synthesized by stoichiometric reactions in methanol using hydrated metal salts containing BF_4^- counterions. The blue or blue-green metal-containing complexes are formed rapidly and are isolated by vapor diffusion using diethyl ether. The complexes have been characterized by

elemental analysis, and preliminary studies indicate that depending on the reaction conditions, either mononuclear or binuclear complexes can be obtained. Additional characterization studies including single-crystal x-ray studies are in progress.

Future work on the MMO enzymatic systems includes more isolations of soluble protein A and the copper-ion-induced particulate fraction plus a more complete biochemical characterization of each by suicide-substrate labeling experiments. In addition, studies using these materials aimed at probing their metal-ligand binding structures, reaction mechanisms, and other general properties of the catalytically active sites in these two biocatalyst materials are also planned. A fundamental part of our research into the development of biomimetic

catalysts is to understand how the active sites in these apparently functionally similar yet distinct MMO systems facilitate C-H breakage in combination with the activation of molecular oxygen.

In addition, we will continue to develop new binuclear ligand systems and to explore the incorporation of Co^{2+} , Fe^{3+} , Cu^{2+} , and Zn^{2+} metal ions into our biomimetic ligand systems. We will then begin to probe both the structural requirements and reaction pathways important for catalysis. Initial studies will focus on the addition of oxygen to the binuclear complexes. In particular, we are seeking complexes that irreversibly bind oxygen and activate it for further reaction (such as with methane). ■

Modeling the U.S. Energy System

A. D. Lamont and R. Koopman

We are developing an economic model of the U.S. energy system that can more effectively evaluate the long-term impact of government policies on reducing greenhouse gas emissions.

The Energy Modeling Project is developing an economic model of the U.S. energy system to evaluate how government policies affect long-term investment and operating decisions within the energy industry, a capability currently not provided by existing DOE policy models. The Lawrence Laboratories Energy Model (LLEM) can project changes in the types of resources and technologies used in the energy system as well as the resulting environmental impact of energy production and use. It is a collaborative project with the Energy Systems Division at Lawrence Berkeley Laboratory (LBL), a leader in developing models and analyzing energy end-use patterns both in the U.S. and in developing countries. In this project, LBL focuses on end-use technologies and economics, and LLNL focuses on energy-supply technologies.

In the late 1970s and early 1980s, LLNL developed a state-of-the-art modeling system and implemented several detailed models of the U.S. energy system to investigate the market penetration of new technologies and to evaluate the impact of these technologies on the environment (Rousseau et al., 1978). The current Energy Modeling Project uses essentially the same approach to model the various energy systems.

The LLEM is a single-region model of the U.S.; each technology and end use represent that technology or end use for the entire country. The LLEM includes 85 energy conversion technologies, 27 end uses, and 13 energy resources. Additional energy technologies can be added as needed.

In FY91, we will use the LLEM to evaluate several tax- and market-based strategies for controlling greenhouse gas emissions from the energy system.

Role of Modeling in Analyzing National Energy Policy

During FY90, the DOE has been working on a National Energy Strategy with certain objectives (DOE, 1990): energy should be inexpensive and environmentally safe, and the energy supply should be relatively secure from disruption by other governments. Unfortunately, these objectives conflict because it is not cheap to expand domestic energy supplies and develop environmentally clean energy. A policy must be formulated that strikes a balance between these goals, and modeling is needed to help evaluate whether a proposed policy provides that balance.

Several energy policies, some of which have been used in the past, are being considered. For example, a proposed tax on gasoline or the Btu content of all fuels would reduce the demand for energy and encourage investments in more efficient end-use technologies. A tax on imported oil would tend to raise domestic prices for oil, encouraging more efficient end uses—much like a tax on all energy—as well as the development of domestic oil

resources. In addition, taxes could be used to reduce CO₂ emissions from the energy system. Facilities could have unrestricted emissions but would have to pay a tax on every pound, providing an economic incentive for self-regulation.

Market-based strategies for controlling emissions have also been proposed. For example, the government could issue permits to energy producers allowing them to emit a certain amount of pollutants. These permits could be bought and sold by producers, but the government would control the number of permits and thus the total amount of emissions. Presumably, those facilities that could reduce their emissions inexpensively would do so and sell their excess permits to other facilities that could not reduce emissions so cheaply. In principle, the energy system as a whole would efficiently reduce its emissions to the desired level under this scheme. Other government policies could explicitly constrain the amounts of emissions permitted or the technologies used.

For some end-use technologies, efficiency standards could be mandated, such as the Corporate Average Fuel Efficiency standards imposed on the auto industry. Short of efficiency standards, the government could also require appliances and cars to carry labeling information about their efficiencies to encourage buyers to consider the life-cycle costs of the product.

Finally, the government could influence the energy system through its research and development policies. These range from the basic research needed to create new technologies to helping technologies to penetrate the market through preferential tax treatments or subsidies.

The LLEM can address the long-term impacts of each type of policy, and we will use it in FY91 to evaluate alternatives.

Economic Theory of the Model

In the long term, we are concerned with the impact of government policies on the structure of the energy system and changes in the resource base. Investment decisions made by energy producers and consumers determine the structure of the energy system by determining the capacity and types of energy technologies and the resources that are used. These investment decisions are strongly influenced by the economic environment (market prices, taxes, regulations, technology costs, demands) surrounding individual producers and consumers. The LLEM captures the impact of government policies on the energy system by modeling their impact on these decisions.

The energy economy is modeled as a network with final demands for energy services at the top and resource extraction at the bottom. Figure 1 shows a simple network. The nodes of the network represent energy conversion processes, resource extraction, and end uses. Additional nodes represent markets for energy commodities such as crude oil or electricity. Each node includes a small model of the costs of production and the investment decision-making relevant to that type of technology or activity.

The input data to the end-use nodes include a trajectory of energy demand through time derived from econometric forecasts. Using LLEM, we have generally extrapolated from the forecasts used by the Energy Information Agency in their analyses. For conversion nodes, the data include the efficiencies of production and the capital and operating costs.

For resource nodes, the data include estimates of the resources available and the costs of developing each increment. As we will show, the input for each node also includes other data needed to represent the impact of government policies on the investment and pricing decisions for the firms represented by the node.

The trajectory of international oil prices through time must be supplied as an input to the model. This is essentially equivalent to assuming that U.S. domestic energy policies do not have a significant long-term impact on the world oil market. We can, however, study the effect of international oil prices on the performance of policies by varying the assumed trajectory of the international oil price.

This set of input data provides enough information to project how the energy demands will be met

and what the costs will be. More specifically, the model finds trajectories of prices for each commodity, the quantity of each commodity produced, and the capacity expansions in each resource and conversion process.

The key to finding these trajectories is the concept of market equilibrium. In this model, technologies compete to supply markets for energy commodities (e.g., coal-fired power plants compete with nuclear and oil-fired plants to provide base-load power in the electric market). When a technology is competing with other technologies to supply a market, the amount that the market demands from the technology is inversely proportional to the price that the technology charges. At the same time, the higher the price that a technology charges, the greater the quantity that it can provide

Figure 1. The energy system is modeled as a network of energy flows. The demands for energy services and the costs of production and resources are the principal inputs to the model.

because it can cover higher fuel and operating costs. We find equilibrium when the market price of a technology evokes a demand that is exactly equal to the amount that the technology can supply at that price. Within an economy, we can find a set of prices for all technologies in all markets such that the demands and supplies are balanced.

The model assumes that the economy follows an equilibrium trajectory through time. Although the economy is seldom in complete equilibrium, in the long term (e.g., 50 y), it will tend to oscillate around equilibrium so the equilibrium assumption is a reasonable one for projecting a long-term average trend.

The system uses an iterative algorithm to find equilibrium prices and quantities. Each iteration requires a computational pass down through the network and a pass back up. Following through these two passes illustrates how the model determines the prices and quantities and how it models the impact of government policies on investment and operating decisions.

The "down pass" computes quantities produced by each process. At the top of the network is a demand for final energy services (e.g., ton-miles of transportation). The energy conversion nodes that provide these services convert a fuel into service. In Figure 1, a diesel truck converts distillate fuel into ton-miles of transportation. The amount of fuel required is computed from the end-use demand and the efficiencies of the conversion process. Working down through the network, we encounter a market that aggregates the demand for a commodity from several users (e.g., diesel trucks,

buses, aviation, and heating). The market is supplied by several suppliers, in this case by both local production and imports. Within the market, demand is allocated between the various suppliers on the basis of their prices.

The demands for commodities are propagated downward through various processing steps. In this example, refined petroleum products are produced from crude oil by the refining process. The crude oil is supplied to the refinery through a market for crude, which, in turn, is supplied by both imports and local production. At the bottom of the network, the total demand for crude oil (or any basic resource such as coal) reaches the resource processes.

The "up pass" starts from the resource nodes. At each resource node, the model tracks the total resource that has been committed, the production that can be obtained from the committed capacity, and the costs of producing from the committed capacity. Inputs to the model include estimates of the resources available and the costs of exploiting each increment of each resource. With this information, the model computes the cost of developing and producing from the next increment of the resource. The resource owner must, of course, recover at least this much in order to produce. In addition, he must recover enough to cover all costs that might arise from proposed government policies. These could include taxes, depletion allowances, lease payments, and costs of environmental regulations. The model does a standard discounted cash-flow analysis to determine the minimum price that the resource owner must charge to cover all costs associated with opening enough new

capacity to meet the demands that have been passed down to that resource process.

The required price is passed up to the next level in the network. In Figure 1, the next node is a market node. The market node computes an average market price from the prices of the suppliers (their required prices need not be exactly equal). This average price is passed on to the nodes above.

At each energy conversion process, the producer's required price for the output is determined by the total cost of production, which includes the price of the input (e.g., crude oil or diesel fuel), the other operating costs, and the capital cost of new capacity (if new capacity is needed). These prices are propagated up through the network until the final demand process is reached.

The end-use demand nodes contain demand functions for the final energy service. This function computes a total quantity of the energy service demanded given the price that has been passed up to it (the greater the price, the smaller the demand). This quantity is then passed back down the network in the next down pass. The algorithm repeats the two passes, making slight adjustments each time until the prices computed on the up pass are in equilibrium with the quantities demanded on the down pass.

The model accounts for the evolution of the energy system by modeling the capacity additions or retirements for each process. The algorithm tracks the capacity at each conversion process, accounting for retirements since the previous period. If the quantity demanded is greater than the capacity, the economic feasibility of adding capacity is evaluated. This is done by computing the discounted present value

of a new-capacity addition, taking into account the capital cost of new capacity, price of the output, price of inputs, and other variable costs. Again, these costs can include any costs or subsidies that arise from government policies. Over time, we can track the market penetration of a technology by tracking the additions to its capacity. We can also track the financial impact of government policies by accumulating the total capital investment made by energy producers over time.

Modeling the Impact of Environmental Policies

Environmental emissions are modeled as a function of the output from each conversion process so that total emissions can be tracked over time. The model also incorporates the cost of an emissions permit in the financial calculations for setting prices and making decisions about expanding capacity for the technology. This machinery allows us to analyze three basic policies: business as usual, taxes on emissions, and an auction for emissions permits.

Under a business-as-usual policy, permits are free and unlimited. The model tells us what will happen without any policy changes. A tax on CO₂ emissions is modeled by placing a price on the permits with no quantity restrictions. The cost of this tax then becomes one of the costs of production for an energy producer and will affect decisions about pricing and additions of new capacity. Finally, to model a simple auction for permits, we note that the purpose of the auction is to limit the total amount of emissions to a particular amount. In the real world, this could be done by limiting the number of permits and auctioning them to energy producers. We can simulate such limits in LLEM by varying the price of the permits until the total number demanded is equal to our target amount.

Future Investigations with the Model

During FY91, we will analyze policies regulating CO₂ emissions, specifically focusing on how taxes

affect carbon emissions. The goal of these analyses is to determine the level of tax required to achieve a given reduction in carbon emissions and the corresponding impact on prices and the structure of the energy system. We anticipate a major shift from high-carbon fuels toward low- or noncarbon fuels. The essential questions are how quickly can this occur, and what will be the impact on energy prices and the capital investment required by the energy sector. ■

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Engineering's ER&D effort provides the enabling technology base for future program requirements. We must anticipate these requirements so that they are sufficiently developed when needed. In FY90, the ER&D program funded four projects in Engineering.

The goal of our Atmospheric Compensated Imaging Systems Project, which began in FY89, is to develop the capability to remove atmospheric turbulence effects from telescope images, thus permitting astronomical objects and satellites to be imaged at the diffraction limit of the collecting telescope apertures. In FY90, we conducted a successful series of experiments at the Air Force Maui Optical Station (AMOS) on Mount Haleakala, Maui. We extracted diffraction-limited features of several satellites, including the Hubble Space Telescope, and imaged objects as far out as 1000 km.

In another project, we are studying the limitations of backward-wave oscillators (BWOs). Our initial goal was to duplicate the experimental work of Soviet scientists and determine if BWOs could produce very-high-power (1-GW) microwaves at high frequencies (9.5 GHz). We found that BWOs do function as oscillators when driven by relativistic electron beams at approximately the expected frequency. However, we were unable to duplicate the high-power conditions reported by the Soviets.

We also are developing a computer model to characterize the formation of superplastic materials and the circumstances under which they deform and fail. Superplastic materials are made by deforming crystalline solids in tension under specific temperatures and strain rates to unusually large plastic strains (often well in excess of 1000%) at low flow rates. Crystalline solids can be deformed close to the desired dimensions, which greatly reduces machining and material costs and minimizes the amount of scrap produced. Unfortunately, during deformation, ultrafine grains can grow and cavities can form (a process called cavitation), resulting in the loss of superplasticity. Once developed, our computer model will be incorporated into finite-element codes to study problems in forming superplastic materials and to provide real-time process control while they are forming.

In the final Engineering report, we discuss our experiments to better understand the causes of friction that occurs at the interfaces of micro-machinery. We constructed polysilicon cantilever beams with end sliders to measure friction at low loads.

In FY90, Engineering received \$691,000 in ER&D funding. ■

EY 90: \$290,000

Atmospheric-Compensated Imaging Systems

T. W. Lawrence and D. M. Goodman

A telescope of aperture D imaging at a center wavelength λ has a theoretical angular resolution of λ/D , the diffraction limit imposed by the size of its aperture. However, ground-based telescopes must view objects through the atmosphere, which is a turbulent, randomly inhomogeneous medium. Local temperature gradients in the atmosphere create random phase fluctuations in the propagation path that can be characterized by a correlation scale length r_0 . A normal long-exposure image contains many realizations of this random fluctuation. The net effect is a blurring of the image, which results in a resolution of λ/r_0 . Thus, resolution is degraded by a factor of D/r_0 and is limited by the atmosphere, not the telescope. For example, the world's largest telescopes, such as the 5-m Hale telescope on Palomar Mountain, California, have a theoretical diffraction-limited resolution of about 0.1 μ rad at visible

wavelengths. However, the correlation length of the atmosphere at visible wavelengths is typically 10 cm; consequently, the resolution of these telescopes is degraded by a factor of 50 (5 m/10 cm). The resulting resolution is 5 μ rad, no better than that of most amateur telescopes.

This limitation applies to all ground-based telescopes and severely hampers our ability to image distant objects. However, recent advances in short-exposure-imaging technology, new computer postprocessing algorithms, and low-cost wavefront correction systems provide methods for imaging astronomical objects and artificial satellites to the diffraction limit of the telescope instead of that imposed by the atmosphere. The impact on large telescopes is immediate. Sparse optical arrays hundreds of meters in diameter can be constructed to image higher-altitude satellites, which could revolutionize ground-based astronomy. The techniques we are developing can also be applied to problems in which random distortions must be removed from a signal or image.

Our approach is based on non-standard imaging techniques used by radio astronomers. The turbulence-induced effects of the atmosphere on an object image can be frozen if the exposure time is reduced to less than the correlation time of refractive-index fluctuations (1 to 10 ms, depending on wind velocity). The instantaneous effects of the atmosphere make a short-exposure image appear to be a superposition of randomly placed copies of the object. The result is called a speckle image because the distortion patterns are analogous to

speckle observed with coherent laser illumination. Methods for extracting object information from a sequence of speckle images are called speckle interferometry and have been used to image star clusters. However, our results are the first to demonstrate that such algorithms, coupled with appropriate hardware, are successful at imaging extended objects, such as satellites, through the atmosphere.

To enhance the speckle-imaging technique, we are also pursuing low-cost technologies that partially compensate the incoming wavefront in real time. The techniques include a magneto-optic spatial light modulator (MOSLM) to correlate the incoming wavefront with an orthogonal basis set of aberration functions (e.g., Zernike polynomials) and compensation with a modal adaptive optic (possibly another MOSLM). We are also considering advanced systems that use both speckle techniques and adaptive optics with new remote-sensing systems for measuring atmospheric perturbations.

Progress

Over the past 3 y, we have put together an impressive technology for imaging space objects to the diffraction limit of earth-based telescopes. We have demonstrated that speckle imaging of satellites can provide enormous help in identifying space objects. We also have developed a comprehensive numerical simulation of our technique and assembled the hardware necessary to conduct a horizontal-path-imaging experiment using a charge-coupled device (CCD)-based speckle camera (Lawrence et al., 1988, 1990);

Johansson et al., 1990; Massie et al., 1990). In addition, we have launched an effort toward a national program aimed at implementing this technology. Our work in FY90 culminated in a systematic investigation of speckle-image reconstruction. With the speckle images obtained during an experiment in Maui and the emerging Atmospheric Compensated Imaging Systems program funded through the Strategic Defense Initiative Organization, we have achieved our original goals.

New Algorithms

We have implemented three state-of-the-art algorithms used by the astronomical community:

- The Labeyrie technique for Fourier magnitude estimation.
- The bispectral (or triple-correlation) technique for Fourier phase estimation.
- The Fienup Fourier iteration technique for imposition of object support and positivity constraints.

We have applied a combination of these techniques to reconstruct objects representative of low-earth-orbit (LEO) satellites. However, when we turned our attention to fainter objects representative of high-earth-orbit (HEO) satellites, we realized that bias effects due to photon and additive noise on the CCD are important.

Therefore, we studied the bias effects of additive noise and derived an analytical expression for bias in the power spectrum and bispectrum. In modifying the algorithms to compensate for such bias, we demonstrated that high-quality reconstructions of dim HEO satellites are possible. However, because these algorithms were not optimal in a statistical sense and did not efficiently incorporate known

constraints, we developed new ones (Goodman et al., 1990).

Our first algorithm was developed to improve estimates of the image phase. It computes the bispectral phase and uses a linear least-squares model to fit the image phase to the bispectral phase. Although the matrix for the linear model is large, it is also sparse; therefore, we used a standard linear conjugate gradient algorithm to compute the phase estimate in a reasonably short time. Then we used the original integration method to resolve the 2π ambiguities in the bispectral phase. This method worked well at moderate to low noise but broke down at higher noise levels.

Our second algorithm used projection onto convex sets (POCS) to improve estimates of the image magnitude. The true power spectral density of a diffraction-limited object is nonnegative and zero at frequencies beyond the diffraction limit. The inverse transform is the object autocorrelation function, which also is nonnegative.

Furthermore, the autocorrelation has finite support because the object has finite extent. Each of these properties of the true power spectrum is a convex constraint on the set of possible power spectra. The estimate for power spectral density obtained by averaging over speckle frames is corrupted by noise and does not satisfy all of these properties.

Because the constraints are convex, we can apply the iterative POCS algorithm to project the estimate obtained from averaging onto the constrained set of possible power spectra. The iteration is similar to the Gerchberg-Saxton algorithm in which constraints are applied alternately in the spatial and frequency domains until convergence to a solution is obtained. Although this algorithm usually

improved image quality, the degree of improvement was problem-dependent and ranged from minor to dramatic.

Our third algorithm obtains the image phase from an estimate of its power spectrum. Because the bispectrum is a third-order moment, phase estimates may be noisier than magnitude estimates. Furthermore, the fact that the object image is nonnegative is important and should be used to reduce error variance. With the POCS algorithm, we used nonnegativity of the autocorrelation to improve the magnitude estimate but have not yet directly used object-image nonnegativity. We developed an improved algorithm that allowed for weighting based on variance of the averaged power spectrum and used a constrained, nonlinear, conjugate-gradient algorithm to compute the estimate. Our results yielded improved estimates, required less computational effort, and often produced crisper images with less "ringing."

Our ultimate goal is an algorithm that simultaneously computes phase and magnitude while allowing for proper weighting of both power spectral and bispectral data and the incorporation of all a priori information.

Speckle-Imaging Experiment on Maui

Given our new algorithms and a better understanding of the noise properties of speckle imaging, we modified our CCD-based speckle camera for an actual satellite-imaging trial. The CCD device was a 1320×1034 chip mounted in a liquid-cooled camera head. The chip was subarray addressable, and the additive noise was 14 electrons/pixel at a readout rate of 500 megapixels/s.

Figure 1 shows the new speckle camera mounted at a secondary focus of the 1.6-m telescope at the

Air Force Maui Optical Station (AMOS) facility. This telescope has excellent satellite-tracking capabilities and uses a fast-steering mirror to maintain image position on a narrow field. We imaged a variety of satellites at ranges out to 1000 km over 2 weeks.

Figure 2 shows images taken of the Hubble Space Telescope (HST) during a sun-lit pass over the AMOS facility. For comparison, a photograph of the HST is included at the top of the figure. Figures 2a, b, and c show sample raw CCD images at various points during the pass. Our camera obtained images at a rate of approximately 10 frames/s using a 150×150 subarray; during a typical 2-min pass, we acquired 1200 images. All images could not be used for the same reconstruction because the object can change its orientation during a pass. This effect is evident in the three reconstructions (Figures 2d, e, and f).

For each reconstruction, 50 frames were averaged, corresponding to about a 5-deg angular segment of the pass. The elevation, range, and sun angle (the angle defined by the observatory, satellite, and sun) at the center of each 5-deg segment are listed under each reconstruction. The field of view of each image is 7.4×7.4 arc s ($1 \text{ arc s} = 1'' = 1/3600 \text{ deg}$).

Comparison of the speckle images with our reconstructions shows that diffraction-limited details are obtained, including the solar-panel cross bars and two relay antennas of the HST. Furthermore, by using our pictures to measure the size of solar panels, antennas, and other features, we can assess the object's power needs and communication capabilities. Because such features are well known, the HST provided an excellent test for our techniques. Figures 3 and 4 show two other satellites before and after image processing, demonstrating diffraction-limited

detail and the power and versatility of our techniques.

New Wavefront-Compensation Technologies

Optical wavefront sensors also can be used in beam control, pointing and tracking, and image formation. Common to all is the determination of wavefront phase or amplitude. Phase is the most important parameter because it has the greatest effect on laser-beam propagation, image fidelity, image-reconstruction quality, and beam-pointing accuracy. Classical wavefront sensors fall into one of two general categories: the Hartmann sensor, and the shearing interferometer. Both types generate a slope or gradient field that can be used with standard numerical procedures to reconstruct the phase field.

The Hartmann device uses an array of lenses smaller than the full aperture to sample subapertures or patches of a wavefront and to bring each wavefront sample to a nominal, focused spot. It monitors the deviation or displacement of the centroid of the spots from their positions given a perfect, unaberrated wavefront. The sensor can create a raw point map of the gradient or slope measurement of the phase field.

A shearing interferometer samples subapertures or patches of the wavefront and divides or shears the samples into two or more components. These components are spatially separated over a calibrated distance. Translated samples are then combined (interfered) on a detector array. Self-referencing allows the shear-induced phase shift to be nulled, leaving the intrinsic phase difference to be detected. The intrinsic phase difference results in a point map that specifies the gradient or slope field of the unknown wavefront phase.

Both the Hartmann and shearing sensors are mature, well-developed instruments. However, they suffer from several limitations that preclude their use or extension to some problems of current interest:

- Shearing interferometers are usually tuned to a particular wavelength and cannot operate broadband.

Figure 1. LLNL camera on the Air Force Maui Optical Station 1.6-m telescope. (a) The 1.6-m telescope. (b) Speckle camera in position on the beamline. (c) Speckle camera optics.

- Both instruments produce slope or gradient information and not a direct phase map; thus, large amounts of computation must be done on each wavefront measurement.

- Aperture and subaperture geometries and dimensions are fixed.
- The mathematical geometry and grid spacings of masks, and the mathematical format of the reconstruction algorithms, are fixed in hardware because they must be high-speed.

To avoid these limitations and produce high-speed, flexible wavefront sensors, we had to meet two general design goals. First, we had to avoid phase-gradient generation and measurement schemes because they are computationally burdensome and analytically limiting.

Second, we had to avoid fixed, precision hardware that is nonalterable.

These goals led to a system concept based on a dynamically alterable, programmable electro-optics device that interacts with a wavefront and provides a direct phase measurement in the form of an electrical or optical signal suitable for immediate readout. This concept suggests a phase-measuring, two-dimensional optical correlator computer in which the phase structure is correlographically mapped. Such a wavefront sensor is by nature a two-dimensional parallel processor and avoids the computational burden of the Hartmann and shearing sensors. It measures phase structure directly with no time delay caused by digital computation or signal processing. This device could enable the construction of large-aperture, high-speed compensation systems that are flexible and low in cost.

During FY90, a prototype correlating optical wavefront sensor system was completed by one of our consultants at the Air Force Weapons Laboratory. The initial system was based on a serial optical correlator using a MOSLM. The apparatus functions as a wavefront sensor by passing an input-aberrated beam through the correlator while a quasi-static sequence of complex-function, computer-generated holograms (CGHs) are introduced by the MOSLM at the correlation plane of the system. If the CGHs are the complex-conjugate representations of a properly chosen set of orthogonal basis functions, the idealized output is a signal representing the relative magnitude of the aberration in question. Because the basis functions are orthogonal, the actual wavefront phase is given by the sum of the various functions weighted by these measurements. We demonstrated this technique with the prototype system.

Figure 2. Comparison of (a, b, and c) short-exposure (speckle) images with (d, e, and f) our reconstructions of the Hubble Space Telescope (HST). The field of view of each image is 7.4×7.4 arc s. Numbers below each set of photos show the range from the observatory to the HST; the elevation (angle between the HST and horizon); and sun angle defined by the sun, the HST, and the observatory.

Necessary procurements to build a duplicate system at LLNL have begun. When completed during FY91, the system will allow us to study the flexibility of the correlating optical wavefront sensor system and to analyze our ideas for extending the techniques to include wavefront compensation and measurement (as an adaptive optical system). Because the wavefront sensor directly measures contributions to a wavefront of various orders of aberration, the aberrations can be directly compensated using a modal (or order) adaptive optic. The adaptive optic could be another MOSLM operated in a conjugate mode.

Future Work

We have developed and demonstrated the capabilities of new image-processing techniques to resolve objects in space to unprecedented levels. We have begun an externally funded program aimed at developing atmospheric-compensated imaging systems. Many new technologies must be pursued to make our final system a success and to extend our techniques to astronomical imaging. We will continue to develop novel technologies, including new wavefront-sensor and adaptive-optic architectures, and to improve our algorithms for more stressful conditions. We also will conduct field experiments that improve our understanding of issues associated with imaging fainter objects. Finally, we will begin to design a multi-aperture demonstration telescope. This device will enable us to use compensated arrays with baselines of hundreds of meters and will facilitate high-resolution imaging of geosynchronous satellites and new features of astronomical objects.

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Figure 3. (a) Speckle image and (b) reconstruction of object s050706. Range = 917 km, elevation = 67 deg, sun angle = 80 deg, and number of frames = 100.

Figure 4. (a) Speckle image and (b) reconstruction of object s311508. Range = 701 km, elevation = 56 deg, sun angle = 50 deg, and number of frames = 50.

High-Power Microwave Generation Using a Backward-Wave Oscillator

J. F. Camacho, E. T. Rosenbury, B. R. Poole, and R. A. Anderson

We are investigating BWOs for high-power relativistic electron beams to determine their capability to generate high-power microwaves. Our goal is to develop a device that can operate in the millimeter-wave regime and to identify mechanisms that prevent further commutation

The Department of Energy and the Department of Defense are considering high-power microwave sources for use in various applications, including directed-energy weapons, high-gradient particle accelerators, plasma heating, and ultra-high-power radar communications and jamming, and for

microwave vulnerability and survivability studies. The backward-wave oscillator (BWO) is a strong candidate for producing these high-power (about 1-GW), high-energy (about 1-kJ), high-efficiency (about 10%) microwave pulses in the centimeter-wave regime.

Backward-wave oscillators belong to a class of microwave sources in which an electron beam drives a slow-wave structure. A typical BWO has a waveguide with walls of periodic corrugations (e.g., sinusoidal ripples). An annular electron beam is injected into the waveguide and guided by an axial magnetic field. Microwaves are generated via an instability in which the slow space-charge wave on the beam couples resonantly with the electromagnetic waveguide modes of the structure. The amplitude of the microwaves grows as a function of time because of the instability and eventually reaches some saturation level. Then the energy produced in the BWO can be coupled out of the device through an appropriate waveguide or antenna system.

Most high-power, high-energy BWO experiments have been conducted in the U.S.S.R., and we were interested in duplicating these experiments to verify the results. Soviet researchers observed 15 GW of output power at 9.5 GHz with 50% instantaneous efficiency using a multiwave Cherenkov generator (essentially a two-stage BWO) (Bugaev et al., 1990a). The energy in the microwave pulse was about 500 J. Using similar devices, they also produced microwave powers of 4.5 GW and 3.5 GW at 27 GHz and 44 GHz, respectively, both with about 500 J of

energy (Bugaev et al., 1990b) and an efficiency of 30%.

These results far exceed U.S. achievements. The most notable BWO experiment performed in the U.S. was conducted by researchers at the University of Maryland. They reported 400 MW of power at 8.4 GHz with 40% efficiency using a plasma-filled BWO (Carmel et al., 1990). The primary challenge at the higher power levels has been to extend the length and increase the energy of the microwave pulse, which terminates sooner than the electron-beam pulse that drives the BWO.

In FY90, we initiated the only current national laboratory effort to research and develop BWO technology. Our initial goal was to perform an experiment based on a BWO apparatus for which the Soviets reported 1 GW of microwave power at 9.4 GHz (Bondar et al., 1983). We hoped both to satisfy DOE intelligence interests and to use this experiment as the starting point in research to solve the problem of BWO microwave-pulse shortening.

During FY90, we developed PC-based computer codes to model and predict BWO performance. In addition, we extensively used CON-DOR, a 2.5-dimensional, relativistic, fully electromagnetic particle-in-cell (PIC) code on LLNL's Cray computer network. Our linear-theory analysis yielded information about the dispersion relation of the system, the required beam parameters, the operating frequency of the device, the growth rate of the microwave energy, and the conditions required to start oscillations. Analysis of the nonlinear physics gave us information about BWO

Figure 1. BWO microwave output power as a function of the injected beam current, where axial magnetic field = 2.2 T, beam voltage = 1 MV, and cathode radius = 8 mm. When the beam current was increased, power increased; however, we did not observe the level of efficiency indicated by our calculations or the results of similar Soviet experiments. Power may be limited by poor coupling between the BWO and waveguide system.

output power, pulse shape, and spectral content. Using this theoretical and modeling work, we established a foundation for analyzing and supporting our experiments.

In the first half of FY90, we designed and assembled our BWO apparatus and performed a series of beam-propagation experiments to ensure production of the required electron beam. Once we demonstrated satisfactory beam generation and transport, we began to generate microwaves. The experiment characterized the BWO's performance over a range of operating parameters, including microwave output power and frequency as a function of several beam parameters (current, energy, and cathode radius) and axial-magnetic-field strength.

Figure 1 shows microwave output power as a function of beam current at a fixed axial magnetic field, beam voltage, and cathode radius. As projected, these data demonstrated that power increased as the beam current increased. The direct measurement of the BWO microwave signal using a high-bandwidth oscilloscope has been a significant diagnostic accomplishment. Figure 2 shows these data, which verify that the BWO is operating as expected, predominantly at a single frequency.

Unfortunately, our BWO has not operated as efficiently as we anticipated. Our experimental results show about 200 MW of microwave power at an 8-kA beam current and 1-MV beam voltage, implying an

efficiency of 2.5%. Our models, however, predicted 200 MW at 1 kA and 1 MV (20% efficiency). Soviet results with a similar device also suggest that higher efficiency and power should be attainable. A key problem may be that the power generated in the BWO is not coupling effectively to the rest of the waveguide system where our diagnostics are situated.

In FY91, we will attempt to identify effects that may limit BWO performance, to optimize that performance through hardware changes, and to develop more advanced diagnostics to study microwave emission and beam behavior. Because the short pulse of our current electron-beam source (50 ns) may limit our investigation of the pulse-shortening phenomenon, we plan to design a new experiment using a beam source with longer pulses (about 100 ns). In addition, we will support our experimental work by extending our analytical and modeling capabilities, particularly in the areas of predicting important BWO parameters and understanding nonlinear BWO physics. ■

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Figure 2. Typical BWO microwave signal as a function of time during a 2-ns window in the microwave pulse. Power is at 8.5 GHz, and bandwidth is approximately 0.5 GHz. The predicted operating point is 8.1 GHz. We measured the signal directly with an electric-field probe situated at the wall of the waveguide section, which is located at the output of the BWO. To our knowledge, a measurement of this type has not been made on a BWO and is a significant diagnostic achievement.

Modeling Superplastic Materials

D. R. Lesuer, C. K. Syn, K. L. Cadwell, and S. C. Mance

We are developing a model that accounts for strain rate changes during superplastic flow and their effects on stress-strain behavior and deformation localization.

Superplasticity refers to the capability to deform crystalline solids in tension to unusually large plastic strains, often well in excess of 1000%. This phenomenon results from the ability of a material to resist localized deformation, much the same as hot glass. Superplastic materials also deform with low flow stress. Thus, materials with superplastic properties provide the opportunity to form complex components into shapes approaching final dimensions. This property greatly reduces machining and material costs and minimizes the amount of scrap produced.

Superplastic behavior depends strongly on the microstructure of the material (such as ultrafine grain structure) and deformation under the appropriate conditions of temperature and strain rate. Unfortunately, ultrafine grains can grow during deformation, resulting in the loss of superplasticity. Cavities can also form and grow during deformation (cavitation) and cause premature failure of a material. Thus, we want to gain a quantitative understanding of these processes and their influence on material forming. The objective of our project is to develop a model of the structural changes and their influence on stress-strain behavior and deformation localization.

Superplastic materials exhibit high elongations because a deformation mechanism known as grain boundary sliding (GBS) is active. The deformation behavior occurs within a relatively narrow range of strain rate. If the strain rate is too

high, a different mechanism called diffusion-controlled dislocation creep (slip creep) is activated, and ductilities are substantially reduced. If the strain rate is too low, then a deformation mechanism known as diffusional flow prevails, and the ductility is reduced relative to GBS. From a commercial standpoint, forming components at high strain rates is attractive because operations can be done with less time and cost. This often means superplastically forming at strain rates close to the slip creep regime. Our work is focused on the two higher strain-rate regimes—GBS and slip creep.

In FY90, we focused on assessing:

- The kinetics of strain-enhanced grain growth and cavitation during superplastic deformation.
- The ability of two rate-dependent constitutive laws to predict the stress-strain behavior when microstructural changes are considered.
- The ability of existing instability criteria to predict strain localization.

In FY91, we will combine these components to provide the desired model.

We are studying two materials that are representative of the two generic classes of superplastic materials. The first is a quasi-single-phase material, a copper-base alloy known as Coronze (Cu-2.8Al-1.8Si-0.4Co). The second is a two-phase alloy (Ti-6Al-4V). In FY90, we began obtaining stress, strain, and strain-rate data on these materials. Because superplasticity is sensitive to the microstructure of material and the testing parameters, we exercised considerable care in characterizing our materials and ensuring that we had good control over the testing variables. We

Figure 1. Stress and cavitated area fraction vs strain for Coronze, a dispersion-strengthened copper-base alloy, at two strain rates and at 550°C. Small cavities form during superplastic flow in many materials. The cavitation area shows an exponential dependence on strain; thus, over most of the strain history, the cavity growth is controlled by plasticity.

programmed our equipment to do testing under machine control. In addition, we obtained a furnace and related experimental hardware that minimizes temperature gradients in the sample (<1°C) and provides excellent temperature control.

Our studies of strain-enhanced grain growth and cavitation extend the work of Caceres and Wilkinson (1984) to include the effects of temperature and higher strain rates. Our studies on the copper alloy show that significant increases in grain size occur with strain and that the grain-growth kinetics show a strong inverse dependence with strain rate; however, temperature dependence is weak. In addition, the grain-size distribution is altered during deformation.

Cavitation is a complex process requiring nucleation, growth, and eventual interlinkage of cavities. The microstructure of the copper alloy consists of single-phase copper with a dispersion of hard, cobalt-rich particles. We observed that cavities nucleate at large particles, grow, and eventually coalesce by intense shear along planes of maximum shear stress. Because we are interested in modeling large plastic strains, we are primarily concerned with the hole-growth and interlinkage processes. Cavity growth should be plasticity controlled at intermediate and large strains; however, void coalescence and eventual fracture are much more complicated processes.

Figure 1 shows cavitation and stress-strain results for the copper alloy at two different strain rates. This figure shows the area fraction of cavities formed in a plane section as a function of strain. Over most of the strain history, hole growth is by controlled plasticity because the cavitation volume shows an exponential dependence on strain, which is typical for processes of this type. For modeling purposes, we are

concerned with the influence of cavitation on the flow stress and strain of the material. To a first approximation, we accounted for these small cavities by correcting for the loss in cross-sectional area. Figure 1 also shows the influence of two strain rates on the strain kinetics of cavitation. One strain rate was in the region of GBS; the other was in the slip-creep region. Deformation in the region of GBS shows substantially more cavitation. This result is expected because particles at grain boundaries are subject to intense deformation and, thus, void formation during grain-boundary sliding.

The rate-dependent constitutive laws we are studying for GBS and slip creep can be expressed (Sherby and Wadsworth, 1989) as:

$$\dot{\epsilon}_{\text{gbs}} = A_{\text{gbs}} \left(\frac{b}{L} \right)^3 \left(\frac{D_{\text{gb}}}{b^2} \right) \left(\frac{\sigma}{E} \right)^2 \quad (1)$$

and

$$\dot{\epsilon}_{\text{slip}} = A_s \left(\frac{\lambda}{b} \right)^3 \left(\frac{D_l}{b^2} \right) \left(\frac{\sigma}{E} \right)^8 \quad (2)$$

where $\dot{\epsilon}_{\text{gbs}}$ and $\dot{\epsilon}_{\text{slip}}$ are the strain rates for grain-boundary sliding and slip, respectively; A_{gbs} and A_s are constants; λ is the minimum barrier spacing governing slip creep (typically the interparticle spacing or the grain size); L is the grain size; D_{gb} and D_l are diffusion coefficients; b is the magnitude of the Burger vector; and E is Young's modulus. Both equations represent steady-state behavior and predict a constant flow stress for a given testing temperature and strain rate at constant microstructure.

We have accounted for microstructural changes during superplastic deformation by correcting experimental data for cavitation, as discussed above, and by substituting the strain-enhanced grain size into Equations (1) and (2). Increases in grain size during deformation cause strain hardening during GBS [Equation (1)] and strain softening during slip creep [Equation (2)].

Figure 2 compares experimental data for three strain rates with the

Figure 2. Stress vs strain for the copper-base alloy at three strain rates and at 550°C. For the two slower strain rates, the dominant deformation mode is grain-boundary sliding. For the faster-rate slip, creep is the dominant mode. Experimental data taken at a constant true strain rate are shown in orange. The stress-strain predictions of two rate-dependent constitutive laws with strain-enhanced grain growth are shown in black.

predictions of these equations. The two slower strain rates are in the GBS regime, whereas the fastest is in the slip-creep regime. In the GBS region, strain-enhanced grain growth and Equation (1) can account for the hardening observed experimentally. However, the analysis does not predict a maximum in the stress-strain curve and subsequent strain softening because grain growth continues until fracture. This strain softening in the GBS region may be caused by the

effects of cavitation; however, the volume of cavities when the softening process starts is quite small ($<5\%$, as shown in Figure 1). Other sources of softening may include a broadening grain-size distribution with strain and the effects of dislocation storage near the grain boundary. Additional analysis is needed to establish the source of softening during grain-boundary sliding in this material.

In the slip-creep region, strain-enhanced grain growth can account for some of the softening observed

experimentally. However, the experimental results clearly suggest that additional softening mechanisms are operating. Accurate modeling of these materials will require more thorough accounting for the softening effects. ■

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Fundamental Mechanisms Governing Friction at Low Loads

D. R. Ciarlo, J. A. Folta, W. J. Orvis, and R. S. Glass

We are performing experiments to better understand the nature of friction between two surfaces in relative motion at low loads.

Our goal is to understand the fundamental causes of atom-by-atom friction that occurs at the interfaces of micromachinery. We are experimenting with very low friction loads (sometimes in the nanogram range) as an extension of our work on the causes of friction in silicon-based microdynamic structures (Ciarlo et al., 1991). Although the friction forces between the moving parts of these structures are very low, they limit operational performance. We have based our studies on the work of Mate et al. (1987), who used an atomic-force microscope (AFM) to measure friction loads that were in the same range as those we are studying.

During FY90, we constructed polysilicon cantilever beams with end sliders to measure low-load friction coefficients. These sliders were attached to silicon-nitride substrates, which allowed us to measure friction between the two in a lateral direction. The areas of the sliders were $100 \times 100 \mu\text{m}$, and the loads were 50 ng. Using a microprobe to apply voltage to the sliders, we measured the force required to move them. Our preliminary calculations showed that the coefficient of friction between polysilicon and silicon nitride, the two materials most used in microfabrication, is 0.2, which is about as low as we predicted.

We have purchased an AFM and anticipate using it in FY91 to scan our surfaces before and after friction experiments. The scanning should determine whether friction is caused by surface roughness or by small

particles that are plowed into the surfaces during operation. Ultimately, we hope to find a method for lowering friction during the operation of micromachinery. ■

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Nuclear Chemistry

C. H. Poppe

The primary mission of the Nuclear Chemistry Division is to provide diagnostic information on the performance of nuclear explosive devices using radiochemical and isotopic techniques. As the Laboratory develops new programs, the Division responds by developing new applications of nuclear science and technology. We combine the chemical and physical disciplines—such as inorganic and physical chemistry, geochemistry, radiochemistry, and nuclear physics—with technological expertise in radiation detection, isotope mass spectrometry, and data analysis, for application to various LLNL programs, including Nuclear Test-Experimental Science, Nuclear Waste Management, Treaty Verification, Safeguards Technology, Environmental Protection, and Laser Isotope Separation. ER&D-sponsored fundamental and applied research in nuclear science and related disciplines has enabled us to develop the scientific and technical leadership needed to most effectively contribute to Laboratory programs.

Our FY90 ER&D projects address two broad research areas: basic research in nuclear (and atomic) science and the development and application of novel techniques for isotope cosmochemistry research.

In FY90, we completed the testing and calibration of a state-of-the-art electron spectrometer to measure the mass of the electron antineutrino and began taking data. Our experiment uses a gaseous tritium source to eliminate all known systematic problems associated with the solid-state interactions of tritium beta decay that occur in other experiments.

While calibrating the electron spectrometer with internal conversion electrons from ^{83}Kr , we found that it is a powerful tool for atomic physics measurements. We were able to directly measure the binding energies of the 2s, 3s, and 4s electrons for charge states up to 13^+ . In addition, we discovered a metastable state where a single 4s electron is left with vacancies in the 3d shell.

In a new experiment for FY90, we began to search for ^{244}Pu double-beta decay using radiochemical milking techniques. We have obtained a tentative result consistent with our predictions.

New projects in FY90 share the objective of exploiting emerging analytical capabilities to address basic research problems in isotope cosmochemistry. These research projects also have high potential for spinning off new applications in environmental research related to our thrust area in environmental radiochemistry. For example, we developed a scheme for dating young, surface geologic features using accelerator mass spectrometry and noble-gas mass spectrometry to make highly sensitive measurements of products of in situ cosmic-ray interactions. In addition, we perfected a new Re-Os radio-metric chronometer ideally suited to dating metal-silicate fractionations such as planetary core formation.

In FY90, the Nuclear Chemistry Division received \$402,000 in ER&D funding. ■

Search for Double-Beta Decay in the Actinides

K. J. Moody, R. W. Lougheed, and E. K. Hulet

Many even-even nuclides that are stable to beta decay are unstable to double-beta decay. In the double-beta decay process, a nucleus with neutron and proton numbers (N, Z) converts to the nucleus ($N-2, Z+2$) by emitting two beta particles with or without the coincident emission of two antineutrinos. Without the emission of antineutrinos, lepton number is not conserved in the decay process. The relative intensities of the no-antineutrino and two-antineutrino emissions test the validity of the gauge models (Vergados, 1983); consequently, considerable experimental effort is going into the study

of double-beta decay. The half-lives for double-beta decay are so long that they have only recently been directly determined (Elliot, 1987).

For our experiment, we chemically purified a 1.45-g sample of ^{244}Pu . This sample constitutes most of the U.S. supply. We adsorbed the plutonium on an ion-exchange column and stored it for 4 months. At the end of this ingrowth period, we isolated a curium fraction and simultaneously processed curium samples from two chemical blanks. The double-beta decay of ^{244}Pu results in the formation of ^{244}Cm in the sample. An excess of ^{244}Cm in the main sample over contamination levels found in the blanks is a signature for the process. From these solutions, we produced counting samples that we analyzed with low-background alpha detectors for more than 220 d.

We observed a significant excess of ^{244}Cm in the main fraction compared with a small amount of curium in the blanks. The gross ^{244}Cm activity in these samples corresponded to a ^{244}Pu double-beta half-life of $2.2 \times$

10^{16} y, which is orders of magnitude shorter than other known double-beta half-lives. However, because some curium was observed in the blanks, we cannot exclude the possibility of environmental contamination of the main fraction.

In FY91, we will clean the reagents and eliminate potential sources of contamination prior to a second milk of curium. With the second milk, using a longer ingrowth period, we expect to produce a publishable result. If we confirm it, we will undertake a large-scale collaborative experiment to determine relative proportions of the two-antineutrino and no-antineutrino components of decay. ■

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Rhenium-Osmium Isotopic Geochemistry

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In Re:Os chronometer development, we corrected problems with sample dissolution, completed calibration of isotopic spikes, and tested our mass spectrometry technique. Applications included analyses of iron meteorites, marine sediments, and Archean shales.

Rhenium-osmium isotopic analyses yield time and provenance information that can be used to study the early evolution of metals in the solar system, the evolution of the Earth's crust and mantle, sediment geochronology, and noble-metal marine chemistry.

Our first goal in FY90 was to test the possibility that our surprisingly high initial results for IAB iron meteorites stemmed from an unknown flaw in our separation procedures (Niemeyer et al., 1989). We found that our usual method of ensuring equilibration between an isotopic spike (discussed in the following paragraph) and dissolved sample did not work for osmium. The problem is removed when the sample is "lightly bombed" in the presence of ethanol. A reanalysis of IAB iron meteorites demonstrates that they are consistent with a closed-system evolution since the formation time of stone chondrites (Niemeyer et al., 1990).

We completed calibration of our isotopic spikes (rhenium and osmium enriched in certain isotopes, added to a sample as part of the analytical procedure). The osmium spikes were calibrated against the same natural standards used in the ¹⁸⁷Re half-life experiment (Lindner et al., 1989), with an agreement of better than 1% for two calibrations. Thus, our future chronological work will be essentially independent of errors in the assumed stoichiometry of the standards in the half-life determination.

In ICP-MS (inductively coupled-plasma mass spectroscopy), we implemented peak-jumping software, so that most analysis time can be spent on the (typically) minor ¹⁸⁷Os peak. We are investigating electrothermal vaporization methods of sample introduction to improve our ICP-MS sensitivity.

Collaborating with M. Koide and E. Goldberg of the Scripps Institution of Oceanography, we have analyzed osmium concentrations in marine sediments. Our method is one of the few sensitive enough to analyze such low-abundance samples. The measured osmium isotopic ratios suggest that the higher osmium concentrations in nearshore environments may be due to anthropogenic sources (M. Koide, University of California, San Diego, 1990).

In FY90, we also surveyed ancient Archean shales with high

Re:Os ratios, which may provide constraints on the Earth's early atmosphere and hydrosphere (Gerlach et al., 1990).

Work in FY91 will concern:

- Low-temperature geochemistry of rhenium and osmium, with potential applications in noble-metal marine chemistry and the use of osmium isotopes as sensitive indicators of heavy-metal pollution.
- Iron and stony chondrite meteorites.
- Silicate samples from the crust and mantle, relating to the Earth's geochemical evolution. ■

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Measuring Neutrino Mass from Beta Decay of Gaseous Tritium

D. J. Decman and W. Stoeffl

We are measuring the mass of the electron antineutrino by studying the beta decay of gaseous tritium with a high-resolution magnetic spectrometer and low background counting system.

The mass of the neutrino is an important parameter in many areas of physics. A neutrino mass of even a few electron volts would have profound effects on cosmology because of the vast number of neutrinos in the universe. A neutrino mass will also change the shape of the beta-decay spectrum near the endpoint energy.

Several research groups have attempted to measure the mass of the electron antineutrino by measuring the beta-decay spectrum of tritium. Unfortunately, the number of beta-decay electrons emitted in the energy region of interest represents only about 1 in 10^9 of the total decays in tritium. Furthermore, several sources of systematic error become important in beta spectroscopy at the relevant energies. Sources of error include the resolution function of the analyzing system, the distribution of final atomic or molecular states in the tritium source, and the background rate.

Researchers in Moscow (Boris et al., 1985) reported the results of a tritium beta-decay measurement and claimed evidence for an electron antineutrino mass of at least 18 eV. The latest results of researchers at Los Alamos National Laboratory (Wilkerson et al., 1987) and in Zurich (Frittschi et al., 1986) do not agree with those of Boris et al.; however, their best value of the neutrino

mass was a nonphysical result ($m^2 = -155 \text{ eV}^2$). This value casts doubt on any upper limit that these groups might claim because it shows a large systematic error in measurements. Our experiment is designed to minimize the sources of systematic error and to make our measurement sensitive to a neutrino mass as low as 7 eV.

To achieve such an ambitious goal, we are using a gaseous tritium source. Our approach minimizes the energy loss in the source itself and uses a system in which the final states can be accurately calculated. Electrons from decays in the gaseous source are guided into a large, toroidal magnetic-field spectrometer by five solenoidal superconducting magnets. The gas is differentially pumped at the ends of the 5-m-long source tube by eight turbomolecular pumps and is then reinjected into the middle of the tube. The radioactive gas is prevented from entering the

spectrometer tank by a 1.5-m-long pumping restriction. The magnetic spectrometer focuses the electrons onto a multisegmented, liquid-nitrogen-cooled Si(Li) detector. The position of the electron impact on this detector determines the energy. Because the detector has high energy resolution, we can distinguish between good events and background from radioactive decays and secondary cosmic-ray interactions.

The magnetic spectrometer operates at a constant field and current setting to avoid thermal stresses and misalignment in the spectrometer hardware. We measure electron spectra by changing the electric potential of the source; for the tritium endpoint measurement, this accelerating potential is nominally 3 kV. In this way, electrons from tritium decays in the source tube have energies near 21 keV and are easily

Figure 1. The K-conversion line of the 32-keV transition of ^{83m}Kr on a logarithmic scale. Smooth lines are the result of a least-squares fit to the main-line and shakeoff satellites.

distinguished from background electrons from tritium contamination in the spectrometer, where the maximum energy is 18.6 keV. Our system is built to accommodate an accelerating potential of 25 kV, which allows us to measure a wide range of electron energies.

In FY90, we improved the knowledge of our system's resolution function by performing a detailed measurement of the line shape of our calibration source, $^{83\text{m}}\text{Kr}$. This isomer, which has a half-life $t_{1/2}$ of 1.8 h, is produced in the decay of ^{83}Rb ($t_{1/2} = 86$ d). The radioactive

krypton atoms emanate from a thin layer of $^{83}\text{RbCl}$ and circulate in our source tube in the same manner as tritium. The isomer decays by a cascade of two electromagnetic transitions that have energies of 32 and 9.4 keV. The K-conversion electrons of the 32-keV E3 transition have an energy of 17.8 keV, which is close to the tritium beta-decay endpoint energy of 18.6 keV. Therefore, this isomer can be used to determine the resolution function of the electron-spectrometer gaseous-source system at an important electron energy.

Figure 1 shows the spectrum of the K-conversion line of the 32-keV transition with a fit to the various shakeup and shakeoff components. These data represent the most complete study to date of such a line shape for an internal conversion line. As yet, no calculations are available that can reproduce the information in this spectrum. The overall intensity of the shakeoff for krypton has been calculated in terms of the sudden approximation (Carlson et al., 1968; Mukoyama and Taniguchi, 1987). The results of these calculations underestimate our measured shakeoff intensity by a factor of 2.

We studied the effects of scattering in our source with an electron gun and deuterium gas. With our proposed source density, less than 5% of all decay electrons undergo an energy loss, thereby contributing only a small correction to our data. We have tested our gas-regulation system with both deuterium and tritium gas and have found that we can keep the gas density in the source tube constant to better than 0.5%.

Our ability to accelerate electrons by more than 20 kV allows us to measure the full beta-decay spectrum from zero energy to the endpoint. The wide range allows us to

Figure 2. (a) Beta-decay spectrum of tritium. (b) Detailed spectrum of the zero-energy electrons detected in this experiment.

use the intense and well-understood portion of the tritium beta-decay spectrum to calibrate system efficiency. In other experiments, efficiency must be extracted from data around the endpoint, yielding one additional source of systematic error. This aspect of our device allows us to make a unique contribution because we can actually measure the electrons from the continuum final states of the ${}^3\text{He-T}^+$ system. Our work represents the first check of calculations of this effect, which are the greatest potential source of systematic errors in such experiments. In addition, we are the first researchers to measure in detail the electron spectrum of beta decay below 1 keV. This energy region is of general interest because it shows the greatest effect of the Coulomb corrections.

Figure 2a shows our data on the full energy spectrum for tritium decay. The spectrum demonstrates that our efficiency function is smooth and easy to determine. Our measured krypton conversion electron and Auger line intensities are in excellent agreement with this efficiency function. Figure 2b is a

detailed spectrum of the zero-energy electrons detected in our experiment. The electrons in this energy region include low-energy beta emissions, electrons emitted from the unbound molecular final states, and secondary electrons from scattering in the source tube. We believe that we can determine the relative contributions of these sources of low-energy electrons by studying the spectrum as a function of source-gas composition.

In FY91, we expect to complete our project and to set the best limit for the neutrino mass of any experiment to date. We plan to continue our studies of zero-energy electrons from tritium beta decay and anticipate that we will be able to explain the problems encountered in previous experiments. More importantly, we plan to perform long-duration measurements in the endpoint region of the tritium beta-decay spectrum. We will conduct these experiments after we install the full-size detector to maximize our count rate. Initial tests indicate that, after a running time of a few weeks, we will obtain data sensitive to a neutrino mass of greater than 7 eV. ■

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Dating Surface Geologic Processes by Measuring Terrestrial In Situ Cosmic Ray Isotopes

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Several nuclides produced by the interaction of cosmic rays with surface rocks have ideal properties for dating surface geologic processes in the <100,000-y time frame. Although these isotopes occur in extremely low natural abundance, analytical techniques now exist that can measure fewer than 1 million atoms of a given isotope.

A premier application of terrestrial in situ cosmic-ray (TISC) isotopes, of which ^3He , ^{21}Ne , and ^{81}Kr are a few, is the determination of the chronology of retreat of mountain and continental glaciers. Rocks that show glacial polish can be sampled to ensure that inherited TISC isotopes have been scraped away and that postglacial erosion has not been severe. This chronology would be an important contribution to understanding the nature of climate change in the Pleistocene.

During FY90, we made considerable progress toward our goal of developing chronometers for dating recent geologic activity. Our most important achievement was the measurement of ^{81}Kr in a suite of

standard samples in which the $^{81}\text{Kr}/\text{Kr}$ ratio was as low as 2×10^{-7} . To perform this measurement, we spent considerable effort eliminating hydrocarbon and ^{81}Br backgrounds. In addition, we selected our first Sierra rocks for analysis and have begun working on techniques to isolate mineral phases from these granites.

We also entered into a collaborative effort with the University of California, San Diego, to measure ^3He and ^{21}Ne in very old Antarctic mountain samples. Our primary goal in this effort was to look for a diffusive loss of ^3He . We have demonstrated that helium is not quantitatively retained in quartz and thus cannot be used as a surface-exposure chronometer. Within the uncertainties of production rates, ^{21}Ne is retained quantitatively. ■

Physics

H. C. Graboske and C. B. Tarter

The Physics Department's ER&D projects seek innovations and new capabilities in areas that contribute to LLNL's major programs of fundamental science, defense, environment, lasers, and technology transfer. They may develop in three ways: (1) as new inventions or ideas, (2) as applications of unique or highly developed Laboratory capabilities to new problems, and (3) as start-up projects that can evolve to self-sustaining levels by attracting independent support. In FY90, one project became a reimbursable program, and three projects became key elements in successful proposals for Director's Initiatives.

For one project in fundamental science, we are collecting data in the neutrino mass experiment. The 15-institution PEGASYS collaboration, led by LLNL, is working on a large, high-energy experiment at Fermi National Laboratory, before continuing work at the Stanford Linear Accelerator Center. We completed the new detectors on the positron-electron scattering project and used them to obtain interesting results from heavy-ion collisions at Lawrence Berkeley Laboratory. We also completed the design of Super-EBIT, an electron-beam ion trap, and are fabricating and installing it. In the MACHO project collaboration, we designed and modified the telescope and assembled the camera to be used to study the Milky Way Galaxy.

Two new projects in fundamental research began in FY90. Using EBIT's highly charged ion source, we are studying the interaction of very highly charged single ions with a crystal surface. We also began to study a project to search for and measure the mass of axions using LLNL's large-volume, high-field magnets, together with microwave cavities fielded by the University of Florida.

In the area of defense, we designed and evaluated the hard-x-ray/gamma-ray imaging spectrometer for use in both space astronomy and treaty-verification applications. We evaluated the FLIT3D radiation transport/hydrodynamic code on LLNL's BBN multiprocessor, and identified software and hardware improvements needed for maximum computing efficiency. In another project, we are developing advanced cryogenic detectors for hard photons (x rays) and charged particles.

To support the environmental and energy programs, we modified our atmospheric chemistry codes to include aerosols, which are now known to be critical elements in predicting cloud formation (a key uncertainty in climate change models). Our development of a large-scale computer model to study incineration processes focused on combustion of CHCs, an important class of hazardous chemical wastes. We also adapted two established ocean general circulation models (GCMs) for use with our atmospheric GCMs.

Three projects support the work of the Laser Programs and technology transfer. We advanced the application of laboratory x-ray lasers to biological holography by designing specialized targets that produce optimal x-ray laser beams. An alternative method of generating coherent extreme ultraviolet (XUV) radiation was greatly advanced by combining our time-dependent Hartree-Fock code predictions with LLNL's new short-pulse laser facility to demonstrate that very high harmonics (XUV wavelengths) can be efficiently generated by table-top lasers. Our theoretical and experimental studies of high-temperature superconductors, conducted jointly with the Chemistry and Materials Science Department, have led to several promising discoveries and a patent for a new type of superconductor.

In FY90, the Physics Department received \$2.37 million in ER&D funding, which includes \$100,000 for capital equipment. ■

Laboratory X-Ray Laser Design

M. D. Rosen, M. S. Maxon, D. C. Eder, R. A. London, and P. Amendt

We are directing the major effort of a laboratory-wide effort to develop the physics and technology of laboratory-scale x-ray lasers for biological applications. In FY90 we achieved a major milestone by demonstrating lasing at the 4.5-nm region, which is optimal for imaging biological structures.

The first successful demonstration of lasing at x-ray wavelengths occurred at LLNL in FY84 when the Novette laser was used to produce soft x-rays of 20.6-nm wavelength. This achievement was recently recognized by the American Physical Society with the 1990 Excellence in Plasma Physics Award. Since the

initial demonstration, our work has focused on understanding the growing body of experimental data in order to improve old designs and invent new ones. We are looking for ways to obtain more coherent lasing at shorter wavelengths, which would make possible biological applications such as holographic imaging of wet subcellular structures.

FY90 Accomplishments

Our FY90 goals were to design long-pulse (500-ps) nickel-like tantalum collisional-excitation targets and short-pulse (20-ps) hydrogen-like magnesium recombination targets, both of which lase at 4.5 nm. We have determined 4.5 nm to be near the optimal wavelength for biological imaging because it maximizes scattering (and signal) while minimizing absorption (and damage) in the sample being imaged.

By mid-FY90, we had taken several steps toward the first goal. Using the Nova II beam, we had demonstrated gain in nickel-like tantalum at 4.5 nm (see Figure 1). Since then, we have gone further and achieved eight gain lengths (i.e., gain times length equal to 8) at 4.5 nm with 20 kW of pump power. We are now analyzing those results and are planning for more optimal performance at lower pump power, which will allow a still greater gain-length product to be achieved.

Looking toward future "tabletop" systems (i.e., laboratory- or room-size), we have designed, and hope to test in early FY91, a nickel-like tantalum target that can lase upon irradiation with only 1 kJ of 1 ω light. This could be the basis for a 1-kJ user-oriented x-ray laser (XRL) facility of room size that would cost perhaps \$1 million to \$2 million.

As another approach, we have been investigating a two-pulse scheme in which lasing is produced with low total pumping energy by using a long-pulse (1-ns), low-irradiance, low-energy (20-J) beam to explode a target foil without heating it much, thus producing a cool tube of plasma. This plasma is then irradiated to a lasing state with a short-pulse (10-ps), high-irradiance beam of 120 J, a total pump energy only about 2% of that required for the conventional scheme with its high-power, long-duration single pulse. Data taken at the National Research Council (NRC) facility at Ottawa, Canada, on a long-pulse/short-pulse nickel-like scheme have been analyzed. Although they are in round-spot, proof-of-principle geometry rather than the line-focus geometry required to produce actual lasing, these data reveal definite

Figure 1. (a) Spectra from laser-irradiated 1.7- and 2.5-cm tungsten foil targets, which clearly show nonlinear growth of lasing line of nickel-like tungsten. (b) Intensity of 4.318-nm line as a function of target length.

sensitivities to intensity and relative timing of the two pulses. For optimal combinations, nickel-like ytterbium was indeed produced, which indicates that lasing would occur in line-focus geometry. This scheme, too, could lead to tabletop lasing.

Experiments on the short-pulse (20-ps) hydrogen-like magnesium recombination scheme ($n = 3 \rightarrow 2$ at 4.5 nm) were performed—also on round spots—at KMS Fusion Inc. (Ann Arbor, Michigan) in FY90. They showed evidence for population inversion in hydrogen-like aluminum and magnesium. Thus, this scheme holds great promise as well. Analysis of these experiments and design of targets for follow-on experiments using Nova are proceeding on schedule. Our initial 20-ps experiments on Nova were plagued by Nova laser problems, which was not surprising since this was the first attempt at propagating and focusing 20-ps pulses in the system. Further attempts will be made in FY91.

For collisional-excitation (e.g., neon-like and nickel-like) schemes as well as for recombination (e.g., hydrogen-like) schemes, the trapping of radiation in the system can lead to a repopulation of the lower XRL level, which destroys gain. In FY90, we made further advances in the proper modeling of the trapping and line-transfer process, and we designed experiments to test the predictions of these improved models.

The femtosecond approach to tabletop XRLs has also been pursued. Here, a plasma produced by a femtosecond laser acts as a radiation source to pump inversions by inner-shell photoionization, which can compete with the Auger process because the pulse is so short. In FY90, we developed a simple model to explain the radiation produced by such short-duration laser-plasma interactions. This model compares well with complex simulations and with experiments.

In past years, we planned experiments at the NRC facility using Thomson scattering to obtain, for the first time, electron and ion temperatures vs time in exploding-foil plasmas such as are used in XRLs. In FY90, the first successful results from these NRC experiments were obtained, albeit in low-Z carbon foils rather than the higher-Z foils expected to be used in XRLs. These results will be analyzed in the coming year.

Theory and design for improving the coherence of the output from our XRLs is also proceeding at a vigorous pace. We have made substantial analytical progress in predicting the coherent output (see Figure 2), obtaining results that compare favorably with simulations using the WAVE code (Feit and Fleck, 1990). These efforts complement the source design issues mentioned above as well as the holography work funded by a Director's Initiative. Experiments were successfully performed at the end of FY90 to measure the coherence of our laboratory XRLs. Detailed analyses will be performed early in FY91, and experiments designed to improve the coherence will undoubtedly follow shortly thereafter.

FY91 Goals

For the nickel-like tantalum laser, we will enhance the signal further using the new synthetic multilayer normal-incidence mirrors for multipass amplification. By gold-tagging specific structures in a biological sample, we have relaxed the fluence requirements on the XRL output to the point where we seem close to being able to take meaningful data and learn something of biological significance about the internal structure of chromosomes. We will continue to design and test 1 ω alternatives that

operate at lower pump power in order to define the requirements for a separate facility to be designed in FY92. For the long-pulse/short-pulse nickel-like approach, the implications of the present data will be assessed, and follow-on experiments will be designed and performed in FY91.

The recombination 20-ps data from Nova will be analyzed, and follow-on experiments will be planned. A critical review and redesign of alternative tabletop approaches, such as harmonic generation and subpicosecond recombination schemes, will also be carried out. Our preliminary analyses of these alternatives have pointed to serious problems with XRL saturation at relatively low irradiances, as well as to issues of insufficient coherence, which we will further quantify and attempt to resolve. The year will see further use of the femtosecond radiation model as new

Figure 2. Relative intensity $I/I(0)$ (dashed curves) and coherence function μ (solid curves) vs transverse position x/a for square profiles with a gain-length product of 20. The effective Fresnel number F_0 and the refraction parameter η serve to parametrize the effects of gain and refraction, respectively.

femtosecond facilities and experiments come on-line worldwide.

Fiscal year 1991 should see NRC experimental results on electron and ion temperatures with higher-Z targets of more direct relevance to XRL modeling. These will be analyzed, and new experiments will be planned if necessary.

Experiments to measure the coherence of present lasers will be performed and analyzed, as well as experiments to improve coherence based on our ideas and designs. Several innovative experiments to test the accuracy of our trapping calculations are also planned.

Long-Range Outlook

The XRL holography project has already produced a spin-off in the form of a project for DNA sequencing via x-ray diffraction, which is funded entirely by the DOE Office of Health and Environmental Research (DOE/OHER). Next year, we will formulate a proposal for FY92 or FY93 in the form of a structural biology initiative, involving the Laboratory and local universities. We anticipate that this could lead to a dedicated XRL holography user facility, half of whose construction and all of whose support would be funded by DOE/OHER. There is a plan, in the context of a direct-drive inertial-confinement

fusion facility using the 2-MJ Nova Upgrade, for a white-area facility (i.e., one without security restrictions) that will include separate target chambers for high-power irradiations and for XRL research. We are also drawing up plans and strategies for obtaining further support from various DOE agencies, with emphasis on trying to identify a larger user community than that represented by biologists alone. All in all, future prospects for XRLs and their applications seem very good. ■

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Ocean Circulation Models

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Atmospheric general circulation models (GCMs) predict that doubling atmospheric CO_2 would lead to eventual global warming of 2 to 5 K. How quickly the warming would occur—and how the real climate would evolve in response to steadily increasing greenhouse gases—will depend primarily on the behavior of the oceans, which are by far the most important heat sink in the climate system (Hansen et al., 1985). This study aims to use an oceanic GCM to explore methods of comparing different ocean models, in order to determine why they often give different predictions of the rate of greenhouse warming.

During FY90, we reviewed results from a simplified analytic ocean model (Wigley and Schlesinger, 1985), which suggest that, for plausible greenhouse scenarios, the ratio of actual to potential global warming (i.e., the ratio of global warming that has already occurred to the total that will occur, for a given level of greenhouse gases) increases with time.

Therefore, the relative amount of unrealized warming that is “in the pipeline” begins as a large fraction but gradually decreases with time. This conclusion from the analytic model, together with the model’s assumption that, in the global average, downward heat flux in the oceans is proportional to the vertical temperature gradient, can be tested with ocean GCMs, which in turn could be validated by comparison with observations of deep oceanic circulation of radionuclides and other tracers.

Accordingly, we initiated cooperative efforts with the Scripps Institution of Oceanography and the Max Planck Institute for Meteorology (MPI) to move a global oceanic GCM to the supercomputers of the National Energy Research Supercomputer Center (formerly the National Magnetic Fusion Energy Computer Center). This model from MPI uses density as a vertical coordinate, allowing a more accurate representation of oceanic transport processes, which take place mainly along constant-density surfaces. We modified the code to permit output of monthly average “data,” and we tested the model for several simulated years under present-day climate forcing. Initially, the code generated too much data, nearly 400 Mbytes per simulated

year, so we refined the postprocessing to allow less than the full complement of data to be kept. We are now making final refinements to the code—including potential vorticity as a conserved quantity—and to the postprocessing system, in preparation for a multiyear “control” simulation of the present-day climate.

During FY91, we will test the response of the model to a common scenario of greenhouse warming; i.e., the model will have its upper boundary conditions specified by surface air temperatures and wind stresses taken from an existing coupled ocean-atmosphere GCM simulation of greenhouse warming. Comparison of the coupled model’s simulation of vertical heat transport in the ocean with our model’s simulation will test the consistency of the two models, thereby providing a preliminary estimate of uncertainty in projections of greenhouse warming due to gaps in our knowledge of the oceans. ■

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Modeling Global-Scale Chemistry and Microphysics and Their Effect on Climate

J. E. Penner

We use a global model to estimate climate effect from particles that are emitted from the widespread burning of tropical forest and grassland. These particles, as cloud condensation nuclei, could potentially increase the reflected radiation of clouds enough to mask any climatic effect from greenhouse gases to date. Preliminary results show that sulfur compounds from global fossil fuel burning could have a similar large impact.

Atmospheric chemistry and microphysics are linked through chemical processes in which gas-phase species such as sulfur dioxide are converted to the particulate phase. Particles affect

clouds by acting as cloud condensation nuclei (CCN). The number of microscopic particles on which water can condense determines the number and character of the drops in a cloud. If larger numbers of particles are available during cloud formation, the cloud drops will be smaller. For the same total liquid-water content, clouds with smaller drops reflect more solar radiation. This reflection of solar radiation can act to cool the climate (Ghan et al., 1990).

We have been developing a model capable of treating the coupled changes due to chemistry, microphysics, and climatic processes (Penner, 1990). During FY90, we estimated particle emissions from the burning of tropical forest and grassland in Africa, South America, Australia, and Indonesia as a source of CCN and calculated the change in droplet size and number that might result from this source of particles (Penner et al., 1991).

Cloud-droplet microphysics was treated in a highly simplified manner. We assumed that in a natural atmosphere, with no biomass burning, the droplet concentration is typically $50/\text{cm}^3$. In the presence of particles from the burning of biomass, the droplet concentration was increased in proportion to the local concentration of particles, up to a maximum of $200/\text{cm}^3$. These two extremes, 50 and $200/\text{cm}^3$, describe typical droplet concentrations over clean marine regions and over continental regions, where particle populations can be high because of local pollution sources. For the same liquid-water content, a higher droplet concentration translates into a smaller droplet size. We used our global model of transport, transformation, and deposition [GRANTOUR (Walton et al., 1988)] coupled to the Livermore version of the National Center for Atmospheric Research general circulation model to calculate the local concentration of particles from the burning of biomass (see Figure 1) and their effects on cloud droplet size and cloud albedo.

Surprisingly, we found that the increase in cloud albedo caused by the particles from the burning of biomass leads to an increase in reflected solar radiation of 1.9 W/m^2 (Figure 2). This value is close to the average increase in the global radiative warming to date due to increases in CO_2 and other greenhouse gases. Thus, it appears that the particles from the burning of biomass might actually mask any climatic change resulting from greenhouse gases.

This calculated "cooling" effect seen for the burning of biomass should be placed in context (Ghan et

Figure 1. Predicted concentration of smoke particles and CCN at the surface of the Earth in July. Contour intervals are 10, 100, 1000, and 2000 particles/ cm^3 .

al., 1989). For the burning of fossil fuels, if we assume that half of the SO_2 emitted is converted to ammonium sulfate particles, we calculate a source strength of 130 Tg/y. Our estimated source of particles from the burning of biomass was 80 Tg/y. Therefore, assuming that the size distribution of sulfur particles and biomass particles is similar and that the effect of these particles on cloud droplet concentrations is similar, we might expect particles from fossil-fuel emissions of SO_2 to increase cloud albedos as well. To calculate the effects of fossil-fuel emissions of SO_2 on climate, we are continuing to refine our treatment of the conversion of sulfur compounds from oceanic phytoplankton emissions of dimethylsulfide and from fossil-fuel emissions of SO_2 to form particles.

Figure 3 shows a recent calculation of the predicted concentration of CCN from our global model of fossil-fuel emissions of sulfur. As shown, the source of CCN from fossil-fuel combustion can significantly increase particle populations over vast portions of the ocean as well as over North America, Europe, and Eastern Asia. Thus, our initial calculations support the conclusion that fossil-fuel emissions of sulfur may tend to cool the climate. ■

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Figure 2. Predicted zonal average of outgoing solar radiation over land for the background case (black line) and for the case with burning of biomass (colored line). The increase in cloud albedo caused by particles from the burning of biomass leads to an increase of 1.9 W/m^2 in reflected solar radiation.

Figure 3. Predicted concentration of particles from fossil-fuel sulfur emissions at the surface of the Earth in July. Contour intervals are 10, 50, 100, 500, 1000, 2000, and 3000 particles/ cm^3 .

Application of High-Intensity Laser-Atom Interactions

K. C. Kulander and M. D. Perry

Short-pulse lasers can deposit a surprisingly large amount of energy in atoms and molecules in a short time. We are studying ways to exploit this concentration of energy

We have been investigating the behavior of atoms and molecules subjected to intense, short-pulse laser irradiation. One goal of our research is to understand the dynamics of photoemission from an atom in the presence of a laser

field, then to exploit this process and construct a source of short-wavelength, coherent radiation. A second goal is to understand the effects of intermediate states on excitation, ionization, and photoemission.

Recent experiments (McPherson et al., 1987; Li et al., 1989) using picosecond optical lasers indicate that high-order harmonic generation in gases may provide a laboratory source of short-wavelength, short-pulse, coherent radiation. We have undertaken a joint experimental and theoretical research effort to better understand this process.

In FY90, we measured harmonic-conversion efficiencies in xenon using our 3-mJ, picosecond, tunable dye-laser system. We observed harmonics up to the 13th order for an incident wavelength of 581.5 nm. In collaboration with researchers at the University of California, Berkeley, we obtained harmonic conversion through the 17th order of 616 nm in neon. In the latter experiments, the peak intensity for the pump laser was 2×10^{14} W/cm².

Our theoretical efforts have focused on two aspects of the problem. Harmonic generation occurs due to photoemission from each atom in a volume of gas into which the pump laser is focused. The overall production at the harmonic frequencies depends on the phase matching of radiation from each atom as the harmonic beam propagates through the medium. The theory of phase matching has been well developed under conditions of a relatively weak pump laser. Under such conditions, the atomic response to the field is perturbative, and the phase-matching integral for a

Gaussian, focused pump beam is well-known. The atomic response to laser fields with intensities greater than 10^{13} W/cm² is not perturbative (Kulander and Shore, 1990). The electric field due to the laser in the intensity range of our experiment is comparable to the intra-atomic interactions so that excitation dynamics and, therefore, photoemission rates are markedly nonperturbative. This means that evaluation of the phase-matching integral over the focal volume becomes nontrivial and requires a numerical solution.

In collaboration with A. L'Huillier from Saclay, we have obtained absolute conversion efficiencies to compare to the French experiments (Li et al., 1989) by numerically integrating Maxwell's equations for the propagation of the harmonic fields. The polarization of the medium at the harmonic frequencies was calculated using our single atom codes. Figure 1 shows the calculated and measured number of photons emitted at each odd-harmonic of the 1064-nm driving field for several peak intensities. Except for the highest-intensity case, the results agree within the experimental error bars, which are estimated to be one order of magnitude. These results are for harmonic conversion in 15 Torr of xenon gas with a weakly focused Gaussian beam and a pulse duration of 36 ps. At the highest intensity, 3×10^{13} W/cm², the medium is appreciably ionized. Because ionization has not been completely accounted for in this model, experimental results are expected to be lower than the predicted yields. We have neglected the change in the index of refraction due to free electrons.

Figure 1. (a) Calculated and (b) measured number of harmonic photons generated in xenon gas for five peak intensities of a 36-ps, 1064-nm pump laser.

The second focus of our theoretical effort in FY90 was to develop faster and more accurate codes for calculating the single-atom emission spectrum. The new codes, which directly solve the time-dependent Schrödinger equation for the atom in the pulsed laser field, use spherical coordinates so that a partial wave expansion of the electronic orbital can be made. This technique has facilitated the development of l -dependent effective potentials for the multielectron atoms we are studying, where l is the electronic orbital angular momentum. The l -dependent potentials give a much more accurate representation of the excited states of the atoms and, therefore, better describe the excitation dynamics. This representation also has allowed us to obtain angular distributions of the photoelectrons, which are a severe test of theoretical predictions.

With such advances, we now have convincing evidence that our theoretical techniques and understanding of the physics of high-order harmonic generation give us an excellent predictive capability.

Such capability will be exploited to optimize the design of an efficient, compact source of short-wavelength, coherent radiation.

We have completed the development of a 9-TW, subpicosecond glass-laser system, which will push our studies of harmonic generation into an entirely new intensity regime. Peak intensities of the focused output of this laser can exceed 10^{18} W/cm^2 .

Finally, we have completed studies of the effects of intermediate resonance states on photoelectron energy distributions from molecular hydrogen. Because of the additional internal degrees of freedom in a molecular system (vibrations and rotations), resonance effects are considerably more complicated than in atoms. Using our picosecond dye laser, we have investigated the shifts and broadening of excited levels as functions of wavelength and intensity as reflected in the photoelectron energy distributions. In addition to the expected effects of the known, field-free molecular levels, we have observed what we believe to be resonant enhancement from transient,

field-induced bound states (Bandrauk and McCann, 1989). These are the first experimental results to indicate that such states exist and can play a significant role in excitation dynamics. ■

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Correlated Peak Structures in Positron-Electron Spectra from Heavy-Ion Collisions

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We have implemented a unique detection system in a new High Efficiency Coincident Lepton Spectrometer to measure correlated peak structures in positron-electron spectra from heavy-ion collisions.

Correlated narrow-peak structures in positron-electron spectra discovered in super-heavy nuclear collision experiments at GSI, Darmstadt, Germany (Cowan et al., 1986) may signal the production and decay of a new family of neutral particle-like objects having masses of 1 to 2 MeV/ c^2 . Elsewhere in this *Annual Report* (Cowan et al., 1991), we describe an experiment that scatters a beam of monoenergetic electrons on a gas of cold,

magnetically confined positrons in a Malmberg-type trap to search for the resonant formation of these objects. An essential requirement for the success of that experiment is the coincident detection of the scattered positrons and electrons with very high collection efficiency.

During FY90, we developed and tested a detection system that achieves nearly 100% positron-electron collection efficiency and is compatible with the magnetic environment of the Malmberg-type positron trap. We successfully implemented a modified version of the detection system in a new High-Efficiency Coincident Lepton Spectrometer (HECLS). The spectrometer will be used to investigate these correlated positron-electron peaks in heavy-ion collisions at the Lawrence Berkeley Laboratory

(LBL) SuperHILAC and 88-in. cyclotron accelerators.

As shown in Figure 1, HECLS has a horizontal, 12.7-cm-diam superconducting solenoid, along the axis of which a projectile ion beam is directed at a thin-foil target, centered in the solenoid's 27-kG magnetic-field region. Four target foils are mounted on a small wheel, which allows us periodically to advance the portion of target foil exposed to the 3-mm-diam beam. Positrons and electrons emitted from the collision are collected over a 4π -sr area and transported by the magnetic field to detectors, which are located both upstream and downstream of the target.

As these positrons and electrons, or leptons, travel from the target region's strong magnetic field toward the detector region's weak magnetic field (~ 1 kG), their helical trajectories adiabatically expand and elongate. The leptons continue to pass through a localized magnetic field that is produced by samarium cobalt permanent magnets located in the fringe field of the solenoid, perpendicular to the lepton's path. This magnetic field deflects them transversely, and the positrons and electrons separate as they spiral, with opposite orbital helicity, into separate arrays of lithium-drifted silicon detectors. Positrons are further distinguished from electrons by the coincident detection of their 511-keV annihilation radiation in sodium iodide detectors. Nuclear gamma rays are detected in a sodium iodide detector

Figure 1. Layout of the HECLS used in the FY90 proof-of-principle positron-electron experiment at the LBL SuperHILAC accelerator. The experiment used a ^{238}U electron beam and a tantalum positron target. B = magnetic field.

located at a 90-deg angle to the beam. Projectile and recoil ions scattered from the target are detected over the full 2π azimuthal area in a heavy-ion detector.

HECLS has been successfully tested at the LBL SuperHILAC accelerator using 6.1- and 6.4-MeV/amu beams of uranium incident on $0.9\text{-mg}\cdot\text{cm}^{-2}$ -thick targets of tantalum. In the proof-of-principle experiment, we achieved an eightfold increase in detection efficiency over that of the Electron Positron Spectrometer (EPOS) at GSI (Cowan et al., 1986) even though we were limited to lower beam currents. We designed several minor upgrades of HECLS this year and will implement them in FY91. They include lepton detectors that are thicker and more segmented, an iron magnet yoke that will better shape the magnetic field, and more efficient heavy-ion detectors. These upgrades will increase the detection efficiency up to 25 times that of EPOS.

As shown in Table 1, the ability to measure the lepton emission energy resolution and angular resolution (i.e., upstream or downstream of the target) with HECLS will be competitive with the capabilities of the GSI EPOS and Orange-Type Beta Spectrometer (ORANGE), which have been active since 1985 and 1987, respectively. However, data-accumulation rates that are 25 to 65 times higher than those of EPOS and ORANGE will be achieved in experiments planned at the LBL 88-in. cyclotron accelerator. HECLS will also exceed the detection efficiency of the \$2.4 million ATLAS Positron Experiment (APEX), planned for 1991 at the Argonne National Laboratory (ANL), by a factor of about 4.5.

The data-accumulation-rate capability of HECLS enables us, for the first time, to systematically investigate how correlated positron-electron

peaks are produced in a heavy-ion collision. We have submitted a proposal to the DOE Office of High Energy and Nuclear Physics for continued funding of an extensive research program using the LBL SuperHILAC and 88-in. cyclotron accelerator facilities. Within a time frame that is competitive with the first APEX runs at ANL, we propose to:

- Increase the world's data sample of positron-electron peak events by almost two orders of magnitude.
- Catalog the peaks and search for additional structures.
- Precisely compare the positron-electron peak energies in various heavy-ion collision systems to determine whether they arise from a set of discrete states.
- Investigate the role of the Coulomb field and nuclear processes in the peak production process.
- Determine whether the peaks are consistent with back-to-back emission from a two-body decay source.

We are now preparing for an experiment in early 1991 at the 88-in. cyclotron accelerator to search for

correlated positron-electron peaks from xenon + uranium collisions. If peaks are not discovered in this experiment, stringent limits will be placed on the role that the strong, collisional Coulomb field has on the production of the peaks, and we will need to reevaluate several theories for the peaks' origins that involve nonperturbative effects in quantum electrodynamics. If peaks are discovered, however, future experiments will be much simpler, and we will be able to use a wider variety of target and projectile nuclides and various nuclear accelerators. ■

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Table 1. Comparison of e^+e^- experiments.

	EPOS	ORANGE	APEX	HECLS ^a	HECLS ^b
Peak detection efficiency, e^+ (%)	9.6	3.3	9.0	31	50
Peak detection efficiency, e^+e^- (%)	1.3	1.5	5.6	11	25
Energy resolution, e^+e^- (keV)	~20	~10	~15	~40	~15
Angular resolution, e^+e^- (deg)	90	60	10	~120	~90
Number of detectors	9	144	432	15	~30
Heavy-ion acceptance	75% of 2π	2π	2π	2π	2π
Beam current (10^{10} s^{-1})	1.2	2.4-4.8	3-6	0.4	3
Data accumulation rates, e^+e^-	≈ 1	~2-3	18-36	~4	~64

^aSuperHILAC accelerator.

^b88-in. cyclotron accelerator.

Super-EBIT: A Trap for Extremely Highly Charged Ions

D. A. Knapp, R. E. Marrs, P. L. Lee, and J. R. Henderson

We are designing and building an electron beam ion trap to produce and trap extremely highly charged ions up to hydrogen-like uranium (U^{91+}). We plan to make the first observations of bound-state beta decay in this trap.

The advent of the electron-beam ion trap (EBIT), which was developed at LLNL, signified a revolution in atomic physics. It provides an intense, stable source of a wide range of ions, from bare oxygen (O^{8+}) to neon-like uranium

(U^{82+}). These ions can be studied spectroscopically or extracted from the trap.

The logical successor to EBIT is an ion trap that will generate even higher charge states. We are designing and building such a device, called Super-EBIT, which will enable us to study the physics of the most highly charged ions possible. One process that we want to study with Super-EBIT is bound-state beta decay, which has never been observed. In this unusual decay mode, an isotope that is stable when it is a neutral atom becomes unstable once its atomic electrons are removed. Figure 1 shows a level diagram for a pair of such isotopes, ^{163}Ho and ^{163}Dy . When ^{163}Ho is neutral, it decays by electron capture into ^{163}Dy . But when the K- and L-shell electrons are removed from ^{163}Dy , it can decay back into ^{163}Ho by creating an electron in a bound orbital directly.

An EBIT consists of a high-current-density electron beam passing through a series of drift tubes (see Figure 2). Ions are trapped radially inside the electron beam by its space charge and are kept axially in the trap by voltages applied to the drift tubes. The primary limitation on the charge states available in the original EBIT is the electron-beam energy, which cannot exceed about 30 keV. The energy is attained by floating the drift tubes at a positive voltage. This scheme is impractical

for the very high energies desired for Super-EBIT, so an electron gun at a high negative potential will be used. Super-EBIT is initially being designed for an electron-beam energy of 150 keV, with an eventual goal of 250 keV.

The ionization states attainable in Super-EBIT will be determined by a balance between ionization and recombination. Two significant recombination processes occur in an electron-beam ion trap: radiative recombination, in which an electron from the beam is captured by an ion, and charge-exchange recombination, in which an electron from a neutral gas atom is stripped off and captured by an ion. The ratio between impact ionization and radiative recombination defines an upper limit for the ionization state attainable for a given ion species and electron-beam energy. Neutral gas is injected into the trap to "cool" the highly charged ions. In studies of various cooling gases, we have determined that neon gas can cool the ions without a significant degradation of the ionization balance.

The requirements for the electron beam for an ion trap are exceptionally stringent. The electron beam must be laminar and "quiet," so that it does not heat the ions excessively, and it must simultaneously have a high current density. To meet these criteria, we need a beam with high "brightness." The electron gun necessary to generate a

Figure 1. Bound-state beta decay of ^{163}Dy to ^{163}Ho . When ^{163}Ho is neutral, it electron-captures to ^{163}Dy from the $n = 3$ and higher shells. When ^{163}Dy is stripped of its atomic electrons, however, it can decay to ^{163}Ho by creating an electron in a vacant bound state.

beam with the required brightness is of the type called a Pierce gun. However, the Pierce gun requires a finely tuned magnetic field with a large field gradient. We designed the electric and magnetic fields for our new electron-gun geometry by a series of computer simulations. Similar calculations were needed to design the fields for the other end of the beam, where it is dumped into an electron collector.

Another area requiring a great deal of design work is the high-voltage system. To observe bound-state beta decay, we need very stable high voltage so that we can keep the ions trapped for hours. We have built a test stand to evaluate various high-voltage standoffs for stability and reliability.

We plan to finish construction of Super-EBIT early in 1991 and make the first observations of bound-state beta decay later in the year. If we accomplish these goals, we will extend them to measure bound-state beta decay of ^{205}Tl to ^{205}Pb . A measurement of this bound-state beta-decay rate could provide a way of determining the flux of low-energy solar neutrinos over the last 15 million y. Low-energy neutrinos from the sun are captured by ^{205}Tl

in the inverse beta-decay process $^{205}\text{Tl}(\nu, e)^{205}\text{Pb}$. The resulting ^{205}Pb decays back to ^{205}Tl by electron capture, with a 17-million-y half-life. The bound-state beta decay has the same nuclear matrix element as the neutrino capture, so measuring its

half-life would give the cross section for neutrino capture. Measuring the $^{205}\text{Pb}:^{205}\text{Tl}$ ratio in deep thallium ore would then provide a measurement of the solar neutrino flux integrated over the lifetime of the ^{205}Pb . ■

Figure 2. Operation of an EBIT. Ions are trapped radially by an electron beam and axially by potentials applied to the end drift tubes. The electron beam also ionizes and excites the ions. We view the resulting x rays through viewports in drift tube No. 2.

Gamma-Ray Satellites

C. J. Hailey

We have developed advanced concepts and hardware for subarcminute-resolution gamma-ray telescopes appropriate for space-based platforms. These sensor systems show promise for both gamma-ray astronomy and nuclear surveillance systems.

The requirements for high sensitivity and subarcminute angular resolution at gamma-ray energies (defined as 0.1 to 2 MeV) place severe demands on a gamma-ray telescope. The telescope, composed of its imaging optic and a gamma-ray detector, must be light and compact. At the same time, it must have a fairly large area to achieve high sensitivity, and the gamma-ray detector must have very good spatial resolution for the telescope to have good angular resolution. We have examined several detectors and one imaging scheme that offer substantial improvements in performance over current gamma-ray telescopes.

The first detector is called the fiber-fed imaging spectrometer (FIFIS) (Ziock and Hailey, 1989). In FIFIS, long bars of thick scintillator crystals capture gamma rays and convert them to optical photons. Some of these photons are detected

by a nonimaging phototube (PMT) to determine the energy of the incident gamma rays. The remaining photons are channeled via optical fibers to an imaging PMT. The registration of a signal in this PMT at a given position indicates which scintillator bar absorbed a gamma ray and, hence, the one-dimensional position of the incident gamma ray.

This detection scheme can provide very high quantum efficiency with high spatial resolution because the bars can be made very thick and narrow. In a detector having a thick crystal mounted directly on an imaging PMT or an array of nonimaging PMTs, the spatial resolution degrades with crystal thickness. FIFIS does not have this problem and, thus, can obtain submillimeter position resolution. Moreover, the bars on many nonimaging phototubes can be attached to a single imaging PMT, so there is a cost advantage for FIFIS because fewer expensive imaging PMTs are required. Since only real events will produce a coincident signal in the nonimaging and imaging PMTs, background events are effectively rejected with FIFIS. By using interleaved rows of scintillators with different decay times, we can also obtain pixel-to-pixel identification and rejection of Compton-scattering events; the Compton and non-Compton gamma-rays will present different decay times in the nonimaging PMT.

A second detection scheme we have evaluated for space-borne applications is the imaging germanium drift chamber (IGDC) (Hailey et al., 1989). The IGDC uses a bar of germanium, cooled to liquid-nitrogen temperature, in which an electric field transverse to the bar length sets up a depletion region. A second electric field is formed along the length of the bar by a doping gradient. Gamma rays interacting in the IGDC form electron-hole pairs. The holes are swept out of the depletion region, initiating a "start" signal in the electronics. The electrons drift to the end of the bar where they are collected, forming a "stop" signal. Thus, the length of drift is directly proportional to the position of the original gamma-ray event. The IGDC has obtained position resolution of less than 500 μm and, in addition, has the excellent energy resolution of germanium detectors.

If we want to obtain true two-dimensional pictures with the IGDC, we must use an imaging optic. To obtain such an image from inherently one-dimensional detectors, we employ a novel technique called rotated coded apertures (ROCAT) (Harrison et al., 1989). This is a powerful generalization of a technique that has commonly been used with two-dimensional detectors for many years. With ROCAT, a large number of one-dimensional image slices of an object, obtained from one-dimensional detectors, can be combined

into a full two-dimensional image. ROCAT, together with either FIFIS or IGDC, offers gamma-ray telescopes with 5 to 10 times better angular resolution than systems previously employed.

Several opportunities for using gamma-ray sensors of the type described here should become available in the near term. These include gamma-ray telescopes to be used on the Space Station Freedom and in the NASA program to launch SuperScout-class satellites. In addition, with their high sensitivity, ruggedness, and compactness, we expect these sensors to offer capabilities beyond the state of the

art for space-based nuclear surveillance systems. We have also examined technical issues related to operation of our gamma-ray telescopes on the minisatellites that are being considered by DOE for several space-based missions. ■

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Three-Dimensional Physics Code for Parallel Processing

P. G. Eltgroth

We are developing a new three-dimensional physics code for massively parallel computation of coupled fluid flow and radiation transport in multimaterial systems. Our approach uses a computer architecture independent of constraints upon the computational framework and explores numerous issues, such as irregular time and space relationships, that may simplify the computations.

The last half of the twentieth century has seen a new development in the field of physics: the advent of large computational systems capable of predicting the behavior of complicated and varied physical phenomena. LLNL has continually been at the forefront of this capability. Several large computer codes designed for running on supercomputers—which use one or a few very powerful processors that can make millions of calculations per second—now embody much of the LLNL expertise in the area of physics modeling. However, we now seem to be at the point of diminishing returns for further development of this “classical” computing style. Limited improvements are expected from new, faster supercomputers or from effort spent in optimizing code performance by conventional means such as vectorization, more expressive languages, and enhanced physics models.

Massively parallel computing—which uses hundreds of individual processors, each about as powerful as a personal computer, to work together on a problem—appears to offer the best hope for big improvements in computational capability. But it may require a new approach to code formulation and organization.

Our classical codes are set much in the style of pre-twentieth-century physics, using a universal time standard to provide a single time reference for the dynamical evolution of a physical system. The special theory of relativity in 1905 demonstrated that this description of nature is not strictly correct because two observers cannot agree on a unique or preferred time standard. Nevertheless, using a single time standard is certainly the easiest way to write a code, and it appears natural to describe a system from the viewpoint of a single observer, given the long-standing use of this technique in classical physics analysis. However, the single-time-standard approach usually forces global synchronization of computation, which is in no way related to the physics model. This synchronization severely limits possibilities for parallel computation.

It is possible and preferable to write code using only local coordination of information, with minimal reference to global data (Eltgroth, 1985). An example is the FLIT family of codes (Eltgroth, 1990b), which uses free-Lagrange independent time steps. These codes use physical causality to determine a local time step that can be used to advance

various regions independently. FLIT codes have been implemented on various parallel processors, including the BBN Butterfly machine at LLNL (see Brooks, 1990).

One of our goals for FY90 was to transfer the FLIT code to the BBN TC2000 (Butterfly II) machine at the Laboratory. Indeed, we had the FLIT2D version running on the computer within a week, an achievement due primarily to the high level of portability written into the code. The computational results were poor, however, as had been expected. In performance measurements on a relatively small test problem with some 3000 cells, using 12 of the Butterfly's CPUs, we observed speedups of 8 of the 12 CPUs, with no scaling improvement for larger numbers of CPUs. On an absolute scale, performance was also poor, never exceeding that of a Cray-X/MP processor.

The fundamental reason for this performance shortfall was the nonuniform memory structure of the BBN machine, which advertises shared memory but exacts penalties for its indiscriminate use. In the FLIT codes, cache memory was rarely reused, local memory was effectively unused, and the shared memory system (used in interleaved mode) was greatly overtaxed.

The changes necessary to the FLIT codes for efficient use of a distributed-memory machine have been planned for some time, since the shared-memory difficulties of the TC2000 had been anticipated. Essentially, the changes involve better data management and a restriction of work regions to a “preferred” CPU. However, our approach has

been to avoid compromising code portability when faced with the need to improve performance. Thus, the code changes are proceeding slowly to ensure that the code will still run efficiently on a large class of future machines.

The original choice of free-Lagrange techniques for the FLIT codes was prompted by the desire to have the minimal amount of spatial constraints upon computation. At the time of this decision, the free-Lagrange approach appeared to be the most suitable. However, our attempts to model three-dimensional systems with asynchronous free-Lagrange techniques gave rise to complex data structures and tangled code (Eltgroth, 1990a). An example of the data structure appears in the FY89 *Annual Report* (Eltgroth, 1990b). In the meantime, we became aware of the smoothed particle hydrodynamics (SPH) approach, which adopts the simplest possible representation of spatial information, where a system is characterized by values at points. Information at points is assumed to

be distributed into the continuum (between points) by a kernel function, freely chosen for optimal performance and accuracy.

Faced with the choice between complicated and simple schemes, and realizing that data management would probably be the critical issue in parallel-processing performance, we decided to abandon the purely free-Lagrange approach and replace it with a combination of the better parts of the free-Lagrange and SPH approaches to solving physical systems. For example, SPH has difficulty with realistic boundary conditions because no geometric information about relationships is kept at the calculation points. Therefore, we use the free-Lagrange approach to identify neighboring points because its inherent topological information is easy to maintain at a system of points. Similarly, the partitioning of variables in space is a complicated procedure in free-Lagrange; thus, to determine physical properties, we use the very simple SPH technique.

To summarize, we have merged the topological simplicity of free-Lagrange with the geometric simplicity of SPH. The result is a new code, as yet unnamed, which is much more compact and manageable. It should be very efficient on a variety of parallel computers. ■

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Gravitational Microlensing

C. R. Alcock, T. S. Axelrod, D. P. Bennett, K. H. Cook, and H.-S. Park

At present, we cannot observe at least 90% of the matter in the Milky Way Galaxy because it emits no light. We are developing a telescopic system to search for this "dark matter" through gravitational microlensing.

The most pressing issue of astrophysics and cosmology is to determine the nature of the ubiquitous "dark matter" in the universe. For our galaxy, overwhelming evidence indicates that at least 90% of the mass is in undetected forms, distributed in a spherical "halo" that surrounds the observable disk of stars (Figure 1). We know this halo exists because of its gravitational pull.

Speculation indicates that this mass is made of hypothetical elementary particles, such as wimps,

axions, or massive neutrinos, or of objects that may weigh between 10 and 80 times the mass of Jupiter (i.e., 0.01 to 0.08 times the mass of the sun). These objects are known as "brown dwarfs." Lighter objects are called "planets" although they do not orbit individual stars. We call them all massive compact halo objects (MACHOs). Because the temperatures at their centers are not high enough to ignite nuclear fusion, MACHOs remain dark and difficult to detect. Our goal is to locate them by using expertise developed in LLNL's Smart Retina Project and the phenomenon of gravitational microlensing.

In 1986, Paczynski suggested that we might identify MACHOs by using gravitational microlensing, which occurs when a MACHO moves close to the line of sight between an observer and a star outside our galaxy. The gravitational field of the MACHO bends the light rays

from the star and acts as a converging lens that amplifies the star's brightness. Because all objects in our galaxy are in motion, a highly characteristic pulse occurs in this amplification provides the MACHO signature.

Because these microlens events are rare, we estimate that we will need regular photometric measurements of millions of extragalactic stars to detect even ten events per year. The best source for background stars is the Magellanic Clouds. These small galaxies at the outer edge of the halo are close enough for us to identify individual stars but far enough to exploit the gravitational lens effect.

In FY90, we began collaborating with groups at the University of California, Berkeley, and the Australian National University (ANU) in Canberra. For this work, we have designed an innovative optical system for ANU's 1.3-m reflecting telescope. The system includes a set of corrector lenses to give the prime focus a very wide field of view (0.5 deg², nearly 100 times greater than a conventional telescope). We will also construct a mosaic of four 2048 × 2048 charge-coupled device (CCD) imagers to increase the collection of data. A dichroic filter will allow simultaneous imaging in two color bands, creating 32 million pixels in the system. We have designed a computer system and plan to develop software for data reduction at the telescope.

In FY91, we will complete most of the engineering for this experiment, including that for the new prime focus. We will also build the CCD cameras, develop the software, and build and install the corrector optics. At the beginning of FY92, we will integrate the system in Australia. □

Figure 1. Schematic of the Milky Way Galaxy. Evidence indicates that at least 90% of the mass for our galaxy is in undetected forms and is distributed in a spherical halo that surrounds the observable disk of stars. To search for this dark matter, we are developing a telescopic system that will detect microlens events.

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EPA 900-100-000

Incineration of Hazardous Chemical Wastes

C. K. Westbrook and W. J. Pitz

A detailed chemical kinetic model has been developed to treat the combustion of several chlorinated hydrocarbons including ethyl chloride (C_2H_5Cl), vinyl chloride (C_2H_3Cl), methyl chloride (CH_3Cl), and other chlorinated ethanes. Using new information on the rates and products of CHC reactions, we constructed the reaction mechanism, and we estimated more accurate thermodynamic properties for the chlorinated species. The new data had a significant impact on the calculated results.

Chlorinated hydrocarbons (CHCs) comprise a significant fraction of hazardous waste. Incineration may be an effective method for disposing of CHCs, but little is known about their combustion chemistry. We have developed a detailed chemical kinetic model to treat the combustion of several chlorinated hydrocarbons including ethyl chloride (C_2H_5Cl), vinyl chloride (C_2H_3Cl), methyl chloride (CH_3Cl), and other chlorinated ethanes. Using new information on the rates and products of CHC reactions, we constructed the reaction mechanism, and we estimated more accurate thermodynamic properties for the chlorinated species. The new data had a significant impact on the calculated results.

We modeled CHC combustion as an idealized stirred reactor, thus simulating some of the physical processes in a highly turbulent incinerator where combustion products are rapidly mixed with the initial reactants. Then we investigated the degree of destruction (destruction efficiency) of each CHC over a wide range of operating conditions. In general, the destruction efficiency was maximized for high temperatures, long residence times, high dilutions, and CHC/oxygen ratios near stoichiometric.

We modeled the effect of pressure from 1 to 100 atm and found the behavior to be complex. The destruction efficiency was highest at 1 atm; it decreased as the pressure was raised from 1 to 10 atm, then increased as the pressure was raised from 10 to 100 atm, though remaining well below its value at 1 atm (Figure 1). These results are significant because they showed, at comparatively little cost and effort, that it probably would not be advantageous to construct incinerators to operate at high pressure. It would have been much more expensive to experimentally assess the influence of high pressure on destruction efficiency.

The reaction-rate constants that exhibited the largest sensitivity with respect to destruction efficiency were related to the hydrogen-oxygen reaction submechanism. In particular, the reaction

leads to multiplication of radical species and an increase in

destruction efficiency of the CHC. The recombination reaction

leads to a loss of hydrogen atoms that might otherwise give radical multiplication through reaction (1). At high pressure, reaction (2) involving three molecules is enhanced over reaction (1) involving two molecules, which causes the drop in destruction efficiency observed in Figure 1 as the pressure is raised from 1 to 10 atm.

In the chlorinated hydrocarbon reaction submechanism, the highest sensitivities were in reactions involving the chlorine atom. If the chlorine atom reacted with the CHC, the destruction efficiency was increased. But if it reacted with other species present in the reacting mixture, the destruction efficiency was usually reduced.

Our identification of the critical reactions in the destruction of CHCs may lead to improved strategies for disposing of these common hazardous wastes. ■

Figure 1. Calculated results of the effect of incinerator pressure on the destruction efficiency of ethyl chloride and the production of other CHCs, specifically methyl chloride and vinyl chloride. The stirred reactor temperature was 1200 K, the ethyl chloride/air mixture was stoichiometric, and the residence time was 0.1 s.

FY90: \$225,000

PEGASYS: A High-Energy Nuclear Physics Facility at SLAC

F. S. Dietrich and K. A. van Bibber

We are planning a detector at the Positron Electron Project storage ring at the Stanford Linear Accelerator Center. This facility will provide a unique opportunity to study the structure of the nucleon by observing the momentum distributions of its constituents, quarks and gluons. In deep-inelastic scattering, the energy transferred to a target is deposited on a single quark. However, early experiments measured only the scattered electron or muon, and even today, little is known about the fate of the struck quark and the mechanism by which it transforms into observable particles (hadrons). Over the past 3 y, we have led a collaboration involving about 60 scientists from the U.S., western Europe, Israel, and the Soviet Union to develop a proposal for a unique facility at the Positron Electron Project (PEP) storage ring at the Stanford Linear Accelerator Center (SLAC). At this facility, we will address the hadron-production mechanism and test the theory of quantum chromodynamics by scattering high-energy electrons from nuclear targets and observing the products of their disintegration. The proposed project is called the PEP Gas-Jet Spectrometer System (PEGASYS).

Deep-inelastic scattering of high-energy electrons and muons has provided the most detailed information available on the structure of the nucleon by observing the momentum distributions of its constituents, quarks and gluons. In

deep-inelastic scattering, the energy transferred to a target is deposited on a single quark. However, early experiments measured only the scattered electron or muon, and even today, little is known about the fate of the struck quark and the mechanism by which it transforms into observable particles (hadrons). Over the past 3 y, we have led a collaboration involving about 60 scientists from the U.S., western Europe, Israel, and the Soviet Union to develop a proposal for a unique facility at the Positron Electron Project (PEP) storage ring at the Stanford Linear Accelerator Center (SLAC). At this facility, we will address the hadron-production mechanism and test the theory of

quantum chromodynamics by scattering high-energy electrons from nuclear targets and observing the products of their disintegration. The proposed project is called the PEP Gas-Jet Spectrometer System (PEGASYS).

Figure 1 shows a schematic view of PEGASYS. The principal features of this facility are a high-energy (up to 15-GeV) electron beam provided by PEP; a gas-jet target that will provide a wide range of target nuclei from hydrogen to xenon; a large-aperture magnetic spectrometer and appropriate detectors to measure the scattered electron and fast, forward-moving hadrons; and additional detectors to measure neutrons and other low-energy decay products from the target nucleus.

After approval by both the Nuclear and High-Energy Program Advisory Committees at SLAC, a proposal requesting \$8 million in funding was sent to the DOE Nuclear Physics Office in November 1989. A successful technical review of the project was held at SLAC in January 1990. The major portion of FY90 ER&D funds for PEGASYS was spent on engineering studies for this technical review together with prototyping the electronic system for triggering the detector and subsequent data readout. While PEGASYS was undergoing review, SLAC personnel began a feasibility study on reconfiguring the PEP ring as a B-meson factory (PEGASYS can be modified to take advantage of the B-factory configuration). However, the DOE suspended its review of PEGASYS until the impact of the B-factory plans on the PEP ring is known.

Figure 1. Plan view of PEGASYS. Electrons entering from the left scatter from nuclei in the target. A scattered electron and most of the target debris are detected in the spectrometer shown to the right of the target.

Because of this delay, the SLAC directorate encouraged the collaborative group to investigate the possibility of rapidly starting its physics program by moving an existing detector, Mark II, to PEP. This detector, which made the first measurements at the Stanford Linear Collider (SLC), will soon be retired from service at the SLC. The PEGASYS collaborative group conducted a summer study that established the feasibility of this option, and a proposal to undertake the move has been written (PEGASYS/Mark II Collaboration, 1990). We anticipate a 2- to 3-y physics program before PEP is modified for a B factory. If this project goes ahead, the cost of the installation (\$2 to 3 million) will be covered by SLAC operating funds.

Figure 2 shows the PEGASYS/Mark II detector, which completely surrounds a 2-m-long target region into which target gases are injected. Detection of particles produced at large angles with respect to the electron beam is superior to that of the original project, whereas the coverage at small angles is not as good. Despite significant differences between the detectors, we have concluded that the major part of the physics program conceived for the original proposal can be addressed with PEGASYS/Mark II.

To test the performance of the neutron detectors planned for PEGASYS, we have implemented the Fermilab Muon Tevatron Experiment (experiment E-665) with an array of detectors that is similar to that planned for the facility at SLAC. In the E-665 experiment, the objectives of which are complementary to those of PEGASYS, 490-GeV

muons are scattered from hydrogen, deuterium, and several heavy targets. Measurements now under way will also yield a first glance at one of the interesting physics topics to be explored at PEGASYS, the response of the nucleus to the color field produced momentarily when a quark is knocked out of one of its constituent nucleons. Information on this topic will be deduced from the spectra of the low-energy (1- to 100-MeV) neutrons we observe. Neutron detectors functioned well during the

portion of the E-665 run that took place during the summer of 1990. Analysis of a small portion of the data sample shows that our measurements should yield an interesting first result on the average energy deposited in a nucleus by deep-inelastic scattering. ■

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Figure 2. Schematic view of PEGASYS/Mark II detector, which is axially symmetric about the beam axis. Products of the interaction between the beam and target nuclei are detected in the drift chamber, calorimeters, and muon counters surrounding the target.

Neutrino Mass Experiment Using Frozen Tritium

O. D. Fackler and M. J. Mugge

We have built an apparatus to determine the electron neutrino mass by measuring the tritium beta decay energy spectrum near the maximum beta energy. Our experimental mass is 1.2 ± 0.4 eV. The resolution is 0.1 eV. The source is a frozen tritium source.

Since the 1930s, astronomers have noted that the rotation curves of galaxies indicate that a large amount of the mass of the universe is invisible. This dark matter is not gas, which would show up as absorption lines in starlight. Similarly, it is not dust, which would appear as luminosity variations that are not observed. Many suggested explanations are being tested. An electron neutrino mass of 5 to 10 eV could explain much of the dark matter because

relic or remanent neutrinos from the big bang have a density of 100 cm^{-3} . A Soviet experiment reported a nonzero neutrino mass in 1980. Since that time, many groups have tried to verify the Soviet results.

In our experiment, we are attempting to measure a radioactive beta-decay energy spectrum near the maximum beta energy. A slight deficiency of electrons from the expected distribution near the measured endpoint would indicate a massive neutrino. The size of that deficiency would indicate the neutrino mass. We have built an apparatus that can surpass Soviet and other experimental efforts to probe neutrino masses of a few electron volts.

Tritium is the isotope generally chosen because it has the lowest endpoint energy of all radioactive isotopes, allowing for the easiest precision measurement of the decay electrons. Tritium must be used in a pure form. The tritium used in

the Soviet experiment and in most other experiments is bound in a complicated organic molecule, which introduces large uncertainties in electron energies from the source. This consequence is due to the large uncertainty in calculating the electronic configuration of the molecule after beta decay. In pure tritium, the excited states introduce a precisely calculable effect on the final energy spectrum.

Figure 1 is a schematic view of our apparatus. Tritium is frozen on a 10-cm-diam aluminium source and placed at the focal point of an electrostatic reflector. Electrons gathered from the source are sent down the axis of the collimator, which selects only those electrons traveling close to the spectrometer axis. The energy of these electrons is then analyzed in the spectrometer and focused onto a detector.

Figure 2 shows the reflector shape, which was computer-designed to

Figure 1. High-resolution spectrometer developed at LLNL. Decay electrons from tritium frozen on the source are gathered by the reflector on the right and sent through the collimator. The collimator accepts a narrow angular spread and passes the 50-cm-diam beam to the spectrometer. By varying the potential of the center grid, only electrons of a higher potential are passed to the focusing electrodes and detector.

Figure 2. Dome-shaped reflector. This device has as much open area as possible to lower backgrounds caused by photon-induced electrons. The reflector works in a manner similar to a searchlight beam, taking the small point-like source and generating a parallel beam.

focus electrons from the small source and send them down the axis of the spectrometer. This reflector is the electrostatic analog of an optical parabola with the source at its focal point. The two additional rings between the dome and the ground plane allow fine tuning of the electrostatic field to improve its properties and to control astigmatism and chromaticity.

The collimator is constructed of 65 thin grids held in position over a distance of 85 to 0.005 cm. Figure 3 shows a detailed view of the front of the collimator. Each aperture into the collimator offers a tunnel through which the electrons must pass. If the direction of an electron is not near the axis of the collimator, it hits a razor-sharp edge and is lost. By making the length of the collimator longer or shorter, we can control the energy resolution of the spectrometer while inversely affecting the data rate.

We chose the electrostatic type of spectrometer because of its large acceptance and high (few eV) resolution. Figure 4 shows the field-shaping electrodes along the edge. By carefully selecting the voltages

Figure 3. Holes in the collimator grids. The holes of each sheet are held in precise alignment, resulting in hundreds of small tunnels. Only those electrons traveling very near the axis pass through the collimator.

placed on these grids, a uniform electric field is set up, which decelerates electrons as they pass through the spectrometer. Electrons with enough energy to pass through the center grid are accelerated to the downstream exit grid. The center

grid is composed of 3000 wires that are 0.002 cm in diameter and spaced 0.02 cm apart. A reflection of light can be seen in the figure from the center grid.

Figure 5 shows the electrostatic focusing electrodes. The three

Figure 4. The spectrometer, made of three parallel wire planes with field-shaping grids along the outer diameter. The grids are visible in this photo as is a reflection from the center grid seen about halfway along the grids. The center grid is made of 0.002-cm-diam wire spaced 0.02 cm apart.

Figure 5. Focusing rings. These rings consist of three nested cylinders that act as an electrostatic lens to focus electrons to the detector. The rings can also be seen in Figure 4 by looking through the spectrometer.

nested cylinders focus the parallel electron beam coming from the spectrometer onto the detector where the electrons are counted. Counting is performed with a thin, scintillating sheet and a photomultiplier tube.

A major part of our efforts during the last 3 y has been designing and fabricating the cryogenic tritium source. This was a challenge because of both ultrahigh vacuum and cryogenic requirements. During

FY90, we completed the source and performed testing to develop operating procedures. This work suggested several improvements to the gas-handling systems, which we implemented. We also performed several tests freezing deuterium, which is a lighter sister isotope of tritium with similar properties. These tests showed that frozen deuterium is stable even if exposed to room-temperature infrared radiation. We then moved the source from the shop where it was built to the experiment in the tritium building. Installation and setup of the source took several months. Figure 6 shows the source positioned next to the 91-cm-diam vacuum vessel. During the last few months of FY90, we installed the remaining experimental components.

In summary, during FY90, we completed and integrated the cryogenic source with our high-resolution electrostatic spectrometer. We have begun systematic calibration and testing and are accumulating data. In FY91, we should be able to set new and significant limits for the electron neutrino mass. ■

Figure 6. The source, located next to the 91-cm-diam vacuum vessel. Tritium is frozen on the 2-K source in the small-diameter cross region on the right. An isolation valve is then opened, and the source is moved to the center of the tank. A thermal-radiation shield surrounding the source is clearly visible.

Director's Initiatives

Exploratory Research and Development FY90

Director's Initiatives

G. L. Struble

Projects selected for funding in the Director's Initiatives category are chosen for their technical merit and for their potential to develop into large, independent, multiyear programs. Each proposal is submitted to the Director by an Associate Director, who agrees to assemble a team of expert scientists and engineers and to manage the project. A board composed of the ER&D Review Committee and invited specialists reviews the proposals and presents its recommendations to the Director for approval. A typical Director's Initiative is funded for no more than 3 y at up to \$3 million/y and is reviewed at least twice a year. In FY90, ER&D supported eight Director's Initiatives, for a total of \$9.735 million, including \$616,000 for capital equipment.

The Director's Initiative support to the Human Genome Project is divided into work on instrumentation and computational problems. Instrumentation efforts focus on automating various aspects of DNA manipulation and on developing an improved procedure to separate and classify DNA fragments. In computational work, researchers are applying new algorithms and computational resources to reconstruct the order of cloned fragments in human chromosome 19.

In a related study, scientists are using x-ray diffraction techniques to analyze DNA sequences and are developing x-ray laser holography to produce high-resolution, three-dimensional images of microstructures within cells.

We also continued to fund a study of nuclear polarization of solid deuterium-tritium (DT). Because polarized DT fuel has a higher fusion cross section, we could use it to fuel a smaller, less expensive inertial confinement fusion (ICF) driver.

In another Initiative, researchers are developing technologies to optimize the use of diode lasers as more efficient, reliable pump sources for solid-state lasers. They also made progress in designing and fabricating novel diffraction-limited microlenses that will substantially reduce the divergence of laser diodes.

The ultimate goal of the Ring Acceleration Experiment (RACE) is to provide a high-power-density, low-cost driver for many applications, such as ICF and a multi-keV x-ray source. In FY90, the RACE program demonstrated energy scaling to 50 kJ of compact torus energy and stable compression by a factor of 2 in radius in a new electrode geometry. The compressed compact torus has an energy and plasma density about 25 times greater and occupies a volume about 25 times smaller than that of the earlier geometry.

LLNL scientists also began collaborative experiments to identify and characterize nuclear shape isomers in nuclei that do not fission. They identified 18 superdeformed (SD) bands in seven nuclei and developed a method to determine the spin of levels in these SD bands. Their results provide the systematic data required to understand the physics of nuclei at large deformation and to explore their potential as an energy source.

In the area of accelerator mass spectrometry (AMS), researchers have developed new levels of sensitivity in the dosimetry of toxic and carcinogenic materials and have applied AMS for the first time in a study of human disease. They also are investigating sediments and humic materials in the global carbon cycle and are using AMS to prepare samples of archeological specimens.

The final Director's Initiative is the massively parallel computing project to develop a new supercomputer concept in which many high-performance microprocessors work in parallel. This system may provide unprecedented computing power for problems of great scientific and technological importance. ■

The Human Genome Initiative

A. V. Carrano

Deoxyribonucleic acid, or DNA, encodes the information necessary to make proteins, the building blocks of life. Proteins are determined by the sequence of the four nucleotides, or information bits, in DNA. In general, the span of DNA nucleotides that codes for a protein is called a gene. There are an estimated 100,000 genes in humans, and fewer than 5000 of them are known. Moreover, <0.1% of human DNA has been sequenced.

National milestones have been established to understand and decipher the human genetic code. The initial goal is to establish a physical map of the human genome. This map is a topographic representation of the linear DNA molecule. Nature divides the genome into its natural units of DNA packing, called chromosomes. There are 24 different human chromosome types, and a map can be created for each of them. The map will identify the location of genes and provide the distance between them as measured in nucleotides. One of the most useful forms of a physical map is a collection of overlapping recombinant DNA clones, which span the length of the chromosome.

Figure 1 shows the steps necessary in creating such a map by the approach we use at LLNL. Once the collection of overlapping clones has been made, the genes can be located

to these clones by a variety of methods used in genetics and molecular biology. Finally, the availability of clones with genes assigned to them will allow scientists to prioritize the DNA for sequencing. For example, it will be important to establish the DNA sequences associated with major diseases before we sequence the DNA that has no known genetic function.

Constructing the physical map of the human genome requires many repetitive, labor-intensive procedures. New methods are continually evolving to minimize the effort and to increase overall efficiency. Many steps in the process are amenable to automation. The computational and engineering staffs at LLNL have helped us in creating a physical map of human chromosome 19. The two projects summarized in the following articles illustrate these interactions.

Instrumentation for Human Genome Research

Instrumentation for Human Genome Research

J. W. Cray, J. L. Walling, D. C. Peters, and A. V. Carrano

To establish a physical map of human chromosome 19, we clone the DNA from this chromosome into recombinant DNA vectors, called cosmids. The average insert size in these vectors is ~40-kilobase (kb) pairs. The goal of the physical mapping effort is to identify a set of cosmids that contain, in 40-kb pieces, all parts of chromosome 19 whose locations along the chromosome-19 DNA are known. Thousands of cosmids must

be screened to find those that contain human DNA fragments that are contiguous along chromosome 19. Contiguous cosmids are identified by digesting DNA isolated from many different cosmids with enzymes called restriction endonucleases to produce DNA fragments of discrete size. Restricted DNA samples from cosmids containing DNA sequences that are partially overlapping (and, therefore, contiguous along the DNA) will have many fragments that are identical

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Steps in constructing a clone map of chromosome 19

Instruments and techniques

In use	Being developed
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Manual methods	Automated robot
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Creating a clone library

A collection of recombinant DNA clones is made from chromosome 19, and each is manually picked into an 8×12 array of small wells on a plastic tray. DNA must be isolated from tens of thousands of these clones.

Isolating DNA

Each clone is manually selected and processed to extract DNA. The multiple steps involved require centrifugations, chemical treatments, and pipetting. A robot is being developed to accomplish this task.

Manual methods	Automated robot
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Cutting DNA into fragments

Isolated DNA from each clone is cut into pieces, which are tagged with a fluorescent dye. These steps are done on a commercially available robotic workstation.

Restriction endonucleases	—
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Sizing DNA fragments

DNA fragments from each clone are placed in the lane of a gel and separated by electrophoresis. As fragments migrate down the gel, they intersect a laser beam, which excites the dye on each fragment. The emitted fluorescence is recorded as a function of time or scan number. The number and size of the fragments for each clone constitute the fingerprint for that clone. Sizing is achieved on a commercially available electrophoresis system.

Fluorescence-based gel electrophoresis	Capillary electrophoresis
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Signal processing

The fluorescence emission and mobility from each fragment must be converted to yield fragment size. Such processing requires correction for spectral overlap of the dyes, peak finding and filtering, normalization, and regression analyses. Software to perform these tasks has been developed at LLNL.

TIMEWARP	MAXENT
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Map building and the contig browser

All clones are compared in pair-wise fashion to find shared fragment sizes. Clones with a significant number of shared fragments are assumed to overlap (form a contig). The contigs are graphically displayed and linked via a relational data base to all other biological and notebook information. The browser was created at LLNL.

Contig browser (prototype)	Contig browser (production)
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Figure 1. Flowchart showing the principal steps required to map human chromosome 19, including some of the instruments now in place or under development at LLNL to automate DNA extraction and fragment identification. The next two articles describe these procedures and innovations in greater detail.

in size. The sizes of the DNA fragments in these samples are identified by separating them using gel electrophoresis.

Our efforts are now rate-limited in three areas:

- DNA must be extracted from thousands of cosmids using labor-intensive methods.
- Cosmids may be mistakenly identified as overlapping because of the chance occurrence of fragments of identical size in two non-overlapping cosmids.
- Gel electrophoresis is time consuming.

Our goal is to develop approaches that will overcome these limitations. Thus, we are developing automated systems to facilitate cosmid DNA extraction and capillary electrophoresis to improve cosmid-overlap analysis.

We are working on an automated robot that can isolate cosmid DNA cleanly and reliably from 200 bacterial cultures per day. This task is accomplished in a series of steps consisting of reagent addition, removal, mixing, column loading and eluent collection, centrifugation, tube transport, capping, decapping, and incubation at constant

temperature. A robot capable of performing each task is being developed in two phases.

In phase 1, which is nearly completed, a flexible, easily modified robot capable of isolating DNA from approximately 50 samples/d is being constructed to gain experience in laboratory automation. This system is based on an xyz translation arm purchased from Source for Automation (SFA), Inc. The translation arm, equipped with appropriate end effectors, is used to process cosmid-carrying bacterial samples in batches of 12 to 32. The upper capacity is determined by the rate at which samples can be processed through the 18 000-rpm centrifuge. The work envelope of each SFA system is 170 cm along the work surface, 61 cm across the work surface, and 46 cm above the work surface. At present, the system has been developed to cap and decap up to twelve 15-ml centrifuge tubes, to centrifuge these tubes, to add and remove liquid reagents from each, and to process the contents of the tubes through resin columns for DNA purification. Liquid addition and aspiration are accomplished using eight

computer-controlled syringe units. The tips of the liquid-handling units can be changed, as required, to maintain sample purity and sterility. The entire system is controlled via two personal computers running a hierarchical laboratory software package combining the SFA and LLNL code. The phase 1 system is limited in speed because it handles tubes one at a time.

Our original goal was to have the phase 1 system operational by May 1990; however, this objective proved to be overly ambitious. We now estimate that the system will be operational by February 1991, if the project continues at its present level. At present, we have developed a good appreciation of the resources required for automation of routine laboratory procedures, and the project has resulted in development of in-house robotics expertise capable of addressing automation tasks as they arise. A phase 2 system will process samples in parallel and, thus, will be able to handle ~200 samples/d.

A second goal of our project is to apply laser capillary electrophoresis to increase the speed and precision with which cosmids carrying overlapping DNA fragments can be identified. In addition, the system is designed to facilitate identification of cosmids carrying guanine-cytosine-rich DNA sequences that may signal the presence of genes. Such identification is accomplished by electrophoresing cosmid restriction fragments through a 50- μ m-inner-diam fused-silica capillary filled with a gel-free electrophoresis buffer (Figure 2). We stain the DNA fragments with adenine-thymine- (AT-) specific dye, Hoechst 33258, and classify them according to the time taken for electrophoresis through the capillary and the intensity of Hoechst

Figure 2. Capillary-electrophoresis system for rapid separation of DNA fragments according to size and for classification of fragments according to both size and Hoechst-33258 binding capacity. See the text for a more detailed description of this system.

fluorescence emitted as the fragments pass through an ultraviolet laser beam. Then we separate the fragments according to size by the molecular sieving effect of linear polymers added to a tris-borate buffer.

Figure 3 shows the fluorescence intensity per unit time, measured for ϕ X174 DNA digested to completion with Hae III, using a commercial buffer system. We performed electrophoresis using a 35-cm-long capillary with a 3-kV applied voltage. During electrophoresis, electro-osmotic buffer flow carries the fragments toward the detector, and the electromotive potential retards their movement toward the detector. In addition, the DNA fragments interact with linear polymers (cellulose derivatives) in the buffer moving toward the detector. The degree of interaction is size-dependent, and larger fragments reach the measurement region first. Fragments are classified according to their Hoechst fluorescence intensity (base composition) and time of arrival at the measurement region (size). Measurement times are typically less than 1 h. This process facilitates cosmid analysis by

speeding the analysis and by allowing us to discriminate between DNA fragments that may be identical in size but not in DNA base composition. In addition, it allows us to identify guanine-cytosine-rich fragments that may contain genes.

Although we have demonstrated proof of principle, we must further

develop the capillary-electrophoresis system before it can be used routinely. In particular, we must improve the reliability of electrophoresis. Our future work will focus on the use of noncross-linked acrylamide as an electrophoresis buffer because this material allows reliable and reproducible separation of DNA fragments. ■

Figure 3. Hoechst-33258 fluorescence intensity vs time for ϕ X174 DNA digested to completion with Hae III (referred to as ϕ X174 DNA/Hae III fragments). Such fragments are classified according to their Hoechst fluorescence intensity (i.e., DNA base composition) and time of arrival at the laser-beam measurement region (i.e., molecular weight or size). The fragment sizes range from 1353 bp at 20 min to 72 bp at 45 min (larger fragments reach the measurement region first). The resolution is about 1.5 bp for mid-size fragments. The composition of the buffer can be changed to optimize the resolution for other size ranges.

Computational Support for the Human Genome Center

E. W. Branscomb, T. R. Slezak, D. O. Nelson, M. C. Wagner, T. M. Yeh, and A. V. Carrano

To analyze the DNA packaged in a chromosome and, hence, the genes encoded by that DNA, we must break the chromosome into relatively small fragments and individually clone each fragment in a microbial host. We

also must know the order that these cloned fragments had in the original DNA molecule even though that information is destroyed by the process of producing the fragments. We recover the information on fragment order by first performing a detailed analysis of each fragment, called fingerprinting. We then use the data from our analysis to reconstruct the chromosome from the smaller fragments. Performing this type of analysis and reconstruction requires substantial computational and mathematical resources.

Feature Extraction and Fingerprint Analysis

Our approach to fingerprinting cloned fragments is based on an automated analysis of DNA fragments obtained from each clone

(Carrano et al., 1989). Restriction fragment fingerprinting involves cutting DNA at specific sites with enzymes called restriction endonucleases and separating the fragments according to size using gel electrophoresis. Analyzing restriction fingerprints for evidence of overlap presents several difficulties. The acquired data are displayed as a random number of possibly superimposed peaks with varying amplitudes and arbitrary positions (see Figure 4 and the discussion of signal processing in the preceding article). The signal is also corrupted by at least two distinct sources of noise: random noise that is uncorrelated from channel to channel, and cross-talk from other samples loaded in the same lane but tagged with dyes of different colors.

We have completed a second-generation suite of robust, reliable programs for signal preparation and analysis in this complex environment. These programs implement methods for random-noise suppression, background subtraction, correction of spectral overlap from different dyes, peak detection, and fragment-size determination.

In FY90, we completed and successfully tested a maximum entropy deconvolver, called MAXENT. MAXENT can recover the locations and relative sizes of peaks from various synthesized test data. However, because of the cost of applying MAXENT to all of the data and its relative sensitivity to estimates of the variance of white noise in the signal, we need to combine this method with other approaches.

In addition to the tasks of noise suppression and data extraction, we have completed a program, called TIMEWARP, to map the locations of peaks to a common coordinate system. TIMEWARP determines fragment sizes from mobility in two steps. First, it uses a dynamic-programming algorithm, which matches peak positions from a known, fiducial standard with the peaks of the local standard run in each lane of the gel. Second, after matching peaks, TIMEWARP interpolates intervening distances using shape-preserving interpolating splines.

We also began the design of a third-generation, hybrid peak detector, which will be more computationally frugal than MAXENT. This detector will use a more rapid method to ascertain the peak on those portions of the signal that it determines to be "easy," while exploiting the analytical capacity of MAXENT on the complex portions of the signal, where such an algorithm is essential.

Figure 4. Inferring overlap from restriction-fingerprint waveforms. (a) Two restriction fragment fingerprints are produced by two cloned fragments from human chromosome 19. Each peak represents the fluorescence emission from a DNA fragment in the clone. (b) From the degree of commonality between the fingerprints produced by the two clones, we estimate the chance that they contain overlapping DNA. Using these estimates, the cloned fragments are then conceptually reassembled (see Figure 5).

Statistical Analysis of Clone Overlap

We recently published a description of our statistical approach to detecting overlap in genomic cloned libraries (Branscomb et al., 1990). In this method, we compute a likelihood ratio and probability of overlap between any two library members on the basis of comparing the members' restriction fingerprints. Our approach assumed complete, error-free data. In FY90, we extended our approach to model the data-generation and measurement process that produces the restriction fingerprint.

Our extension proceeds by building a probabilistic model describing the complete data-generation process, from overlap to peak detection, in terms of a network of interdependent random variables. (This type of network has been called a Bayesian network or probabilistic influence diagram.) The probabilistic model includes separate, but dependent, random variables for the fragment-matching and peak-detection processes. Separating data generation from data capture allows us to build more elaborate error models while localizing the effects of the models on the overall calculation.

Our error models contain two components. First, we model the nature of the measurement process itself, which contains intrinsic errors. Because of differences arising from day-to-day chemistry, host stability, and other factors, fingerprints of putatively identical DNA may not be absolutely identical. For example, differences in signal-to-noise ratios from fingerprint to fingerprint can result in peaks that are resolved in one data set but not in another. Second, we model the somewhat idiosyncratic nature of DNA migration in an electric field (i.e., during

gel electrophoresis). Restriction fragments of identical nominal size can nonetheless migrate at slightly differing speeds according to their precise base-pair composition.

Bayesian networks let us model explicitly the dependencies among the random variables making up our probabilistic model, and they provide a convenient, efficient, and computable representation. In addition, Bayesian networks make explicit the distributed nature of the probability calculation and its amenability to parallelization (i.e., several computers performing similar but independent calculations).

Reassembly of Clones

We assemble the fingerprinted cosmids (i.e., the cloned pieces of chromosome 19) into overlapping sets, called contigs, using a depth-first greedy algorithm that examines the overlaps with highest confidence first. This algorithm dynamically maintains a near-minimal spanning path. A spanning path is an ordered subset of the contig members that span all of the linear DNA represented in the contig. Our algorithm has demonstrated successful reconstruction of Monte Carlo-simulated cosmid clones and has been verified on actual chromosome-19 data by independent techniques.

General Computational Support

A large part of our computational support consists of maintaining general hardware and software resources that are adequate for the project, including large amounts of raw computer power. Providing such power affordably has proved to be a challenging task. For example, we need substantial amounts of computer power to compute the

probability of overlap between each pair of cosmid clones. For 8000 clones, approximately 32 million overlaps are possible, and we can expect only about 50 000 to be valid.

We would ordinarily require a supercomputer to complete calculations of this nature in a reasonable amount of time. However, we have developed a simple mechanism to use all spare CPU cycles on 22 workstations on our Local Area Network to perform this computationally intensive task in parallel, using machines that were already available for other purposes. This method is easily applicable to any task, with independent calculations running on any collection of Unix computers (Slezak et al., 1989).

Data-Base Support

The data for chromosome 19 alone will require at least 1000 megabytes of storage. We also need a logical organization so we can access data in new ways. The major research laboratories involved in the national Human Genome Initiative have selected the Sybase Relational Data-Base Management System to store their data. This system features an advanced server/client architecture that allows front-end software to run on inexpensive client workstations. The system allows us to contact one or more data-base servers that might be on the same machine, the same network, or across the continent without having to know where the data reside.

We have adapted our data-acquisition, analysis, and display programs to interface directly with the data base, allowing us to progress from individual clone-fingerprint waveforms to multiple, spanned,

reassembled islands of clones without any manual data handling. We have also included a large portion of the traditional "lab notebook" data into our data base, allowing us to trace DNA from the initial clone construction through the entire reassembly process. A data-base interface library for the C programming language, which we developed in FY90, allows us to program at a higher level of abstraction. This library also concentrates all data-base-specific details to a single, small piece of

code, giving us the freedom to easily port the code to other vendors' data bases, if desired.

In addition to our local needs, we must be responsive to the needs of outside collaborators and other researchers working on the Human Genome Initiative. Thus, we are promoting the server/client data-base model as the best way to meet the needs of all user groups. We have shown that secure, controlled access to human genome data bases is possible via Internet access. The data-base

system provides individual accounts with passwords and allows data access to be specified as "read only" or "write" on individual columns in tables. Such features allow us to create customized views that control precisely the desired degree of data access.

To meet the requirements of some collaborators in keeping certain data private until published, we have shown that it is easy to time-stamp data and to create views that allow only individual records to be seen by outsiders after a specified amount of time has passed. Our expectation is that we can demonstrate that data can be stored in distributed fashion and shared under local control. Such an approach obviates the need for all data to be centralized in one massive repository, which, because of size and complexity, would become a tardy and incomplete representation of the true data.

Graphical-Interface Development

The volume and complexity of human genome data require graphical display tools for analysis. We have constructed a "contig browser" (Figure 5) using the X11-Windows system and graphics libraries developed at LLNL. This browser has been coupled to our data base and extended to incorporate tools for viewing and comparing raw and normalized fingerprint data. The browser also provides general data-access and visualization functions by featuring access to graphical tools provided by the data-base vendor and access to custom programs developed by LLNL.

Biologists use the contig browser to peruse the output of the automated contig-reassembly process. A user selects the desired reassembly

Figure 5. Reconstruction of an image produced by the LLNL contig browser graphical-interface tool. This figure presents a reassembled "island" of cloned chromosome-19 fragments. Each clone is indicated by its unique identifier number (e.g., F22095). Clones along the bottom rows that are linked together by colored bars are a subset of the island's membership; the "smallest" subset spans the island end-to-end. Various intensities of color indicate the confidence of clone overlap determined from the fingerprint data: intense color (red for the actual display, but shown in orange here) means higher confidence and, generally, larger amounts of overlap. The clone inside the box has been selected with a graphical "mouse" to show, using the color-confidence code, the clones overlapping it. The surrounding buttons allow the user to search for and display different clones and islands and to see various biological facts known about individual clones, such as genes present on a given clone.

stored in the data base; then the browser extracts all relevant data from the data-base server over the Local Area Network (or Internet). A user can view islands of overlapped clones (contigs) and can quickly access all data-base information about those overlaps. Biological information regarding the clones on the map (gene loci and other known links or attributes) is shown on demand. The user can invoke a separate viewing function to see as many

as four raw fingerprint waveforms at once, or to call up other windows to obtain direct access to the data base for detailed queries that cannot be done graphically. ▀

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X-Ray Holography

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Work on x-ray laser holography for structural analysis of intact biological cells and organelles. For the analysis of DNA sequences, we are applying x-ray diffraction techniques to determine the ensemble average of the sequence of labeled bases along the individual elements of crystalline DNA. For x-ray laser holographic imaging, we will apply three-dimensional (3D) x-ray holography to elucidate the structure of objects measuring a few tens of nanometers, such as 30-nm chromatin fibers, nuclear pores, and nucleic-acid replication complexes in living cells. We will use existing laboratory x-ray lasers to produce flash x-ray holograms of the biological structures.

Our work on x-ray holography has two principal goals: the development of x-ray diffraction techniques to analyze DNA sequences, and the development of x-ray laser holography for structural analysis of intact biological cells and organelles. For the analysis of DNA sequences, we are applying x-ray diffraction techniques to determine the ensemble average of the sequence of labeled bases along the individual elements of crystalline DNA. For x-ray laser holographic imaging, we will apply three-dimensional (3D) x-ray holography to elucidate the structure of objects measuring a few tens of nanometers, such as 30-nm chromatin fibers, nuclear pores, and nucleic-acid replication complexes in living cells. We will use existing laboratory x-ray lasers to produce flash x-ray holograms of the biological structures.

DNA Sequence Analysis Based on X-Ray Diffraction

DNA sequences are usually analyzed with procedures based on electrophoresis techniques, which

are relatively slow. A potentially faster method exploits x-ray diffraction techniques. In this approach, we amplify the target DNA sequence to produce many identical copies and label selected sites along the DNA fragments (e.g., all bases of one type) with atoms that scatter x rays efficiently (e.g., with high-atomic-number elements, such as iodine or mercury).

We make the labeled DNA fragments linear and orient them with their long axes perpendicular to a beam of coherent x rays. Then using high-energy x rays with a wavelength smaller than the required resolution (typically ~ 0.15 nm), we will determine the distribution of labels along the target DNA sequence by Fourier analysis of the resulting coherent x-ray scattering pattern. The Fourier transforms of these patterns from four samples of the same target DNA sequence yield the information needed for complete DNA-sequence analysis. Thus, each of the four samples will have only one of the four types of bases labeled. We can interpret the relative positions of labels along copies of the target DNA sequence by labeling one end of each copy of the target DNA sequence with a distinctive, high-atomic-number label. This approach will allow us to specify the positions of the labels relative to the distinctive end label. In general, the end label will be separated from the target DNA sequence by a distance that is comparable to the length of the target DNA sequence.

We require large numbers ($\sim 10^{12}$ to 10^{14}) of essentially identical copies of the target DNA sequence

to produce an interpretable diffraction pattern without significant radiation damage in the sample. We will obtain such copies either by clonal multiplication in a bacterial host or by an in vitro amplification method, such as the polymerase chain reaction (PCR). We will introduce DNA base labels by continuation of the PCR reaction in the presence of one halogenated iododeoxyuridine (I-dUTP). Such labeling results in the incorporation of a halogen at the position of each base of one type. For example, all thymines will be labeled with an iodine by amplification in the presence of I-dUTP. Labeled copies of the target DNA sequence will be arranged in a near-linear form with their long axes perpendicular to the coherent x-ray beam for determination of the positions of the high-atomic-number labels along the target DNA sequence. We will attempt to obtain linear positioning of the fragments using several methods, including parallel positioning in an electrophoretic environment, orientation in a DNA fiber, and orientation in a liquid crystal.

A proof-of-principle experiment is now in its initial stages. This experiment incorporates a rotating anode source using a copper target to produce 0.154-nm K-line radiation. A pyrolytic carbon crystal eliminates broadband radiation from the source. Sufficient spatial coherence is produced with a pinhole and by placing the sample DNA far from the source. We are using a 30-base-long synthetic DNA sequence with two labeled bases and one reference label as the object sequence in the preliminary experiments. We obtain the oriented DNA sample distribution by

using either a liquid crystal or by pulling the DNA in a fiber. Then we record the diffraction patterns on x-ray film. Computer simulations of this experiment, which include all of the atoms within the DNA sequence, indicate that the labeled sequence should be clearly obtained from the predicted diffraction pattern (Yorkey et al., in press). To date, we have obtained high-quality x-ray diffraction patterns of oriented, unlabeled DNA fragments. In FY91, we will initiate experiments with labeled DNA fragments.

X-Ray Laser Holography

Much work in modern biology is directed toward analyzing the function of macromolecular structures (e.g., chromatin-enzyme replication complexes, nuclear-pore structures, structures of protein complexes, and elements of the golgi apparatus) smaller than the resolution limits of light microscopes (~200 nm). This trend will probably increase as the international genomics effort provides genetic information about the proteins that mediate cell function. We are developing x-ray laser holographic imaging to meet the new requirements for biological microscopy.

Our technique uses the coherent beam from an x-ray laser to produce a flash x-ray hologram of the object under investigation. Then we numerically reconstruct the hologram to provide a 3D object image with high spatial resolution. We are investigating several aspects of the microscopic technique, including:

- X-ray scattering physics.
- X-ray hologram simulation and numerical reconstruction.
- Development of x-ray holography geometries.
- Development of an x-ray laser.

The optimal wavelength for producing an x-ray hologram of biological objects in water has been

determined by a detailed investigation of the physics of coherent x-ray scattering from biological materials, such as protein (London et al., 1989). The optimal wavelength is the x-ray wavelength that maximizes coherent x-ray scattering and simultaneously minimizes x-ray absorption. This optimum is about 4.4 nm, just outside the so-called "water window" between the carbon and oxygen K edges (4.37 to 2.3 nm), which was previously believed to be the optimal wavelength regime. A secondary optimum allowing more penetration is 2.4 nm.

We have also investigated damage caused by the x rays that are absorbed by an object during the creation of an x-ray hologram. Production of an interpretable hologram requires that sufficient photons be scattered to allow detection of approximately 100 coherently scattered photons from each object pixel. Such scattered photons must be produced on submillisecond time scales to prevent biological and Brownian motion from compromising the resolution.

For small (~30-nm) objects, heat conduction is ineffective on submillisecond time scales, resulting in significant heating of a sample. We have calculated that achieving a spatial resolution of 30 nm would result in sample heating to approximately 6000 K. This heating would cause rapid hydrodynamic expansion, compromise the resolution, and destroy the object. If the exposure time is less than 50 ps, the object pixels do not have sufficient time to expand much beyond their initial size. Thus, a hologram must be produced before x-ray heating-induced explosion can destroy the object. Such effects force the use of pulsed x-ray sources, such as x-ray lasers, that have the necessary high

peak powers for producing holograms in the required time scales.

The problem of x-ray damage can be greatly reduced if we label the structures of interest in a biological object with high x-ray scattering cross-section materials, such as gold. Microspheres of gold as small as 1 nm in diameter can be attached to cellular structures with the technique of immunogold labeling. With specific structures labeled in this manner, we can significantly reduce the x-ray fluence required for imaging. Using 30-nm gold microspheres reduces the required x-ray fluence by a factor of 60, and the object attains temperatures approximately at the boiling point. Because the resulting expansion is reduced, exposure times of a few hundred picoseconds, well within the capabilities of existing x-ray lasers, become feasible. Larger gold spheres will result in temperatures below the boiling point and in negligible expansion. In this case, the exposure time is limited by the time scales for biological motion, and other x-ray sources, such as synchrotrons, can potentially be used.

We have developed computer codes to simulate and reconstruct x-ray holograms of 3D objects. We use these codes to evaluate different holography systems and to determine their realistic performance capabilities. To validate the codes, we test their predictions and reconstructed images against visible-light holography experiments with known objects. Our simulation studies indicate that multiple-view holography will be required to analyze the 3D structure of macromolecules with a resolution of a few tens of nanometers. Multiple views are required because the depth resolution is proportional to the square

of the hologram f -number. Typical holography systems will have f -numbers of ~ 5 because of the spatial distribution of scattered x rays. Thus, the depth resolution that can be achieved from a single hologram with a 4.4-nm x-ray laser is ~ 100 nm. Holograms produced simultaneously at different angles can be combined to achieve both high 3D spatial resolution and reduced noise. We have demonstrated this technique in numerical simulations of x-ray holograms of simple protein rods and spheres in water.

We have conducted preliminary holography experiments using an x-ray laser and initiated the design of a conceptual x-ray holography system capable of producing high 3D spatial resolution of biological objects. In our initial experiments, we used a selenium x-ray laser with a wavelength of 20.7 nm to make Gabor holograms of 8- μ m-diam carbon fibers and 10- μ m-thick gold bar figures (Trebes et al., 1987). These experiments demonstrated that x-ray lasers are sufficiently intense and coherent to produce holograms in exposure times of ~ 200 ps and that multilayer x-ray mirrors are sufficiently flat and smooth to be used in phase-sensitive x-ray experiments.

We have also produced the conceptual design of an x-ray laser holography system for biological applications. This system is based on Fourier transform holography and uses multiple views. In this system, different spatially coherent portions of the x-ray laser beam are directed by multilayer x-ray mirrors along different lines of sight through the object. A reference scatterer adjacent to the object consists of a gold disk or sphere. X rays scattered from the object interfere with x rays scattered from the reference scatterer at detectors placed along each line of sight produced by the optic. We use x-ray film as a detector, although a charge-coupled device detector could also be used. When three views are recorded, the x-ray laser must have an output energy of ~ 50 μ J to produce interpretable holograms on each detector, resulting in a 3D resolution of 30 nm. The object is maintained in an aqueous environment in an x-ray transparent boron nitride holder.

Finally, the production of holograms of biological objects requires a spatially coherent x-ray laser operating at a wavelength of ~ 4.5 nm. X-ray lasing has been demonstrated at 4.483 nm using a nickel-like tantalum laser (MacGowan et al., 1990).

Experimental efforts to measure and improve the spatial coherence of this laser are under way. We have developed a novel diagnostic for measuring the spatial coherence of an x-ray laser on a single laser shot. This diagnostic is based on partially coherent diffraction techniques and has been tested on a 20-nm laboratory x-ray laser. In FY91, we will measure the spatial coherence of our 4.483-nm-wavelength tantalum x-ray laser and will begin x-ray laser holographic studies of labeled human chromosomes. ■

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Nuclear Polarization of Solid Deuterium-Tritium

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Kulsrud et al. (1982) predicted that deuterons and tritons with their magnetic moments aligned in the same direction will undergo nuclear fusion with a 50% higher cross section than that for deuterons and tritons without such alignment. More (1983) showed that the nuclear polarization would survive a high-power laser compression. These findings could translate into an inertial-confinement fusion driver of one-half the size and cost as that needed for unpolarized fuel. The form of the fuel would be solid deuterium-tritium (DT).

The micro-Kelvin temperatures needed to thermally align nuclear moments are not achievable with the 1-W/mol radioactive-decay heat of tritium. Larger electron magnetic moments must be used to transfer polarization to the nuclei in a microwave-irradiation process called "dynamic nuclear polarization" (Souers et al., 1986). Such a process would be carried out in the more easily accessible temperature range of 0.5 to 4 K.

The major problem with nuclear polarization is the short longitudinal nuclear relaxation time T_1 of the nuclei in solid hydrogen. This time

constant describes the loss of nuclear magnetic energy and polarization memory. The T_1 values range from tenths of a second for the triton to seconds for the deuteron, where hundreds of seconds are needed for polarization. These time constants are unusually short in solid hydrogen because of the free molecular rotation. The $J = 1$ molecules possess electric quadrupole moments that split the $J = 1$ rotational energy level into a band. ($J = 1$ indicates the first excited molecular rotational state of the hydrogen molecule. For H_2 , D_2 , and T_2 , quantum symmetry rules prevent the $J = 1$ to 0 transition except at a slow rate.) This band siphons energy from the nuclear magnetic moments, and the mechanism is called EQQ after the electric quadrupoles. The problem can be fixed in solid HD by waiting for the $J = 1$ molecules to catalyze away. In solid DT, however, the tritium radioactivity creates new $J = 1$ molecules with a 200-h time constant. Normal DT, which contains 25% T_2 , 50% molecular DT, and 25% D_2 , contains too many $J = 1$ molecules to allow any hope of a long T_1 . We have synthesized enriched 85 to 90% molecular DT and raised T_1 to 1 s (Souers et al., 1988).

We had hoped that the deuteron T_1 would be longer than that of the triton because it is smaller and interacts more weakly with its surroundings. Figure 1 shows our FY90 measurements. The T_1 value at low $J = 1$ D_2 is one order of magnitude below that expected from the other hydrogens. Our best explanation (although doubt still exists) is that the tritium

radiation damage creates an electric field gradient (EFG), which interacts with the nuclear electric quadrupole moment of the deuteron to create a new pathway for nuclear relaxation. When a high-power nuclear magnetic resonance (NMR) transmitter was used, T_1 increased from 2 s to more than 10 s, that is, to the value expected by EQQ. We suggest that the high-power transmitter triggers constant "heat spikes" representing hydrogen atom recombinations. Such miniexplosions may anneal the crystal lattice and temporarily remove the EFGs.

Heat spikes are due to atomic recombination as evidenced by the disappearance of the electron spin resonance (ESR) signal. At the same time, light flashes appear, with most of the energy in the near infrared. If the DT sample is old, the sample

Figure 1. Longitudinal relaxation time T_1 for deuterons at 6 to 7 MHz and 2.5 to 5.3 K. Open data points represent regular DT undergoing $J = 1$ to 0 de-excitation; solid data points are for enriched 90% molecular DT. These data differ markedly from those of both solid HD and D_2 in the same temperature range.

darkens, possibly from the precipitation of helium bubbles. Finally, the heat spikes release so much energy that the sample is physically thrown around the sample cell, even at 1.4 K. Such sample motion has been seen through windows and explains the sudden loss of NMR signal measured repeatedly at low temperatures. The spikes confirm the energetic nature of the atoms stored in solid hydrogen and suggest a variant for rocket fuel. In frozen DT at 1.4 K, we have stored 3000 parts per million of hydrogen atoms. In one experiment, we found that inclusions of neon atoms appeared to increase the ESR signal, perhaps indicating the trapping of larger numbers of hydrogen atoms.

The difficulties at low temperatures led us to consider whether nuclear polarization might be possible at relatively "warm" temperatures, such as 15 K. We hoped that the EQQ mechanism might weaken

as thermal motion increased. We also anticipated a possible increase of the deuteron T_1 with NMR frequency. For normal DT and samples with about 1% $J = 1$ hydrogen, we measured the deuteron relaxation times. We found that T_1 increased from 2 s at 1.4 K to about 10 s at 15 K, but we found no NMR frequency dependence. The increase means that the radiation damage at low temperature is annealed out, and the EFG mechanism is removed. However, the 10-s relaxation time means that the EQQ mechanism is still present in the warm solid.

We have considered the EQQ mechanism theoretically. Two $J = 1$ hydrogen molecules that are one molecular distance apart interact via their electric quadrupoles to create energy gaps of 0.5 to 5 K. As the molecules move farther apart, the quadrupole interaction decreases as the inverse fifth power of distance.

For 0.1% $J = 1$ hydrogen, we expect the molecules to be 10 molecular distances apart and the energy gap to drop to 1 MHz, much less than the NMR frequency. The band structure of EQQ disappears and is replaced by the energy splittings of $J = 1$ molecules that happen to come close to each other. There is reason to believe that the EQQ mechanism will be largely gone for $J = 1$ concentrations of 0.01 to 0.1%, and T_1 should increase.

Figure 2 shows our current knowledge with estimated relaxation times for the triton and deuteron in solid DT as a function of the temperature and $J = 1$ T_2 or D_2 concentration. The NMR frequencies are 30 MHz for the triton and 6 MHz for the deuteron. Two regions occur for the deuteron. The region shown with EFG is the measured area in which the EFG mechanism adds the new relaxation mechanism to the deuteron. The region shown with no EFG is the sheet predicted by EQQ theory, which is apparently obtained when the high-power transmitter induces continuous heat spikes in the sample.

It is clear that the long relaxation times are to be found at low $J = 1$ concentrations where we also hope that the relaxation times will increase with increasing NMR frequency. To confirm this expectation, we have constructed an adsorption column designed to remove $J = 1$ hydrogen from molecular DT at 23 K. The variability of adsorption on alumina of the spherical $J = 0$ molecules and the oblong $J = 1$ molecules is the key to their separation. Using our computer model of the column, we predicted removal of $J = 1$ hydrogen below 0.01%. We have mounted the first column in a 1.4-K Janis dewar inside a 13-T magnet. Within six months, a Raman spectrometer will

Figure 2. Estimated longitudinal relaxation times T_1 for the (a) triton and (b) deuteron in solid DT adsorbed in thin layers to silica aerogel. The heavy black lines represent data taken at 30 MHz. The gray region shows estimates based on the (temperature)⁻⁷ dependence seen at 4.3 to 6.5 K. Such dependence is expected from phonon relaxation.

be prepared to analyze the composition of the purified DT. We will study pure DT both at low temperature and at 15 K.

In earlier work, we froze layers of regular DT inside silica aerogel foam (Souers et al., in press). We found that T_1 increased with the inverse seventh power of temperature, a dependence expected in spin-lattice relaxation. Here, the EQQ mechanism is overcome by the electric-field gradients on the foam walls, and long relaxation times are obtained. If we extrapolate the T_1 values to 0.5 to 1.4 K, we can expect

T_1 s of thousands of seconds, at least near the foam walls. Figure 2 clearly shows that lower temperatures increase T_1 . In FY91, we plan to place on-line a 2-mW Oxford He³ cryostat capable of reaching 0.5 K and to experiment with and without silica aerogel. ■

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Semiconductor Lasers and Micro-Optics Technology

J. J. Snyder and M. A. Emanuel

We are developing technologies that will optimize the use of diode lasers as efficient pump sources for solid-state lasers. New laser diode architectures will offer improved efficiencies and better mode control. Inexpensive micro-optics will efficiently couple diode laser light to solid-state laser media.

Solid-state lasers pumped by large arrays of semiconductor diode lasers are potentially more reliable, efficient, compact, and inexpensive than other medium- to high-power laser sources. However, the expected advantages of diode-laser pumping are predicated on developing reliable, efficient, and low-cost diode lasers and on the ability to efficiently couple diode-laser light into the solid-state laser material.

Current approaches to pumping solid-state lasers with arrays of diode lasers are based on proximity coupling, optical fiber coupling, or simple lens systems for transporting diode-laser light to the solid-state laser material. Proximity coupling is simple and can replace flashlamps directly in some designs, but the pump flux density is limited by diode-laser packaging and cooling restraints. Optical fiber coupling offers modest increases in pump flux density and substantially greater versatility in diode-laser packaging. Lens systems can provide the large increases in pump flux density required by new solid-state laser architectures, but optical efficiency is reduced by the complex lateral mode structure and strongly divergent light emitted by diode lasers.

In FY90, we improved our capability to design and fabricate semiconductor laser diodes with enhanced reliability and efficiency

and improved lateral mode control. We also made significant progress in designing and fabricating novel diffraction-limited microlenses that can be integrated with laser diodes to substantially reduce their divergence. The low-divergence beams emitted from these integrated structures can be efficiently coupled to solid-state lasers using readily available standard optics.

Semiconductor Lasers

Our laser-diode facility incorporates a metal-organic chemical vapor deposition (MOCVD) crystal-growth reactor, which is nearly operational. Although the MOCVD facility is complete, experiments have not commenced because of delays in receiving approval to proceed from the DOE Environmental Safety and Health (ES&H) organization. When approved for operation, the MOCVD facility will provide high-quality III-V laser-diode structures for application in novel solid-state laser pumping schemes and basic research into the devices and materials themselves.

We are planning several projects on semiconductor lasers for FY91. The threshold current and efficiency of AlGaAs laser diodes are critical parameters in many applications, particularly in solid-state laser pumping. The parameters are determined primarily by wafer quality and device design. We plan to correlate these parameters to laser-diode fabrication processes and to determine process parameters for demonstrating diodes with low threshold current and high efficiency.

Figure 1. Photograph of a preform for an elliptical fiber lens. The glass preform is ground on a numerically controlled machine.

Modifying a conventional AlGaAs-based laser diode by placing an InGaAs strained-layer quantum well in the active region allows access to the 870- to 1100-nm wavelength range. Such InGaAs strained-layer lasers are potentially useful for various solid-state laser pumping applications. Experimental results from various groups (Larrson et al., 1989, and references therein) suggest that the efficiency and temperature sensitivity of such lasers can approach those of conventional AlGaAs-based lasers; however, the technology has not yet been developed to the point that such desirable characteristics can be realized simultaneously and reproducibly.

InGaAs strained-layer lasers have unusual resistance to the growth of defects that limit the useful life of similar AlGaAs-based devices. The realization of such strain-induced longevity in the conventional AlGaAs laser wavelength regime of 750 to 870 nm would have tremendous impact on the cost and reliability of diode-pumped solid-state laser systems. We plan intensive developmental work in this area.

Diode lasers generally exhibit unfavorable (multiple and unstable) mode behavior in the direction parallel to the junction (Kressel and Butler, 1975). Such poor lateral mode control makes the efficient use of diode lasers difficult in some applications. We determine lateral mode quality primarily by the configuration of the emission stripes (i.e., stripe width and stripe-to-stripe spacing). We plan both theoretical and empirical work to devise configurations that show improved lateral mode behavior.

Micro-Optics

Typical laser diodes have an emission aperture of approximately 1 μm in the dimension normal to the

electrical junction. The small aperture results in a beam divergence on the order of 60 deg (numerical aperture of ~ 0.5), which makes the design of efficient optical systems using diodes difficult. Our work on micro-optics has concentrated on developing diffraction-limited cylindrical lenses about 200 μm wide with numerical apertures of 0.5 or greater. These lenses are attached directly in front of a laser-diode bar to collimate the fast axis (normal to the junction) of the output light, thus reducing its numerical aperture by about two orders of magnitude.

Most techniques available for fabricating micro-optics are limited in numerical aperture to 0.3 or less for a single surface and do not provide aberration-corrected, diffraction-limited performance. Such lenses typically provide good optical performance only for numerical apertures of about 0.15 or less; thus, for example, only about one-half the emitted power of a laser diode can be focused onto the solid-state laser. Computer-generated diffractive optics can, in principle, provide diffraction-limited performance at high numerical apertures. However,

in practice, performance is limited by available lithographic resolution, and the cost of optics is high. An alternative is the use of simple, round optical fibers as cylindrical microlenses (Baer et al., 1989). A round lens has a large numerical aperture (up to 0.9, depending on the index of the glass). However, because of spherical aberration, its diffraction-limited numerical aperture is much smaller.

In FY90, we fabricated shaped optical fibers as diffraction-limited, 200- μm -wide cylindrical lenses with a numerical aperture exceeding 0.6. We make the lenses from glass preforms by heating the glass and drawing it into a fiber in a manner similar to that used for fabricating standard optical fibers. The preform we use has an elliptical cross-sectional shape designed to provide the focusing properties we need. Figure 1 shows a typical preform. During the fiber-drawing process, the cross section of the glass preform shrinks, but the shape is unchanged. The resulting fiber has a cross section identical, except for scale, to the cross section of the original preform. Figure 2 shows a preform stub after

Figure 2. Photograph of a preform stub after drawing. The tapered section is fire polished, and its cross-sectional shape is retained as it is reduced.

Figure 3. SEM picture of a $200\text{-}\times\text{220-}\mu\text{m}$ elliptical lens. Collimated incident light on the curved surface is brought to a line focus at the opposite (planar) side.

drawing; Figure 3 is a scanning electron micrograph (SEM) of a shaped fiber microlens.

An interferogram of one of the elliptical lenses, shown in Figure 4, demonstrates a diffraction-limited aperture of 0.6. For comparison, Figure 5 is an interferogram of a $200\text{-}\mu\text{m}$ -diam, round cylindrical lens (actually a section of standard optical fiber). Comparison of the two figures shows that the round cylindrical lens has substantially more spherical aberration than the elliptical lens. ■

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Figure 4. Interferogram of the microlens shown in Figure 3. The straight fringes indicate diffraction-limited performance over about a $150\text{-}\mu\text{m}$ aperture, which corresponds to a numerical aperture of 0.6.

Figure 5. Interferogram of a round, $200\text{-}\mu\text{m}$ -diam cylindrical lens. The distorted fringes indicate the presence of spherical aberration, which limits the diffraction-limited numerical aperture.

Compact Torus Accelerator

J. H. Hammer, J. L. Eddleman, C. W. Hartman, H. S. McLean, and A. W. Molvik

We have demonstrated energy scaling to 50 kJ of compact torus energy and stable compression by a factor of 2 in radius in a wire electrode geometry. The compressed compact torus has an energy and plasma density 25 times greater and occupies a volume 25 times smaller than that of the earlier geometry.

A new type of accelerator based on acceleration of magnetically confined compact torus (CT) plasma rings (Hammer et al., 1988a) holds the promise of a high-power-density, low-cost driver for many applications, such as weapons-effects testing (Hammer et al., 1988b), weapons physics, and inertial confinement fusion (Hartman et al., 1988). The CT confines plasma in a toroidal configuration similar to a tokamak or other magnetic confinement device, but it differs in that it requires no external coils to produce the field. Plasma currents and image currents in conductors produce the magnetic field, which is "frozen" into the CT by its high electrical conductivity. This property, along with the observed robustness, allows the CT to be translated over large distances, accelerated to high directed kinetic energy, and focused to high power density. The accelerating force can be provided by an azimuthal magnetic field injected behind the CT, which exerts an axial $J \times B$ force on a CT confined between coaxial electrodes.

We embarked on the Ring Acceleration Experiment (RACE) at LLNL with the objective of providing

a proof-of-principle demonstration of the four phases of accelerator operation. As shown in Figure 1, the four phases are:

- CT formation under conditions suitable for acceleration.
- Precompression of rings prior to acceleration for inductive energy storage.
- Acceleration.
- Focusing of rings.

Our experiments before FY90 employed fast ring formation (on an Alfvén-wave transit time) and acceleration in straight coaxial electrodes. We demonstrated formation of resilient, stable, magnetically confined rings along with acceleration to velocities as high as 3×10^8 cm/s (Hammer et al., 1988a). We investigated many aspects of the dynamics of CT ring acceleration, including limits on the accelerating force, ring shaping in response to acceleration, and possible Rayleigh-Taylor instability. We achieved good correspondence with two-dimensional (2D) magnetohydrodynamic (MHD) and other models, giving the desired proof of principle of acceleration for CT rings in the straight system.

Our experiments with the straight geometry culminated in the first quarter of FY90 with a demonstration of energy scaling to the maximum achievable using the present facility. We subsequently installed a new set of electrodes to test the precompression concept and observed stable compression of rings in good quantitative agreement with 2D MHD simulations (Molvik et al., 1990). These results give us confidence in our next step, which is to combine all four phases of acceleration for the final proof-of-principle test at high power density ($>10^{11}$ W/cm²).

One goal of RACE is to demonstrate scaling of the concept to larger energies, as required for many applications. The key to operation at high accelerating voltages (>60 kV) was improved operation of the gun that forms the CT. As the accelerator voltage and current are increased, we must simultaneously increase the CT magnetic fields to prevent "blow-by" of the accelerating field. Operating the gun at higher voltage raises the CT magnetic fields but also tends to increase the amount of follow-on

Figure 1. The four stages of a compact torus accelerator: formation, precompression, acceleration, and focusing.

plasma that can interfere with acceleration. We eventually solved the problem through improved surfaces (plasma-sprayed tungsten in the gun and tantalum liners in the acceleration section) and a conditioning regime of hundreds of moderate-energy shots. The tungsten coating and conditioning regime were modeled after the successful technique developed on the Compact Torus Experiment (CTX) at Los Alamos National Laboratory.

Improved gun operation allowed us to test the scaling properties of both CT formation and acceleration over a fairly wide range of energies. As a guide in scanning parameters, we employed MHD scaling, which consists of scaling the gun, solenoid, and accelerator voltages V proportionally while varying the pressure in the pulse gas valves as V^2 . The

rationale for MHD scaling is that the time constant and impedance of the device are fixed by the geometrically determined inductance and bank capacitance, so the gun and accelerator currents should be proportional to V . The CT and accelerator magnetic fields should be proportional to current and, hence, proportional to V . To keep the plasma time scale matched to the bank time scale, we must fix the characteristic MHD velocity, the Alfvén velocity $\propto B/(n)^{1/2}$. The injected gas density should then be varied as $B^2 \propto V^2$. The scaling is only approximate, however, because the distribution of gas in the gun is not independent of the fill pressure in the gas valves, i.e., viscous effects during valve opening vary with gas pressure, so the density is not strictly proportional to the fill pressure.

Figure 2 shows how the CT kinetic energy, as calculated from the measured circuit parameters, scales with accelerator voltage. The CT energy calculated in this way is in approximate agreement with the mass inferred from interferometer data (assuming hydrogen) and the observed velocity. Two families of shots are shown: those with early gas timing (gas-valve fire time 250 μ s before gun fire time) and late gas timing (150 μ s before gun fire). Both families of shots have an upward trend of the CT magnetic field with increasing voltage, although the early gas timing gives consistently larger fields for the same gun voltage. The higher fields (hence higher efficiency) of the early gas timing may arise because the gas has expanded to fill more of the gun, and the resulting plasma more effectively links the previously emplaced solenoid field.

The acceleration data are reasonably consistent with a fixed efficiency of about 20% over one order of magnitude in accelerator bank energy. An efficiency of 20 to 25% is predicted by the zero-dimensional ring acceleration code (RAC) over a fairly broad range of masses, e.g., 100 to 300 μ g at 100-kV accelerator voltage. The efficiency is lower than the 30% obtained previously because of the shorter acceleration section installed to accommodate x-ray generation experiments and losses in series resistors installed for fault protection in the recently added crowbar circuit. The CT velocities were in the range of 5×10^7 to 1×10^8 cm/s for the early gas timing shots and varied from 1 to 2×10^8 cm/s for the late gas timing shots. All of the shots in the scaling sequence are shown so that the shot-to-shot variations are evident.

In summary, our scaling studies demonstrated ring acceleration in the linear geometry with the accelerator bank energy varying from 20 to

Figure 2. CT kinetic energy calculated from circuit data vs accelerator bank voltage.

200 kJ. Therefore, acceleration probably can be achieved at higher energies by increasing the accelerator length and bank energy while keeping the current and CT magnetic fields fixed or, alternately, increasing the gun, solenoid, and accelerator energies proportionately while keeping the geometry the same.

Following the energy-scaling tests, we reconfigured the accelerator to provide a 2:1 radial compression ratio, as shown in Figure 3, with compressed inner and outer radii of 3.5 and 8 cm, respectively. The precompression cone was followed by a 1-m-long straight acceleration section, which we subsequently increased to 1.7 m. The objectives of these experiments were to demonstrate stable precompression to higher energy and plasma densities, the transition from conical to straight acceleration electrodes, and inductive storage of the accelerating field energy. MHD modeling indicates that precompression is an important prelude to obtaining a tight focus at the end of the acceleration/focusing stage.

An additional advantage of the precompressor geometry is the greater stability against tilt and other modes, which allows formation of the CT on time scales long compared to the Alfvén-wave transit time. In parallel with installation of the new electrode set and tests of ring formation and precompression, we fabricated a 1- μ H inductor, which was added to the gun bank, and a new 11-kV, 240-kJ "slow" gun bank. We changed the gun banks to take advantage of the successful slow ring-formation technique developed at Los Alamos, which allows greater control of CT density and impurity content.

For all gun-bank configurations, we have been able to form CT rings

that are nearly symmetric and localized to the precompressor region. Figure 4a shows data from an array of four magnetic probes measuring B_z , the axial field, spaced every

90 deg at the beginning of the precompressor. Even when the CT shows a large asymmetry early in time (Figure 4b), the asymmetry dies out within about 30 μ s, leaving

Figure 3. The RACE apparatus. This device consists of a magnetized plasma gun, a precompression cone, and a 1.7-m-long coaxial accelerator.

Figure 4. Magnetic-field magnitude vs time at four azimuthal positions on the outer electrode at $z = 0.12$ m. (a) CT formation within the precompressor is usually stable and symmetric. (b) The early portion of CT formation is occasionally asymmetric but becomes symmetric within 30 μ s. These data are for hydrogen plasmas with the 10- μ s-risetime gun bank.

an azimuthally symmetric and quiescent CT by the time the accelerator is fired at 60 μ s. These data demonstrate that the precompressor cone meets the goal of providing stability against tilt modes.

The rings have a mean electron density in the range of $2 \times 10^{15} \text{ cm}^{-3}$, B_z fields of 4 to 8 kG, and decay in $\tau = 50$ to 100 μ s before compression.

After the rings are formed, the accelerator bank is discharged, and we observe a clean sweep up and compression of the CT fields and plasma. The dynamics of the CT are in striking agreement with 2D MHD simulations using the Lagrangian TRAC code. Figure 5 shows TRAC simulations of the precompressor geometry, with the location of the

magnetic probes and the helium-neon interferometer shown in the left-hand column for comparison.

We selected the free parameters, CT mass and magnetic energy, in the TRAC simulation to approximately match the magnetic field strength in the formation region and the observed density. Figure 6 shows the accelerator current, magnetic probe data, and interferometer data for the experiment and TRAC simulation. Of particular note is the timing of the sudden drop in magnetic-field strength in the precompressor, showing that the code accurately predicts the time at which the accelerating magnetic pressure exceeds the CT equilibrium field pressure and begins to compress the CT. The good agreement between the time of arrival and CT field strength in the straight acceleration section is further confirmation of the CT dynamics and that the compression approximately conserves magnetic flux. (In its present version, TRAC is an "ideal" MHD code; i.e., we assume infinite electrical conductivity.)

Rapid phase shifting of the interferometer led to a factor of ~ 2 uncertainty in the density for this case, although the timing and qualitative behavior of the experimental and computational density traces are in accord. Toward the end of the straight section, the simulated CT lags behind the experimental CT, presumably because the CT mass in the simulation (100 μ g) is somewhat too large. The CT in this particular shot near the end of the straight section has undergone partial flux decay, although many shots show negligible flux loss.

The time to expel the CT from the precompressor after the accelerator is fired varies from about 10 μ s at higher accelerating voltages to 20 to 40 μ s at lower voltages, which, in all

Figure 5. Zone boundaries and magnetic flux contours at (a, b) $t = 0$, (c, d) $t = 10$, and (e, f) $t = 12 \mu$ s for a TRAC simulation of the RACE precompression electrodes. Diagnostic locations are shown in the left-hand column for comparison with the experiment.

cases, is many times the characteristic Alfvén-wave transit time for the CT. In studying the low-voltage cases, we found that the magnetic field of the CT, decaying linearly to zero in about 60 μs , decreases faster than the crowbarred accelerator field and that the CT is expelled from the precompressor only after the CT magnetic-field magnitude has decayed to the order of the accelerating field. This finding is suggestive of a force balance between the acceleration field and the CT field pushing against the conical electrodes, as expected on

long time scales where CT inertia is unimportant (Molvik et al., 1990).

To evaluate the forces involved in compression, we compared the acceleration field with the CT field at an axial location of 0.43 m immediately after the precompressor cone. Figure 7 shows the results of 152 such shots. Here, the acceleration field following the CT and measured at the 8-cm radius of the outer straight electrode, B_θ , is plotted vs the peak value of the CT poloidal magnetic field, B_p . The magnetic-field values are averaged over three probes at 0.43 m. We found that B_θ

increases approximately linearly with B_p . The robustness of this result is shown by the close grouping of data taken under three sets of conditions: hydrogen plasmas with either a slowed fast-gun bank (10 μs) or a slow-gun bank (60 μs), and argon or neon plasmas with the slow-gun bank. The result is reasonable because the CT exerts a force against the precompressor cone proportional to B_p^2 directed toward the large end of the cone. This force opposes the force of the acceleration field that is proportional to B_θ^2 ; thus, in the quasi-static case where the forces

Figure 6. Comparison of data with a TRAC simulation. (a) Accelerator current, (b–e) magnetic probe signals at various axial locations, and (f) interferometer data showing post-compression line-averaged density in the straight section. B_z = axial field.

balance, we expect B_θ to be proportional to B_p .

The straight line in Figure 7 is the prediction of the TRAC code in which the CT mass was chosen to scale proportionately with the CT

magnetic energy ($M = 100 \mu\text{g}$ at $U_M = 3.1 \text{ kJ}$ and $B_{CT} = 5 \text{ kG}$ before compression by an accelerator voltage of 80 kV). The agreement is quite satisfactory. Data can lie off the line because of errors in probe

measurements of about 20% or because of effects that are not included in the code, such as ohmic decay of the CT.

The CT accelerates to velocities of 2 to $6 \times 10^7 \text{ cm/s}$ in the 1.7-m straight section used during these pre-compressor experiments. As shown in Figure 8, for example, the CT accelerates after emerging from the pre-compressor as indicated by the increasing slope of the line through the magnetic probe profiles, reaching a velocity of $2.3 \times 10^7 \text{ cm/s}$. Furthermore, the torus remains stable as indicated by the nearly constant shape of the magnetic profiles throughout the acceleration period.

Parameters achieved on RACE are already suitable for two applications. First, the 20-kG magnetic fields, 0.16-m diameter, and 10- to 100- μg ring masses are appropriate for fueling and current-drive experiments in tokamaks. Second, an "opening switch" or power-amplifying device with amplification factors of about 100 should be possible because of the large disparity in time scales between the compression phase (10 to 30 μs), during which the acceleration bank energy is being inductively stored, and the risetime of the accelerating field following the CT ($\sim 300 \text{ ns}$).

We are fabricating a 7-m-long, 3:1 tapered acceleration/focusing electrode set to replace the present 1.7-m-long straight acceleration section. We will conduct experiments in the new geometry in FY91. Modeling calculations using the TRAC code and the zero-dimensional RAC code predict ring kinetic energy in the range of 50 to 100 kJ with a final focus radius of 2.7 cm. The ring power incident on a target is predicted to be $>10^{11} \text{ W/cm}^2$. Further modifications of the acceleration/focusing electrodes to

Figure 7. The CT in quasi-static pressure balance during compression in the conical electrodes. We observed close agreement among data obtained under three sets of conditions: hydrogen plasmas with either a slowed fast-gun bank (10 μs) or a slow-gun bank (60 μs), and argon or neon plasmas with the slow bank. The straight line is the prediction of the TRAC 2D MHD code.

Figure 8. Poloidal magnetic field vs time. The CT is accelerated and remains stable after emerging from the pre-compressor as shown by the displaced, but similar, profiles from each magnetic probe for this shot. Probes measure the axial, or poloidal, magnetic field B_p . The vertical offsets are proportional to the axial locations of each probe. Gray line shows the CT trajectory. This shot uses hydrogen plasma with the 60- μs -risetime gun bank.

allow convergence to smaller radii are predicted to give incident power densities $>10^{12} \text{ W/cm}^2$.

The ultimate goal of the RACE program and possible multimegajoule upgrades is to provide a high-power-density driver for application to inertial fusion, magnetically insulated inertial fusion, a multi-keV x-ray source, and other high-energy-density applications of interest. At each succeeding phase of driver development, increased power densities and, hence, increased performance will be attainable with predicted power densities $>10^{15} \text{ W/cm}^2$ at bank energies $>10 \text{ MJ}$. Because of the relatively low power of the driving

capacitor bank and the high efficiency of acceleration, the CT accelerator has the potential of providing an economical tool ($\sim \$3/\text{J}$ of energy on target) for studies at high total energy and high energy density. ^{as}

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Identification and Characterization of Shape Isomers by Nuclear Spectroscopy

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We have identified 18 superdeformed bands in seven elongated nuclei in the $A \approx 190$ region. Our results provide the systematic data required of a testing ground for the physics of nuclei at large deformation. We have also developed a method to determine the spin of levels in the superdeformed bands.

Shape isomers are states in elongated nuclei that are long-lived because the decay to nuclear states with a normal shape is inhibited simply by the change in shape. Fission isomers are well-known examples; they exhibit pro-

late shapes with a 2:1 axis ratio. We are looking for similar nuclei in which fission is forbidden. Shape isomers have been suggested as possible high-energy-density storage media, but to serve as such media, they must exist with a useful half-life. In addition, it must be possible to produce the isomer in useful quantities, and a mechanism must be available to release the energy on demand.

We are at the beginning of an experimental program to identify and characterize nuclear shape isomers in nuclei where fission is forbidden. By their very nature, our experiments provide information on properties of nuclei at large deformation. Our results, therefore, test and help refine theoretical models of these excitations. We have concentrated

on studies of nuclei in the mercury region, with atomic weight $A \approx 190$, because theories have suggested that nuclei in this region can exist at large deformation and may possess shape isomers. Although shape isomers have not been identified in the $A \approx 190$ region, we have recently demonstrated that nuclei in this region possess superdeformed (SD) bands, that is, a series of levels that exhibit the properties of a nucleus with nearly a 1.65:1 axis ratio.

Typical Superdeformed Band

Figure 1 shows a typical coincidence gamma-ray spectrum signaling an SD band in ^{192}Hg . We obtained this spectrum at the Lawrence Berkeley Laboratory 88-in. cyclotron facility using the 21-detector array HERA spectrometer and the heavy-ion reaction $^{176}\text{Yb}(^{22}\text{Ne}, 6n)^{192}\text{Hg}$. Figure 1 shows the most easily observed signature of an SD band: a band of 10 to 15 gamma rays exhibiting a regular, close spacing indicative of a rotating nucleus with a large moment of inertia. Properties of SD bands are deduced from these gamma-ray energies and intensities.

Extent of the Region of Superdeformation near $A \approx 190$

We have identified one or more SD bands (for a total of 18) in ^{192}Hg , ^{193}Hg , ^{194}Hg , ^{194}Tl , ^{195}Tl , ^{194}Pb , and ^{196}Pb . Figure 2 is a map of these nuclei as well as those studied by other laboratories. Current nuclear physics research activity around the world is concentrated on defining

Figure 1. Spectrum in coincidence with the transition $E_\gamma = 496$ keV and the sum of all other band members (except the 215- and 732-keV members). Members of the SD band are labeled by transition energy. Known ^{192}Hg low-lying transitions are identified by spins and parities. Asterisk indicates transitions in ^{193}Hg .

the limits of this region of superdeformation. Searches have been made in the neutron-deficient nuclei ^{188}Hg , ^{189}Au , and ^{188}Pt with negative results. Studies with heavy-ion reactions show that all of the more neutron-rich nuclei in this region possess SD bands. Access to many nuclei in this region with heavy-ion reactions (^9Be or heavier) is limited by currently available target-projectile combinations. No results for nuclei with $Z > 82$ have been reported, although studies of bismuth, polonium, and radium nuclei are planned or under way.

Spins of Superdeformed Band Members

We have developed a procedure to estimate level spin for the SD bands. Information on level spin is important for a detailed comparison with theoretical predictions. In particular, apart from level lifetime, this information is a key identification of

the spin-0 bandhead. The regular behavior and spacing of the transition energies of SD bands, together with other measured properties, suggest that they follow the well-known rotational formula for level energy as a function of angular momentum:

$$E_I = \hbar^2/2[I(I+1) - K^2], \quad (1)$$

where I is the moment of inertia, I is level spin, and K is the K quantum number. The low gamma-ray energies indicate that they are near the bottom of the cascade where the level spin I can be extracted reliably since the transition energy is most sensitive to spin change at low spin. In practice, we use an expansion in rotational frequency ω because it converges better than an expansion in spin.

The level spin as a function of rotational frequency is given by the relation:

$$I^* = 2\alpha + 4/3\beta\omega^3 + \dots, \quad (2)$$

where $I^* = I(I+1)$ is the spin, α and β are the inertial parameters, and $\hbar\omega = E_\gamma/2$ is the rotational frequency. The corresponding equation for the dynamic moment of inertia is:

$$J^{(2)} = 2\alpha + 4\beta\omega^2 + \dots \quad (3)$$

A least-squares fit to Equation (2) is made using I_f (the final state spin), α , and β as fitting parameters.

As shown in Table 1, 16 SD bands in the $A \approx 190$ region have been fitted to extract the spins of the levels. In this table, bands are identified by the nuclide and the energy of the lowest-energy band transition. Band energies were first fitted with I_f , α , and β , all free parameters. The value of I_f was then fixed to the nearest integer or half integer, and α and β were fitted again. Final fits are shown.

Figure 2. Map of the known SD bands in the $A \approx 190$ region. The property displayed is the experimental dynamic moment of inertia $J^{(2)}$. The scales of all plots are identical. Different types of data points indicate separate SD bands in nuclei with multiple bands.

The spin values of the lowest band members I_i deduced by this procedure are very close to integer or half-integer values, as they should be. Direct evidence that the spins deduced by this method are correct has not been obtained because transitions connecting the SD bands to normal states have not been observed. However, these results are consistent with observed feeding to levels of known spin, giving indirect support to the deduced spins. The SD bands we have identified in ^{194}Pb and ^{196}Pb exist down to spin 6, the lowest spin observed for superdeformed states outside the actinide region.

Moments of Inertia

Two moments of inertia are of interest in the characterization of the SD bands. The kinematic moment of inertia is related to the first derivative of the excitation energy with spin $I^{(1)} = \hbar^2 I / \omega$ and is associated with the overall rotation of the nucleus. The dynamic moment of inertia is related to the second

derivative $I^{(2)} = \hbar^2 / dI/d\omega = 4 \hbar^2 / \Delta E_{\gamma}$. The term $I^{(2)}$ describes the response of the nucleus to torque. The term $I^{(1)}$ can be determined reliably in the $A \approx 190$ region because we have determined the level spins. From Equation (2), $I^{(1)}(\omega = 0) = 2\alpha$. Table 1 shows the values of α for the 16 SD bands fitted with Equation (2), with the spin fixed at the nearest integer or half integer. They cluster in two groups with values of $4.415(13) \times 10^{-2}$ and $4.672(10) \times 10^{-2} \hbar^2/\text{keV}^2$, respectively. The grouping suggests that the SD bands have a common starting point, depending on whether they are yrast, i.e., lowest energy level for a given spin ($\alpha = 4.415 \times 10^{-2}$), or two quasi-particle ($\alpha = 4.672 \times 10^{-2}$) bands. This result can be understood qualitatively because calculations show that the deformed minimum in the potential-energy surface at a constant mass quadrupole moment is roughly independent of A and Z . The increased moment of inertia of the quasi-particle SD bands relative

to the yrast SD band in the same nucleus reflects blocking (suppression of properties associated with filled orbitals or paired particles) near the Fermi surface by excited quasi-particles.

Excitation Energy of Superdeformed Bands

An important question about SD bands is their excitation energies. The precise way to determine the excitation energy of a band is by identifying the gamma rays that connect the levels in the SD band to normal levels in the nucleus. To gain insight into the excitation energy of SD bands in these nuclei, we have assumed that the SD band becomes yrast at the transition, which exhibits one-half the maximum band intensity. In ^{194}Hg , that transition connects levels with spins 36 and 34. To improve the determination of the yrast sequence of normal states in ^{194}Hg , we have extended the known level scheme to approximately spin 34. A reasonable positioning of the SD band in ^{194}Hg results in the spin-10 SD band member being about 2.2 MeV above the lowest 8^+ level. The spin-0 SD bandhead is then at about 4 MeV after extrapolation. Three-dimensional, microscopic Hartree-Fock calculations, which yield an SD minimum that is 4 to 5 MeV above the ground-state minimum, agree with our estimate of the excitation energy for the SD bandhead in ^{194}Hg .

Alignment in Superdeformed Bands

An unusual property we observed in our data is the equality of the transition energies for certain pairs of SD bands in different nuclei (Figure 3). The transitions of an excited SD band in ^{194}Hg (band 2) are within about 1 keV of those of the yrast SD band of ^{192}Hg over a spin range of

Table 1. Results of least-squares fits to Equation (2) for 16 superdeformed bands.^a

Nuclide	Level energy, E_{γ} (keV)	α ($10^{-2} \hbar^2/\text{keV}$)	β ($10^{-6} \hbar^2/\text{keV}^3$)	Spin value, I_i
^{190}Hg	360.0	4.129(3)	8.51(6)	14
^{191}Hg	350.6	4.444(5)	6.48(10)	29/2
^{192}Hg	214.6	4.366(2)	9.03(8)	8
^{194}Hg	254.3	4.430(1)	8.49(4)	10
^{194}Pb	169.7	4.398(4)	8.33(16)	6
^{196}Pb	215.8	4.363(5)	6.13(14)	8
^{191}Hg	292.0	4.713(3)	5.21(8)	25/2
^{191}Hg	311.8	4.709(4)	6.14(9)	27/2
^{193}Hg	293.6	4.663(7)	6.45(18)	25/2
^{193}Hg	234.5	4.662(8)	6.37(25)	19/2
^{194}Hg	201.2	4.677(2)	6.23(8)	8
^{194}Hg	262.5	4.682(4)	6.38(11)	11
^{193}Tl	228.1	4.777(5)	5.07(2)	19/2
^{193}Tl	248.3	4.773(4)	5.92(11)	21/2
^{194}Tl	268.0	4.988(5)	4.08(14)	12
^{194}Tl	240.5	4.767(8)	5.30(19)	10

^aFinal fits are shown, as described in text. Values in parentheses for α and β represent uncertainties.

$20\hbar$ (Figure 4). Furthermore, a second excited band in ^{194}Hg (band 3) has transitions with energies that are close to the midpoint energies of the ^{194}Hg band and, consequently, to the midpoint energies of the ^{192}Hg band. This equivalence of frequencies, within about 0.1%, is unexpectedly small and is not understood at present.

We can determine the alignment of the ^{194}Hg bands relative to that of the ^{192}Hg band at the same rotational frequency because we know the level spins. These alignments are plotted as a function of rotational frequency in Figure 5. It is remarkable that this aligned angular momentum reaches a value of $1.00(2)\hbar$ over the last half of the observed bands. A plausible explanation of this result is that the intrinsic spins ($1/2\hbar$ each) for the two extra neutrons have aligned with the rotational axis, while the orbital angular momentum remains strongly coupled to the nuclear symmetry axis. Such behavior at high spin is related to the concept of pseudospin.

Search for Shape Isomers by Light-Ion Reactions

Heavy-ion reactions have led to the identification of superdeformation at low spin in the $A \approx 190$ region, but we have not yet found a spin-0 long-lived shape isomer. The SD cascade does not continue to the bandhead but, rather, depopulates suddenly before the bandhead is reached. Theorists are now beginning to address the question of depopulation of the SD bands at low spin before the bandhead is reached. The best idea currently is that superdeformed bands do exist down to the bandhead, but that gamma-ray branching out of the band dominates decay between low-lying members of the band. Nevertheless,

Figure 3. Rotational spectra of (a) the ^{192}Hg band, (b) one of the excited SD bands in ^{194}Hg (band 2), and (c) the second excited band in ^{194}Hg (band 3). The close agreement in the transition energies is evident in spectra (a) and (b). The transition energies in spectrum (c) are midway between those in spectra (a) and (b).

Figure 4. Differences in the energy of the transitions in band 2 in ^{194}Hg (round data points) from those of the band in ^{192}Hg as a function of rotational frequency. The square data points are the differences of the energies of band 3 in ^{194}Hg from the midpoints of the ^{192}Hg energies.

Figure 5. Relative aligned angular momentum for band 2 (round data points) and band 3 (square data points) in ^{194}Hg relative to the band in ^{192}Hg as a function of rotational energy.

the shape isomer might be populated directly by light-ion reactions, or possibly beta decay, and studied by sensitive spectroscopic techniques. We have, therefore, begun experiments to produce and identify shape isomers by light-ion reactions.

We are producing ^{196}Hg in a light-ion $^{197}\text{Au}(p,2n)$ reaction and using electron spectroscopy to identify high-energy $E0$ transitions, a signature of a shape isomer. We have completed a trial experiment on ^{90}Zr to evaluate the merits of internal conversion vs internal pair production to identify high-energy $E0$ transitions. We have recently received a thick electron detector that was designed and constructed especially for this project, and we are performing a high-statistics experiment to identify shape-isomer candidates in ^{196}Hg .

Population of Shape Isomers in Radioactive Decay

We are searching for population of shape isomers in the beta decay of neutron-deficient thallium nuclei. Alpha decay is an energetically possible shape-isomer decay mode that can be identified with high sensitivity. Because the barrier for alpha decay is decreased at the end of a nucleus with an elongated shape, alpha emission from an SD state might be enhanced.

We have looked for alpha emission by shape isomers in ^{186}Hg and ^{188}Hg populated by beta decay of ^{186}Tl and ^{188}Tl , respectively. Within experimental error, we did not observe an alpha transition in the 7.0- to 12.0-MeV energy range that would correspond to decay from an SD state to the ground state of the daughter. For ^{186}Hg , we set an upper limit of $10^{-6}\%$ for such an alpha transition.

Search Techniques in Multiparameter Data

The use of heavy-ion reactions in conjunction with large multidetector arrays has resulted in a vast amount of multiparameter data. More than 10^8 three-parameter (three high-resolution energy signals) events are not uncommon. Current analysis techniques use projections of these data into one dimension and, in the process, forfeit much information. We are investigating the use of two dimensional arrays to search for patterns in the data that result from features, such as SD bands. We are encouraged by our tests using the ^{192}Hg data that contain a well-established SD band. Our first tests have used simple truncation techniques and a Fourier-transform mask with the known spacing of the SD band in ^{192}Hg . The SD band is clearly evident in the two-dimensional array when analyzed in this manner. We hope to extend this work to three dimensions to search for weak SD bands or bands in which interference from other, stronger transitions appears in projected data.

Conclusions

The aim of our experimental program is to locate shape isomerism in nuclei where fission is forbidden. Theoretical calculations suggest that the $A \approx 194$ region will include shape isomers, and we are concentrating our efforts there. We are searching with both light- and heavy-ion reactions as well as those with radioactive decays. We have identified rotational bands in elongated nuclei in the $A \approx 194$ region using heavy-ion reactions. Unfortunately, the cascades do not extend to a spin-0 bandhead in the nuclei studied to date. Together with our collaborators, we have identified 18 SD bands

in seven nuclei; other groups have identified another ten SD bands in this region. We have developed a method to determine the spin of SD band levels and have found that the lowest spin in known bands in this region is spin 6 in ^{194}Pb . We have also developed substantial systematic data on superdeformation in the $A \approx 194$ region. Our work provides a testing ground for the nuclear theory that predicts shape isomerism in nuclei where fission is forbidden. ■

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Multidisciplinary Accelerator Mass Spectrometry

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...the frequency of stomach cancer in the U.S. Our initial measurements, shown in Figure 1, were one order of magnitude more sensitive than the previous best technique (³²P postlabeling) and were three to five orders of magnitude more sensitive than the previous best quantitative techniques using immunoassay methods. As shown in Figure 1, we did not reach the normal abundance limit for detecting ¹⁴C in biological materials (¹⁴C:¹²C ≈ 1:10¹²); our

2-amino-3,8-dimethylimidazo(4,5-f)-quinoxaline (referred to by its short name, MeIQx). DNA adducts are defects formed by the covalent binding of certain chemicals to DNA. MeIQx is a mutagen and carcinogen produced by cooking meat at high temperature, and its consumption may be partly responsible for the observed frequency of stomach cancer in the U.S. Our initial measurements, shown in Figure 1, were one order of magnitude more sensitive than the previous best technique (³²P postlabeling) and were three to five orders of magnitude more sensitive than the previous best quantitative techniques using immunoassay methods. As shown in Figure 1, we did not reach the normal abundance limit for detecting ¹⁴C in biological materials (¹⁴C:¹²C ≈ 1:10¹²); our

samples were handled in laboratories previously used for ¹⁴C tracer studies and, thus, were contaminated.

In FY90, we performed similar measurements with the acutely toxic chemical dioxin (2,3,7,8-tetrachlorodibenzo-*p*-dioxin, or TCDD). TCDD is a potent animal carcinogen that causes tumors, teratogenic effects, and other biological defects, but whose metabolic products do not bind to DNA. Figure 1 also shows data for these measurements. Improved protocols for sample preparation that remove all contamination allowed us to reach the natural abundance level of ¹⁴C. This accomplishment demonstrates that labile ¹⁴C in bulk tissue does not contribute background to the measurements. As expected, dioxin metabolites did not bind to DNA,

In FY89, we began a multiyear program to exploit the new LBNL capability in accelerator mass spectrometry (AMS). The thrust of the AMS program is threefold:

- Develop new research applications, especially in the biomedical sciences.
- Exploit the high-throughput capabilities of our spectrometer for research in archeology, the geosciences, and environmental studies.
- Develop new technologies and optimized spectrometers for spinoff to the marketplace.

In FY90, we made accomplishments in all three areas.

Biomedical and Clinical Sciences

In our initial experiments during FY89, we measured the dose-response curve for the production of DNA adducts by the mutagen

Figure 1. Measurement of binding MeIQx and TCDD metabolites to mouse liver DNA expressed as multiples of Modern, the current ¹⁴C content of living material. The MeIQx data represent an improvement of greater than one order of magnitude over previous techniques.

except at the highest doses (corresponding to LD₅₀, the dose at which 50% of the animals die from toxic effects) for mice. Such effects were expected given the toxicity of dioxin at that dose. The significance of our measurements with TCDD and MeIQx is not merely that AMS may be used to study genotoxicity at low doses but that AMS has great potential value as a screening tool for genotoxicity in drugs or other chemicals of potential interest. Our results were published in FY90 as the first biomedical application of AMS (Turteltaub et al., 1990).

In similar experiments with collaborators at the National Cancer Institute, we have begun measuring adduct formation in the lymphocytes of monkeys given doses of 2-amino-1-methyl-6-phenylimidazo(4,5-*b*)-pyridine (PhiP), a possible human carcinogen. Detection of adducts in lymphocytes will allow us to make time-history studies of damage and repair mechanisms.

Although the sensitivity demonstrated in our measurements is approximately that required to study mutagens and carcinogens at

the levels to which humans are environmentally exposed, greater sensitivity would be useful to study the dose and time dependence of DNA damage and repair processes. The instrumental sensitivity of AMS for detecting ¹⁴C is at least two orders of magnitude below the natural abundance limit of ¹⁴C. To take advantage of this potential gain, we have begun a program of growing ¹⁴C-depleted research organisms using food wholly or partially synthesized from petroleum stocks. Table 1 summarizes the results of our initial work. We have shown that methanotropic bacteria grown on ¹⁴C-depleted methane contain 1% of the normal ¹⁴C concentration.

In a much more difficult experiment, we showed that the first generation of mice grown on a diet containing natural, but indigestible, cellulose plus protein synthesized from petroleum show a ¹⁴C depletion of 30%. If the ¹⁴C content can be reduced to 1 to 10% of normal in successive generations, a valuable research animal will be produced. Such a host would be useful for chemical dosimetry and genotoxicity research and for fundamental work in nutrition because any natural compound added to the diet would be inherently tagged with ¹⁴C.

In a collaborative experiment with researchers at the University of California, San Francisco, we conducted the first experiment using AMS to investigate a human disease. We used AMS to demonstrate that alkanes, single-bonded carbon chain compounds, present in the skin of patients suffering from ichthyosis (a congenital disease causing stiffening and scaling of the skin) were exogenous. The alkanes were transported to the test area of the skin from portions of the body being treated with petroleum-based emollients. We

performed these measurements using small patch samples from the skin of two individuals. Previous research using conventional ¹⁴C-decay counting required scraping samples from 16 individuals and was subject to contamination. In contrast, we separated ¹⁴C-free alkanes from Modern skin lipids by column elution. (Modern is defined as the 1950 radiocarbon content of the biosphere.) Our work shows that clean separation of labeled and unlabeled compounds will be possible with this commonly used chemical technique.

In FY91, we will shift our efforts to time-dependent studies of damage and repair processes, investigating dose-dependent effects. In a parallel Laboratory Directed Research and Development (LDRD) project, we plan to develop a radioimmunoassay technique that will make possible low-level dosimetry without the use of ¹⁴C labels.

Archeology, Geosciences, and Environmental Studies

AMS was first extensively developed and applied as a radiocarbon tool for studying human artifacts. In collaboration with archeologists, we continue to access tools that can be applied to reconstruct the paleoclimatology records useful for testing computer models used to predict climate change. A particularly significant accomplishment at LLNL in FY90 was the development of a new technique for sample preparation that will help remove uncertainties in such records. The age and dietary information obtained from carbon isotopic determinations on ancient bone is often contentious because of possible contamination of bone protein either by exogenous materials in situ or contamination in laboratory procedures.

Table 1. Radiocarbon content of materials and tissues in AMS biological studies.

Substance	Fraction of Modern ^a
Corn oil	1.2
Acrylamide	0.1
ANU sugar ^b	1.51
Methanotropic bacteria	0.01
Normal mouse diet	1.3
Depleted mouse diet	0.78
Depleted mouse tissue	0.5–0.7
Depleted mouse liver DNA	0.7

^aCurrent biological material is ~1.2 Modern. Modern is defined as the 1950 radiocarbon content of the biosphere.

^bAustralian National University sugar—a calibration standard.

A method we invented and tested at LLNL in FY90 (Nelson, 1991) uses ninhydrin (7,2-dihydroxy-1,3-indandione) to release the carboxyl carbon of free amino acids directly as CO_2 . The reaction is structurally specific, so it is highly selective and discriminates against all contaminants except foreign amino acids. Furthermore, because the reaction product is CO_2 , the method eliminates the nondiscriminatory combustion that is the normal first step in preparing a sample for conventional or AMS radiocarbon analysis. Interference from foreign amino acids may also be eliminated because no reaction takes place with intact protein material. By extracting for analysis only insoluble protein, hydrolyzing this protein to its constituent amino acids, and then reacting them, we avoid all possible contaminants except insoluble protein. Insoluble protein is an unlikely contaminant in bone. In initial tests on bone samples previously dated to 53 000 y before present, this sample preparation method and analysis on our spectrometer gave a date of 54 000 y. The agreement is excellent and represents the oldest date we have measured.

In FY90, we measured more than 100 samples to investigate the origins and cycling processes of sedimentary and dissolved organic carbon. These reservoirs, often referred to as humic material, play a large but poorly quantified role in the global carbon cycle and have been proposed as significant sinks for anthropogenic CO_2 . Sedimentary and dissolved organic matter is composed of complex mixtures of organic compounds present in low concentrations. Thus, the small-sample capability of AMS allowed detailed study of these materials for the first time.

In our first project, we measured the radiocarbon ages of organic material in marine sediments from both cores and suspended traps for samples taken from the continental margin of the east coast of North America. The measurements tested the hypothesis that primary photosynthetic production in this organic-rich area serves as a major sink of anthropogenic CO_2 . The age of both bulk organic matter and of foraminifera shells showed that the source of the organic matter is resuspended older sedimentary matter rather than new production. In addition, the longer core records showed that this mode of deposition has been operating since just after the end of the last glacial period. We conducted this work with Lamont-Doherty Geophysical Observatory collaborators as part of the DOE Shelf Edge Exchange Processes (SEEP) program.

The SEEP radiocarbon results raise the question of what determines how much organic carbon is preserved in sediments. We are studying samples of the well-characterized sediments of Cape Lookout Bight, North Carolina, to test different fractionation techniques in an attempt to separate and characterize organic matter of marine and terrestrial origin.

In support of new research in the atmospheric sciences and hydrology, we have developed the accelerator and spectrometer tunes necessary for AMS measurement of ^{10}Be , ^{26}Al , and ^{36}Cl . In FY91, we will measure ^7Be and ^{10}Be in the troposphere and stratosphere to provide atmospheric chemistry and mixing data to constrain global circulation models. We will use measurements of ^{36}Cl in natural groundwaters to characterize the hydrology of the Yucca Mountain repository site and

water samples containing bomb-induced ^{36}Cl to monitor the leaching of radioisotopes from test points at the Nevada Test Site. The latter effort is one component of work on tracing the movement of these contaminated groundwaters toward aquifers used as water supplies for agriculture or human consumption.

Finally, we will continue to measure bomb-induced ^{36}Cl in building materials from Hiroshima. This project, scheduled to receive National Academy of Science support in FY91, will move from the Nuclear Structure Laboratory at the University of Rochester, New York, to the LLNL tandem accelerator as ^{36}Cl AMS measurements become routine.

Spectrometer Performance and Technology Transfer

In FY90, we performed more than 1000 ^{14}C AMS measurements using less than 20% of the beam time of our tandem accelerator. Of the ~1000 samples, approximately 600 were for LLNL research, 200 for University of California collaborators, and 200 for other institutional collaborators. Absolute accuracy of 0.5% was attained in the measurement of Modern samples in one run, although accuracy of 1 to 1.5% was more typical. We expect that continued improvement in spectrometer optics and accelerator and transport-component stabilization will yield a routine accuracy of 1% or better.

We have improved the spectrometer control system to include computer-driven "flat topping" of the major components of the beam transport system, allowing optimum operating points to be determined and set quickly. We also added an autoscaling feature to the controls, allowing quick and accurate switching between isotopes. In FY91, the

ion-source sample changer will be put under control of the data-acquisition computer, completing the hardware necessary for automated operation in routine analysis. We have demonstrated a spectrometer capable of 5000 to 10 000 AMS measurements annually. On two occasions, we measured more than 100 research samples in 24 h, which is the capability necessary for biomedical production work.

We have made two advances in sample-preparation technology. An occasional problem with lithium contamination in graphite samples has been traced to lithium from the quartz tubes used in combusting samples. By changing from quartz to Pyrex, we eliminated the contamination and reduced the cost of sample preparation. More importantly, a new technique for preparing hundreds of samples in parallel has been demonstrated to produce a char-like material suitable for the 3 to 10% accuracy necessary in much biomedical work. Comparison of material prepared in this fashion with more conventional graphite

samples shows that we can now perform single experiments involving hundreds of samples in a timely and cost-effective manner.

New capabilities have been developed in two directions. We obtained the ^{10}Be and ^{36}Cl tunes for the spectrometer in FY90. Research operation with these isotopes will begin in FY91. We have also begun developing a tritium AMS capability. In FY90, we completed scaling experiments with hydrogen and deuterium beams to investigate isotopic fractionation effects. A sample-preparation technique involving combustion, subsequent reduction on hot zinc, and absorption in titanium was used to prepare the materials placed in our sputtering source. We will begin experiments with tritium-tagged materials in the first quarter of FY91. The second switching magnet added to our laboratory can function as a tritium accelerator mass spectrometer. The tritium AMS capability, which was originally conceived of for arms-control applications, will now be used for biomedical and environmental monitoring.

To support possible clinical uses of AMS in the future, we have designed two notional spectrometers, one optimized for tritium AMS only and one optimized for both tritium and ^{14}C AMS analysis. The latter machine, shown in Figure 2, would occupy less than 93 m² compared with the more than 280 m² needed for the present research spectrometer. Such a machine, costing less than \$1 million and capable of simultaneous measurement of tritium and ^{14}C fraction, might access a market comparable to that for current computed axial tomography (CAT) and magnetic resonance imaging (MRI) scanners. After scientific proof of utility and technological demonstrations are completed with our existing spectrometer, we envision finding outside support and a potential industrial collaborator to construct a prototype spectrometer and associated sample-preparation systems. We filed five patent disclosures during FY90 on technical aspects and potential applications of such spectrometers.

In summary, our AMS initiative continues to produce both scientific and technological results of value in several disciplines. A transition to external funding appears probable and will provide an excellent test of the entire LDRD concept. ■

Figure 2. Optics layout for a small spectrometer optimized to measure tritium and radiocarbon fractions simultaneously by AMS.

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Massively Parallel Computing Initiative

E. D. Brooks, III

...are developing and implementing a new computer concept in which large numbers of microprocessors work in parallel to provide unprecedented computing power for the complex computational problems of interest to the Laboratory.

We are on the threshold of a fundamental change in the approach to building supercomputers. Whereas traditional supercomputers use one or a few powerful but expensive processors, the new approach will use large numbers of high-performance and relatively inexpensive microprocessors working in parallel. In recent years, great performance gains have been made in microprocessors, and they are beginning to overtake traditional supercomputers for scalar applications. Moreover, within the next few years, they may rival supercomputers in vector-processing performance. This will make possible a new breed of supercomputers, each composed of hundreds or thousands of high-performance microprocessors.

To help the Laboratory exploit these new machines, we proposed the Massively Parallel Computing Initiative (MPCI) to study massively parallel computing, which includes the participation of many groups throughout LLNL as well as active collaboration with other DOE laboratories, industrial research and development groups, and universities. In this study, we focus on short-term progress that

will enable the Laboratory to exploit massively parallel machines for production use in the 1992–1993 time frame. The study incorporates some long-term research as well, including studies of parallel language development and parallel graphics-rendering algorithms.

An essential component of the project is an on-site parallel-processing machine of substantial size together with the core staff to adequately support it. In selecting a machine, we chose one that can support a wide range of parallel-programming models because we do not know which model, if any, will have general utility at LLNL. Therefore, to support both message-passing and shared-memory applications, we selected the BBN TC2000 multiprocessor (also known as the Butterfly II), a scalable machine manufactured by BBN Advanced Computers, Boston, Massachusetts.

We also have access to remotely sited message-passing machines and single-instruction, multiple-data (SIMD) machines.

During FY90, we launched an aggressive effort in parallel computing. Half of the 126-processor BBN TC2000 was delivered midyear (shown in Figure 1). While waiting for delivery of the machine, we selected the parallel-programming models to be supported on it. We implemented initial versions of these models using the Alliant FX8 and available remotely located parallel machines. Then we transferred the programming models to the BBN TC2000 shortly after it arrived at the Laboratory.

We also developed the first version of a gang scheduler for the BBN TC2000. The gang scheduler provides for time sharing of the multiprocessor resource among several users, a feature that is lacking

Figure 1. The BBN TC2000 multiprocessor, also called the Butterfly II. In FY91, we will upgrade the machine to its full complement of 126 processors.

on other parallel machines. In addition, we developed the communication infrastructure required for such a heavily used computing resource, including internal and external network connections, LLIX and Telebit modem connections, X display support, and dump facilities.

The MPC1 project has broad participation from applications groups inside and outside the Laboratory, as reflected by the number of user accounts we maintain on the BBN TC2000 (currently 128). Some of the parallel-programming models used by our research collaborators are the Parallel C Preprocessor (PCP), the Parallel Fortran Preprocessor (PPF), the Livermore Message Passing System (LMPS), the Uniform System (supported by BBN Advanced Computers), the Intel message-passing system (available remotely on Intel iPSC machines), and Connection Machine Fortran (available remotely on SIMD machines).

The PCP, PPF, and LMPS programming models, which are all supported by our staff, are frequently changed in response to requests and suggestions from experienced users. In addition, some members of the research staff work in computer science-related areas, for example, to further develop the Sisal programming language.

Among the applications being pursued by our research staff are the following:

- Three-dimensional, finite-element neutron transport.

- Cold-matter Monte Carlo photonics.
- Linear algebra.
- Symbolic implicit Monte Carlo photon transport.
- Solution of Maxwell's curl equations.
- Molecular dynamics used in tool-cutting simulations.
- Mixed-zone Eulerian hydrodynamics.
- Geophysical fluid dynamics.
- Independent-time-step hydrodynamics.
- Parallel image rendering.
- Lattice-gas hydrodynamics.
- Plasma simulations for fusion energy devices.

We have obtained good performance for scalar-dominated codes, as was expected from the relatively high performance of current RISC microprocessors on scalar-dominated code. Using 32 processors on the BBN TC2000, we obtained performance as much as ten times that of a single-processor Cray X/MP on symbolic implicit Monte Carlo photon transport. For a partially vectorized code, such as the three-dimensional neutron-transport code presented in Dorr and Ferguson (1990), we have obtained performance equal to that of a single-processor Cray Y/MP using 48 processors. We also have worked on highly vectorized codes, obtaining nearly 20% of Cray Y/MP performance using 48 processors on a linear-system solver.

Future massively parallel machines will be far more powerful. The fastest microprocessor currently available is four times as fast, on

scalar-dominated code, as the Motorola 88000 used in the BBN TC2000. Single-chip vector processing units with 200-MFLOP performance were announced in FY90. We expect that large-scale parallel machines incorporating such vector-processing units will appear in 1 to 2 y.

In FY91, we should be able to reap substantial rewards, given the major investment in parallel-processing infrastructure expended during this first year of the project. We plan to bring our parallel-programming models to maturity and to further develop the gang scheduler to incorporate fair-share scheduling. We also want to bring on-line the high-performance, full-color graphics display, which includes sophisticated equipment for generating movies using parallel image-rendering algorithms. In early FY91, we will upgrade the BBN TC2000 machine to its full complement of 126 processors. Finally, we will begin to develop a code to demonstrate the capability of highly parallel machines on algorithms that couple radiation and hydrodynamics, which is typical of applications at LLNL.

Massively parallel machines will offer the Laboratory an entirely new computational capability in the near future. This project is helping LLNL prepare to take advantage of the new breed of supercomputers. ■

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Laboratory-Wide Competition

Exploratory Research and Development FY90

Laboratory-Wide Competition

S. Niemeyer

ER&D annually awards funding to LLNL investigators following an open, Laboratory-Wide Competition. These awards provide seed funding outside the usual programmatic channels for new scientific and technological initiatives that are still in a germinal stage. Such projects are also of a relatively small scope, with a funding ceiling of \$200,000/y for each project. The goal of the ER&D Laboratory-Wide Competition is to identify those projects of the highest scientific and technical caliber and to fund them for the short term (generally 1 to 2 y, but no more than 3 y), after which they would be able to generate their own funding from other sources.

We used two primary criteria in the FY90 proposal selection process. First, we judged the projects on their inherent scientific and technological quality. Second, among the highest-quality projects, we gave priority to those with the most potential impact on the Laboratory or the scientific community. In addition, we assessed the likelihood of successful completion of each project on the basis of the quality of the team of researchers and their research plan.

The Research Committee responsible for evaluating the projects consisted of 22 members appointed by and reporting to the Director. Chosen to represent the wide diversity of scientific and technical disciplines pursued at the Laboratory, the members serve for a maximum of 3 y. After all written proposals had been submitted, each principal investigator was interviewed by two members of the Committee, who then reported to the full Committee. The Committee ranked each proposal in order of overall scientific and/or technological excellence and pertinence to the Laboratory's mission, so as to make optimal use of the available funds.

Funded projects range from laboratory experiments, both small and large, to theoretical work and computational simulations. The span of disciplines is impressive, as one can readily determine by the following short reports for each of the funded projects. For example, a team of Biomedical researchers engineered new mammalian cell lines that have enhanced sensitivity to detecting chemically induced mutations. Investigators from the Chemistry and Materials Science Department constructed a model of butane cracking and used it to show that although pressure accelerates cracking under most laboratory conditions, it inhibits cracking at temperatures and pressures relevant to petroleum exploration. A team from the Physics Department developed a technique for efficiently extracting highly charged ions from an electron-beam ion trap and began to exploit the new opportunities for studying ion-matter interactions with slow ions (eV) carrying up to a few hundred keV potential energy.

A theoretical study by researchers from Magnetic Fusion Energy identified a parameter regime wherein intense, pulsed lower-hybrid waves can be used to heat and drive current in the core of reactor-grade tokamak plasmas. As a final example of FY90 projects, a team from Lasers used x-ray irradiation to produce plasmas at densities and temperatures of interest to astrophysics, inertial confinement fusion, and atomic physics; in addition, they designed a Schwarzschild microscope suitable for focusing x-ray lasers for use in nonlinear optics experiments.

In FY90, the Research Committee considered 134 proposals requesting \$17 million in funding. Of these, we granted 27 awards for a total of \$3.292 million, which includes \$40,000 for capital equipment. The awards ranged from \$25,000 to \$200,000; the average FY90 award was \$125,000. Fourteen projects were renewals of projects that were funded in FY89. Of the thirteen newly funded proposals, four were for multiyear projects. ■

Structure of Human Sperm Chromatin after Sperm-Egg Fusion

B. F. Brandriff, L. A. Gordon, and D. Pinkel

Using highly specific DNA probes to stain sperm chromatin after sperm-egg fusion, we determined the arrangement of repetitive sequences that formed pronuclei and the distribution of DNA replication sites during genetic recombination in sperm chromatin

The DNA-protein complex in a mature sperm nucleus exists in a highly condensed, genetically inert form. Following fertilization, the paternal complex, referred to as chromatin, must be reactivated genetically to function as one-half of the new organism's DNA complement, with the other half contributed by the maternal parent. Much of this transformation is brought about by biochemical molecules stored in the egg cytoplasm in preparation for fertilization and the beginning of embryogenesis.

Our objectives were twofold. First, we wanted to examine the arrangement of certain repetitive DNA sequences specific to centromeres. Centromeres are regions present on all chromosomes that function in the orderly segregation of chromosomes at cell division. Defects in this process in sperm or egg cells lead to a condition called aneuploidy, in which either too many or too few chromosomes are present in an individual's cells. One well-known example of aneuploidy

is Down's syndrome. Here, an extra chromosome 21 is present, resulting in serious physical and mental problems in a child. Aneuploidy is also frequently associated with tumor cells.

The centromeric DNA sequences used in our study were from two classes of repetitive DNA called satellite DNA and alphoid DNA. In repetitive DNA, the same short sequence of base pairs is tandemly arranged and repeated many hundreds or even thousands of times.

The interrelationship between centromere function and repetitive DNA sequences is still largely unknown. To address this question, we took advantage of the uniquely extended chromatin conformation in the male DNA complement. The greatly expanded distribution of male chromatin is one characteristic of the reactivation process once sperm-egg fusion has taken place. Thus, we focused on the period during which chromatin is extended in length and volume to examine the arrangement of repetitive DNA sequences known to be located at the centromeres of human chromosomes.

Our second goal was to determine the spatial distribution of the initiation sites of DNA replication in the fertilized-egg stage, also known as the zygote stage. The purpose of this experiment was to test the hypothesis that zygote chromatin exists in an extended spatial distribution regardless of the state of DNA replication. Work on somatic cells shows that DNA is most highly decondensed during the

replicative part of the cell cycle. We wanted to show that the highly extended conformation of zygote chromatin is an inherent characteristic of that stage, rather than one dependent on replication status, because chromatin conformation has been implicated in the regulation of gene expression.

For this work, we used a model system involving fusion between individual human sperm cells and hamster eggs. Such fusion activates the hamster eggs so they process human sperm chromatin correctly during the first cell cycle, after which they die. This model system made it possible for us to use molecular DNA probes available for specific regions of human chromosomes, including centromeres. The cloned probes were hybridized to pronuclei fixed on microscope slides. They were detected with fluorescently labeled reporter molecules viewed through a light microscope equipped with fluorescence capability. In our two-color detection system, each probe was labeled with a different fluorescent dye. Using this system, we analyzed the arrangement of alphoid and satellite DNA sequences on human chromosomes 1, 7, and 9.

On chromosome 1, we found that alphoid and satellite sequences did not intermingle but, instead, were distributed in adjacent blocks along the chromatin fiber. Both of these blocks were located in the long arm of the chromosome. On chromosome 7, in contrast, one alphoid repeat block (referred to as MGB) was located on the short arm,

and a second block (tet) was located on the long arm. On chromosome 9, fluorescence signals from enzymatically generated probes (supplied by H.-U. Weier of the LLNL Biomedical Sciences Division) for alphoid and satellite sequences were intermingled, possibly because of the techniques used in the synthesis of these probes.

We also used an enzymatically generated probe for a 17-base-pair repeat sequence known to be associated with the kinetochore, the region of a centromere that is most active in chromosome segregation. In early experiments, we screened kinetochore probes mapping to multiple centromeres. We identified one probe mapping to a single chromosome, most likely reflecting the chromosome-specific, nonrepetitive sequences flanking the repetitive sequences. Using the results obtained to date, we will next construct a high-resolution map of the centromere of chromosome 19, for which many probes generated at LLNL are available.

To examine the spatial distribution of DNA replication sites, we pulsed fused eggs with bromodeoxyuridine (BrdUrd), an analog of one of the four bases in native DNA. We then located the incorporated BrdUrd with two fluorescently labeled antibodies after fixation of pronuclei on microscope slides. Many replication sites, shown as the white spots in Figure 1, were distributed throughout the pronucleus. We estimate approximately 4000 sites. This result is in contrast to findings for somatic cells (cells other than sperm and egg), in which

replication sites are organized into higher-order structures numbering no more than 300 per nucleus. As DNA replication progressed, the discrete pattern disappeared and continuous labels began to outline completely replicated DNA strands.

The large number of replication sites visible in the zygote pronuclei, compared with those in somatic nuclei, may well indicate that pronuclear chromatin exists in a state of highly extended, undercondensed spatial distribution. The same conclusion is supported by the highly extended state of repetitive DNA sequences seen near centromeres. A two-color analysis combining replication-initiation sites and DNA-probe signals confirmed that the extended chromatin existed independently of DNA replication activity.

Chromatin organization is involved in regulating replicative and transcriptional activities of the

genome. Therefore, we surmise that an extended spatial distribution may function in the genetic reactivation of the paternal genome after its quiescence in the mature sperm head. The expanded conformation may also underlie the special vulnerability (Russell et al., 1988) of one-cell newly fertilized eggs to mutations. Our novel approach to studying centromere structure will help elucidate the function of repetitive DNA sequences in the orderly segregation of chromosomes. Once such mechanisms are elaborated, we will have a better understanding of defects that interrupt this process and lead to aneuploidy, which represents a major health hazard. ■

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Figure 1. Zygote pronucleus labeled with an antibody to detect DNA synthesis. Each white spot represents an initiation site for DNA replication. Approximately 4000 such sites are observed per pronucleus.

Bioremediation of High-Explosive Wastes

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We have defined conditions under which microorganisms can degrade RDX and HMX and have designed a flow reactor for the continuous treatment of high-explosive-contaminated water.

Research and development activities involving high explosives (HE) at federal facilities, including DOE laboratories, have resulted in the contamination of soils and groundwaters. Environmental regulations can require that remedial actions and controls be adopted that remove such contamination to acceptable levels. Water-quality criteria as low as 105 µg/L have been proposed for persistent HE contaminants, such as hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) (Etnier, 1989). Unfortunately, no technologies are currently in place to address the treatment of such dilute HE residues in the environment. Nevertheless, bioremediation (the application of biodegradation technology for the transformation of hazardous waste to harmless byproducts) is the only strategy that offers the versatility needed to treat HE in environmental media. The objective of our research is to develop a biological system for the remediation of HE-contaminated wastewaters so that environmental quality can be preserved.

Our research consists of two parts: the isolation and adaptation of a microbial population (consortium) optimized for the degradation of two common HE compounds, RDX and octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX); and demonstration of a flow reactor for the continuous treatment of HE-contaminated wastewaters and aquifers. Our approach relies on the adaptation of an indigenous consortium of aerobic microorganisms to carry out the degradation process. We selected this approach because recalcitrant contaminants, such as HE compounds, are more likely to be completely degraded by a diverse population than by a single species of microorganism. The increased likelihood is due to the broad spectrum of metabolic capabilities required to degrade such compounds to innocuous byproducts (e.g., carbon dioxide and water).

Microorganisms were obtained from HE waste streams and were cultured in batch reactors that received low concentrations of HE compounds along with a variety of nutrient preparations. Such treatment stimulates the growth of diverse microbial populations so that natural selection enhances the metabolic capabilities of microorganisms favoring HE degradation. In our initial experiments, we discovered that degradation of RDX required the presence of a separate carbon source to maintain a viable microbial population. This process, known as co-metabolism, occurs

when the compound of interest (RDX in this case) is degraded but does not provide enough energy to sustain microbial metabolic activity. This phenomenon makes the degradation of contaminants to environmentally acceptable concentrations (e.g., <100 µg/L) difficult because the microorganisms tend to metabolize the most readily available carbon source rather than the contaminant of interest. We addressed the problem by assessing the suitability of several carbon sources that will support microbial activity and HE degradation. We found that peptone (a mixture of protein derivatives) is a superior carbon source for supporting the co-metabolism of HE compounds.

We have demonstrated the biodegradation of RDX and HMX under aerobic conditions. Using ¹⁴C-labeled compounds, we verified that these explosives can be degraded to concentrations of <100 µg/L with carbon dioxide as the principal transformation product. The occurrence of this transformation under aerobic conditions is significant because the process does not lead to the formation of hazardous byproducts (e.g., hydrazines), which are known to arise from the anaerobic degradation of RDX and HMX.

We are developing a treatment system for the remediation of water contaminated with low levels of HE. We have constructed a bench-scale flow reactor designed to optimize the degradation of low concentrations of HE compounds

(Figure 1). The bioreactor consists of a large (50-L) batch system in which microorganisms are maintained on a constant diet of nutrients and HE compounds. Microorganisms grown in the system are intermittently pumped into a 4-ft-high, fluidized-bed biofilm reactor in which they become attached to suspended sand grains. Thus, we can maintain a high density of biomass that is adapted for HE degradation. Furthermore, the degradation of HE compounds present at low concentrations can be achieved in the fluidized bed because alternative carbon sources are held at minimum concentrations. We are conducting our research in cooperation with scientists from the University of California, Los Angeles, who have extensive experience in the development of bioreactors for the treatment of industrial wastes (Stenstrom et al., 1989).

The results of our studies are helping us to design an optimum bioremediation strategy for HE compounds that will subsequently be tested in the field. By establishing a system for the biological degradation of RDX and HMX, we will be in a position to rapidly develop treatment strategies for other structurally similar HE

compounds, including those that are under development (Alire, 1990). ■

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Figure 1. Diagram of the fluidized-bed treatment system developed for the biodegradation of high-explosive compounds.

Development of an Experimental Model of Human Hemopoiesis

M. G. Pallavicini and R. G. Langlois

We are using a novel approach to attempt to create an experimental model of human hemopoiesis. Primitive human hemopoietic cells are introduced into fetal mice in utero prior to the development of rodent immunological competence.

The hemopoietic system constitutes a complex, self-sustaining tissue that continuously generates cells, which then mature into blood cells. Hemopoietic tissue is located in the bone marrow, spleen, liver, and lymph nodes, where it is maintained by a small population of stem cells that self-renew, proliferate, and differentiate, ultimately producing mature cells in the circulation.

Much of the knowledge about the hierarchical structure of hemopoiesis and its regulation is based on data generated in mice. The lack of an experimental model, in which human stem cells proliferate and mature to form functional

cells in multiple lineages, has precluded in-depth analysis of the regulation of the human immune system. A better understanding of the structure and regulation of human hemopoiesis would (1) improve our understanding of the mechanisms responsible for human malignancies, such as leukemias, and for immunological disorders, such as AIDS; and (2) aid in the design of therapeutic strategies for modulating hemopoiesis, as in cancer chemotherapy and gene therapy.

To maintain human hemopoiesis in rodents, we must circumvent their immune system so that the human cells are not rejected. During the first and second trimester, both rodent and human fetuses are immunologically incompetent and, thus, lack the ability to recognize foreign antigens. Acquisition of the ability to discriminate between "self" and "nonself" is a critical process in hemopoietic development and occurs shortly before birth and/or during the first few days after birth.

We proposed a novel approach for establishing functional human hemopoiesis in animal model systems. Our approach is to transplant

human stem cells into murine fetuses prior to the development of rodent immunocompetence. Human fetal liver tissue, which contains hemopoietic stem cells, was introduced transplacentally into the circulation of murine fetuses during the second trimester. The presence of human cells in newborns and young animals was monitored using (1) sensitive DNA-based strategies that detect human nucleic acid sequences (fluorescence in situ hybridization and in vitro DNA amplification using the polymerase chain reaction), and (2) a flow-cytometric assay that measures the expression of a human red cell protein (glycophorin). Several murine models were used for transplantation of human cells, including mice with genetic defects manifested in the erythroid system (anemia) and in lymphoid lineages (lack of lymphocyte subsets).

Transplacental injection of murine fetuses is complicated. The small size of first and early second trimester fetuses limits visualization of fetal/placental anatomy as well as the injectate volume and number of cells that can be introduced. Furthermore, about 40 to 50% of the injected fetuses abort.

We have demonstrated that the increased abort rate results from the injection process itself and not from the introduction of human cells.

Our initial studies were directed toward measuring the percentage of human cells that lodge in fetal tissues at short time intervals after transplacental injection.

Fluorescence in situ hybridization with a DNA probe specific for repeated human centromeric sequences was used to detect human cells. Our data indicated that the seeding efficiency of human cells in rodent liver 1 h after injection ranged from 0 to 1%.

Approximately 25% of the injected fetuses did not contain human cells. The frequency of human fetal liver cells in rodent fetuses at longer intervals after transplantation (4 to 144 h) ranged from 0.01 to 0.1%.

These data suggest that it will be advantageous to develop techniques for introducing a higher cell number or injecting tissue containing a higher frequency of human stem cells (human fetal liver is composed of various subpopulations including hemopoietic stem cells). We are conducting studies to determine the feasibility of intraperitoneal injection of human cells into the rodent fetal abdomen. Flow-cytometric analysis and sorting of immunofluorescently labeled fetal liver cells are being used to obtain

preparations enriched in human hemopoietic stem cells. In addition, human hemopoietic stimulatory factors are being supplied to rodent mothers carrying fetuses injected with human cells. We anticipate that these regulatory factors will increase the frequency of human stem cells by stimulating their proliferation and differentiation in the rodent microenvironment.

Despite the technical limitations described above, we have had some promising successes to date that demonstrate immunological tolerance of human cells in young mice. In one mouse that received human cells (3×10^5 human fetal liver cells at 18 weeks of gestation), human cells were detected in blood and bone marrow 2.5 months after birth. The frequency of nucleated human cells in blood and bone marrow, as measured by fluorescence in situ hybridization, was 1.0 and 0.15%, respectively. Furthermore, approximately 13% of the peripheral blood erythrocytes were of human origin (measured using the glycophorin assay).

Detection of human cells 2.5 months after birth suggests that a human stem cell population was contributing to the formation of erythrocytes. In addition, the presence of human glycophorin on erythrocytes in mouse peripheral

blood (albeit at levels two- to three-fold lower than in typical human red cells) suggests that selected human cell surface proteins are being expressed. Thus, modulation of tolerance by transplantation of human cells in utero appears to abrogate immune rejection of the injected cells.

These data provide a basis for continuing studies to increase the level of human cell engraftment and to establish a model of human hemopoiesis in which human stem cells self-renew, proliferate, and differentiate to produce progeny in multiple lineages. Successful development of a model of human hemopoiesis in rodents will require demonstrating that multiple human hemopoietic lineages can be maintained for extended periods, i.e., 12 months. Establishment of chimeric mice carrying a human hemopoietic system has broad and significant ramifications for studies of human hemopoiesis in health and disease. Widespread application of the model for research studies will depend on maintaining relatively high frequencies of human cells (i.e., 10 to 20%) in the rodent environment. These goals will be the focus of studies in FY91. ■

Isolation and Characterization of the Dioxin Receptor

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We are attempting to produce a monoclonal antibody that binds the dioxin receptor found in mammalian cells by using an antidioxin antibody we previously isolated.

The dioxin receptor, also called the Ah receptor, is a protein found in the cytoplasm of mammalian cells that binds:

- Chlorinated and brominated dioxins.
- Halogenated dibenzofurans, biphenyls, and azo(xy)benzene congeners.
- Polyaromatic hydrocarbons, such as 3-methylcholanthrene and benzo(a)pyrene.
- Certain nonhalogenated aromatics, such as 5,6-benzoflavone.

The recognition and binding of these compounds is influenced by many factors, including the chlorination pattern, the degree of

planarity, and the three-dimensional space occupied by the ligand. Dioxins, especially those isomers with chlorines in the lateral positions (the most toxic isomers), bind to the Ah receptor with the greatest affinity.

We are interested in the dioxin receptor because of the biological effects thought to be mediated by that receptor complex following exposure to dioxin. The biological effects include atrophy of the thymus gland, which is important in developing the body's immune system, induction of various drug-metabolizing enzymes, promotion of tumors, and teratogenic effects. The mechanism(s) by which the dioxin-Ah receptor complex regulates these diverse biological responses is not understood, in part, because of the lack of probes for the receptor. Current models suggest that following ligand binding, the complex migrates to the cell nucleus, binds to specific sites

on DNA, and influences gene induction. The induced genes are then presumed to be responsible for the biological effects.

Our main goal is to isolate a monoclonal antibody that recognizes the Ah receptor. The antibody will then serve as a probe for studies designed to isolate and characterize this receptor. Because of the small amount of receptor present in cells, routine methods to produce an antireceptor antibody have been unsuccessful. We propose to use a unique antidioxin antibody that we have already produced at LLNL. The antidioxin antibody will serve as an immunogen resulting in the production of an anti-idiotypic antibody that should also bind the receptor (see Figure 1). The anti-idiotypic approach has been successful by other researchers studying different receptor systems in which limited amounts of receptor made the use of standard antibody methods inadequate.

Figure 1. Production of anti-idiotypic antibody that should bind the dioxin receptor. (a) The shaded area denotes the potential receptor site around one form of dioxin, whose chemical structure is shown. (b) Our previously generated antidioxin monoclonal antibody. The entire light (L) chain and a portion of the heavy (H) chain make up the region of the molecule that binds dioxin. This region of the antibody, when isolated, is referred to as the Fab fragment. The remaining portion of the antibody molecule is referred to as the Fc fragment. (c) An anti-idiotypic antibody is an antibody to the Fab portion shown in (b). The anti-idiotypic antibody should also bind the dioxin receptor shown in (a).

To generate an anti-idiotypic antibody that binds the Ah receptor, we must complete the following objectives:

- Generate Fab fragments of our antidioxin monoclonal antibody.
- Immunize animals.
- Evaluate the efficiency of immunization.
- Develop an enzyme-linked immunosorbent assay (ELISA) to screen the resultant hybridomas for anti-idiotypic activity.
- Complete cell fusions to produce hybridomas.
- Identify and clone putative anti-idiotypic monoclonal antibodies.
- Confirm the identity of anti-Ah receptor antibodies using a receptor-depletion assay.

We made progress during FY90 toward producing an anti-Ah receptor antibody and completed the generation of Fab fragments, as shown in Figure 1. The term Fab fragments refers to the antigen binding region of the antibody molecule, that is, the

entire light chain and a portion of the heavy chain. Using these Fab fragments, we have developed and evaluated a direct-binding ELISA and a competition ELISA (cELISA) for screening the hybridomas resulting from fusion experiments. (Hybridomas are hybrid cells produced by fusing an antibody-producing lymphocyte with a tumor cell and used to culture a specific antibody.) These assays are needed to evaluate the effectiveness of the immunization protocol. In both assays, the Fab fragments serve as an antigen. We also have completed a series of immunizations and have evaluated the sera from these animals for anti-idiotypic antibodies. In most cases, the animals appear to be well immunized.

Our main effort in FY91 will be to perform cell-fusion experiments generating hybridoma cells. In a typical experiment, we generate approximately 10^4 hybridomas,

which we then evaluate for specific anti-idiotypic antibody. The initial screens employ the direct-binding ELISA, then positives are confirmed using the cELISA. These hybridomas will be analyzed in a receptor-depletion assay to confirm their anti-Ah receptor character. The receptor-depletion assay will be performed at Purdue University.

We will continue the cell-fusion experiments until we identify a useful anti-Ah receptor antibody. Once such an antibody is produced, it will be used in experiments to characterize the Ah receptor. We will couple immuno-precipitation experiments with analysis by gel electrophoresis. Experiments of this type will allow us to characterize the physicochemical parameters of the Ah receptor. ■

Novel Mammalian Cells for Studying Chemical Mutagenesis

L. H. Thompson and J. S. Felton

We have used recombinant DNA techniques to generate engineered mammalian cell lines that have enhanced sensitivity to studying chemically induced mutations.

When cells within an organism are exposed to environmental mutagens, two processes largely determine the magnitude of genetic effects. First, metabolic conversions can activate compounds to electrophilic derivatives, which react with cellular DNA, and can detoxify reactive species. Second, the damage produced in DNA by covalent binding of metabolites is subject to removal by repair processes. Unrepaired DNA adducts, however, often cause mutations, which may initiate cancer or have other deleterious effects. Mammalian cell lines routinely used for the assessment of mutagenic or carcinogenic chemicals do

not express the cytochrome P450 monooxygenase enzymes that are normally expressed in cells in mutant animals and are needed for the metabolism of nonpolar organic chemicals (Thompson et al., 1987; Gonzalez et al., 1989). Thus, new cell lines that have specific cytochrome P450 enzymes will be advantageous for studying chemically induced mutations.

We introduced the genes for two cytochrome P450s (Gonzalez et al., 1984) into the widely used Chinese hamster ovary (CHO) cells using a mutant line that cannot repair bulky-adduct DNA damage. (Technically speaking, for greater ease of handling, we used cDNA "minigenes," which have only the protein coding regions.) After identifying the appropriate subclones that expressed the P450 genes, we genetically reverted the repair defect to a state of repair proficiency. Thus, for both cytochrome P450 genes, we produced pairs of genetically similar cell lines differing in repair capacity

while having matched levels of metabolic capability.

We used the transformed cells expressing the P450 genes to study a class of dietary mutagenic compounds known as amino-imidazoazaarenes (Felton et al., 1986). These compounds are produced during the cooking of fried beef and other meats. They are carcinogenic in rodents and monkeys (Ohgaki et al., 1984; Esumi et al., 1989) and perhaps also in humans.

The mouse cytochrome P₃450 (IA2 subfamily) gene was chosen because of its ability to confer metabolic activation of heterocyclic amines, such as IQ [2-amino-3-methyl-imidazo(4,5-f)quinoline] and PhIP [2-amino-1-methyl-6-phenylimidazo(4,5-b)pyridine], mutagens produced in cooked beef. In FY89, we introduced the P₃450 gene by DNA transformation procedures into repair-deficient UV5 cells, which are highly sensitive to killing by ultraviolet (UV) radiation. Clones of cells that expressed this gene were identified by preparing cell extracts and testing for the ability of the extracts to convert IQ to mutagenic metabolites in the Ames/*Salmonella* bacterial test. Clone UV5P3 was reverted to repair proficiency by treating with an ethylating agent (mutagen) and selecting for clones that were resistant to UV radiation. A resistant clone, 5P3R2, was identified that expressed the same level of P₃450 protein activity as UV5P3 cells when tested in the *Salmonella* test.

In FY90, we used the UV5P3 and 5P3R2 cell lines in dose-response assays of cell killing and mutation with PhIP (Figure 1). Parental UV5 cells, which lack the expression of

Figure 1. (a) Cell survival and (b) induced mutations at the *aprt* locus vs concentration of PhIP mutagen. (The *aprt* gene is involved in metabolism of precursors of DNA.) The frequency of mutant cells was determined after 48 h of exposure and after 6 d for mutation expression.

P450 genes, were completely nonresponsive to PhIP, but the derived UV5P3 cells were sensitive to concentrations as low as 0.1 $\mu\text{g}/\text{ml}$. As expected, 5P3R2 cells were more resistant than UV5P3 cells because of their capacity to repair DNA adducts produced by PhIP. Two important conclusions result from these experiments. First, PhIP-derived adducts are highly toxic and mutagenic in mammalian cells, particularly in the absence of repair. Second, cells are able to efficiently repair these adducts and thereby eliminate their mutagenic potential. These findings have implications for understanding the dietary significance of the carcinogen PhIP.

Similar experiments were done using the mutagen/carcinogen IQ, which is much more potent than PhIP in the *Salmonella* mutation test. However, we found IQ to be only weakly mutagenic in both UV5P3 and 5P3R2 cells. Thus, PhIP and IQ clearly differ in their relative potency between bacteria and mammalian cells. Such major differences point out the limitations of using data on bacterial mutations to infer relative human risk from exposure to these compounds.

The second cytochrome P450 gene we used in FY90 was the mouse P₁450 (IA1 subfamily) gene. The P₁450 protein has a substrate specificity that differs from that of P₃450. The P₁450 protein can metabolize benzo(a)pyrene, 7,12-dimethylbenz(a)anthracene, other polyaromatic hydrocarbons, as well as certain cooked-food-related mutagens, such as Trp-P-2 [3-amino-1-methyl-5H-pyrido(4,3b)indole]. We used procedures similar to those for the P₃450 gene but with several improvements to produce repair-deficient and repair-proficient cell lines that expressed the P₁450 gene. Figure 2 shows the expression of the P₁450

gene for experiments in which cell extracts were used to produce mutations with Trp-P-2 in the *Salmonella* test. No mutagenic activity was seen in UV5 parental cells, whereas similar dose responses occurred in UV5P1 (repair-deficient) and 5P1R1 and 5P1R3 (repair-proficient) lines. We confirmed the expression of the P₁450 gene in these lines using the 7,8-diol of benzo(a)pyrene in this assay and by measuring the enzymatic conversion of benzo(a)pyrene to the 3-OH product.

In summary, we have constructed a series of cell lines that have wide applications to both testing for mutagenesis and studying the pathways of metabolic activation of environmental mutagens. ■

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Figure 2. Bacterial mutations in the *Salmonella* test produced by Trp-P-2 mutagen in the presence of S9 extract fractions prepared from four cell lines. Three independent experiments were done; representative standard error bars of the mean are shown for 5P1R3 cells.

Application of ^{67}Zn Mössbauer Spectroscopy to Hole- and Electron-Doped CuO_2 Superconductors

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We are applying ^{67}Zn Mössbauer spectroscopy to study hole- and electron-doped CuO_2 planes in the high- T_c superconductors $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4/\text{ZnO}$ and $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4/\text{ZnO}$ to determine the carrier dynamics and the role of magnetic interactions in pairing the charge carriers.

Particle-hole symmetry is a property of single-band models of the CuO_2 planes, which focus on the partially occupied band of one-electron states describing the delocalized carriers (Anderson, 1987). In contrast, the two-band description assumes that the oxygen (2p) levels are filled and that the intrinsic holes are in the copper (3d) states leading to Cu^{+2} ions with a localized spin responsible for the observed magnetic behavior (Emery, 1987). With p -type doping, holes occur in the oxygen 2p orbitals, which move through the plane flipping the copper spins. Because the oxygen 2p orbitals of the undoped CuO_2 plane are filled, the additional electrons resulting from n -type doping must go onto the copper sites producing Cu^{+1} ($3d^{10}$) ions. The electron carrier moves through the CuO_2 plane, annihilating rather than flipping the copper spins. The seemingly different carrier dynamics for p - and n -type superconductors in this model is one basis for arguing against particle-hole symmetry. However, introduction of some hybridization of the 2p and 3d bands may introduce a limited and more complicated form of carrier symmetry.

During FY90, we carried out Mössbauer spectroscopy experiments on ^{67}Zn situated at copper sites in hole-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and electron-doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$. The 93-keV transition between the spin 5/2 ground state and the spin 1/2 first excited state is a Mössbauer transition. The electric-field gradient (EFG) at the ^{67}Zn nucleus splits the spin 5/2 ground state into three levels, whereas the spin 1/2 excited state remains unsplit. For a single-line source, the Mössbauer spectrum consists of three unequally spaced lines from which the EFG tensor components (including their absolute signs) at the ^{67}Zn nucleus can be determined. The width of the 93-keV transition in ^{67}Zn is exceedingly narrow, $\sim 5 \times 10^{-11}$ eV, corresponding to an energy resolution $\Delta E/E$ of $\sim 5 \times 10^{-16}$. This high energy resolution permits accurate measurement of changes in the isomer shift and EFGs as a function of doping levels.

We have produced ^{67}Zn in situ at copper sites in a copper foil using an alpha-particle beam from the 88-in. cyclotron at Lawrence Berkeley Laboratory by the reaction $\alpha + {}^{65}\text{Cu} \rightarrow {}^{67}\text{Zn} + 2n$. Gallium-67 decays (78-h half-life) by electron capture to excited levels of ^{67}Zn , which then decays through the 93-keV Mössbauer transition. In

Charge carriers in the high-temperature superconductor (high- T_c) $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ are understood to be holes in the normally filled O^{2-} valence shells introduced by substituting a divalent ion, such as Sr^{+2} , for La^{+3} . The discovery of electron superconductivity in similar cuprates has significant implications for the understanding of high-temperature superconductivity (Tokura et al., 1989). The new n -type Ln_2CuO_4 compounds (where Ln = praseodymium, neodymium, or samarium) are doped with Ce^{+4} , and oxygen is removed by annealing, which results in additional electrons. If the additional electrons go into the CuO_2 planes, important questions arise regarding the particle-hole symmetry of the perovskite planes

addition, we produced ^{67}Ga by irradiating a ZnO sample with protons at the tandem accelerator at Los Alamos National Laboratory.

To measure a velocity spectrum, we developed a piezoelectric velocity transducer along with the associated control electronics in association with Ranger Scientific, Inc. We checked the transducer prototype with a ^{57}Co source and ^{57}Fe Mössbauer spectroscopy and verified its satisfactory performance.

The experimental setup consists of an irradiated sample attached to the piezoelectric transducer and a standard absorber, ZnO enriched in ^{67}Zn , both contained in a liquid-helium cryostat. Radiation is

detected by a NaI detector located outside the cryostat. We have assembled and tested the completed ^{67}Zn Mössbauer experimental configuration at LLNL and Los Alamos. In FY91, we will conduct ^{67}Zn Mössbauer experiments on $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ and $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$.

We also fabricated and characterized $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ and $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$. In addition, we prepared $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{Cu}_{1-y}\text{Zn}_y\text{O}_{4-\delta}$ samples and determined their superconducting transition temperatures by superconducting quantum interference device (SQUID) measurements. The measured superconducting temperatures were 24, 17, 11.5,

and 7 K for $y = 0, 0.01, 0.02$, and 0.05 , respectively, demonstrating the relatively strong detrimental effect of zinc doping on superconductivity in these compounds. ■

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Changes in Material Structure over Extremely Short Periods

Q. C. Johnson

We are developing new techniques that will allow us to elaborate on changes over extremely short intervals in crystalline microstructure undergoing intense shock-wave compression.

Much of what we know today about the detailed structure of crystalline materials comes from studies using x-ray diffraction. The arrangement of atoms and their positions, which are precise in many cases to 100 fm, allow us to formulate models. Measurements of similar precision on materials undergoing shock-wave compression would enable us to better model the material response in such states. However, common x-ray sources lack the intensity to function over extremely short time intervals. We are exploring a method to overcome this limitation.

Our approach uses the Cornell High-Energy Synchrotron Source (CHESS) operating in its single-bunch mode to deliver a series of intense, 120-ps pulses of polychromatic x rays at 2.56-ms intervals. A pair of x-ray shutters selects a single pulse. A detector that registers scattered radiation from a foil in the synchrotron beam provides the precise timing signal for firing an electric gun. Thus, the shock wave reaches the crystal front surface within a few nanoseconds of the arriving x-ray pulse. A two-dimensional detector array (Kodak image-storage plate) will record the resulting Laue diffraction pattern.

We have built and characterized a plane-wave generator that delivers 10- to 40-GPa shock-wave pulses without damaging the nearby detector array. Our generator consists of a small capacitor bank that discharges through a strip-line conductor to a load (a narrow plate in the strip line). The sudden current vaporizes the load and ionizes the vapor into a plasma that pushes against and accelerates a thin plastic flyer sheet. The flyer hits the sample

at a velocity of 5 km/s in less than 2 μ s after the electric pulse goes through the gun. We have reduced time variations between the triggering of the electric pulse and the arrival of the shock wave at the sample surface to less than 10 ns.

In our experiments, the x-ray pulse must reach the crystal just as the shock wave arrives at the sample surface. We send the x-ray detector signal from the pulse, which comes just before the fast shutter opens, through a suitable time delay to make the gun fire at the correct time. The shock wave then coincides with the x-ray pulse.

The size, intensity, and elongation of Laue spots observed during our shock experiments will yield a wealth of information about crystalline structure, and we will examine each Laue spot for substructure. In FY90, we improved the timing circuits, made a synchrotron-compatible shock gun, and developed modeling software for analyzing Laue images. In FY91, CHESS personnel will complete the development of fast shutters, and we will schedule a dedicated, single-bunch run using the storage ring. ■

Effect of Pressure on Hydrocarbon Cracking

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We are rapidly consuming petroleum resources close to the earth's surface and must drill deeper for oil and gas. Deeper holes are expensive, and at greater depths, oil is converted to gas, which is less valuable than oil. Because of these value differences, we must know how deep we can drill and still find oil before time, heat, and pressure convert it. The effect of pressure on "cracking" (the conversion of oil to gas) is controversial and poorly understood. Laboratory experiments indicate that although oil forms 3 to 5 km deep at 125 to 150 °C and gas forms 5 to 9 km deep at 150 to 250 °C, increased pressures at greater depths may have preserved oil from conversion. However, estimating the reaction rate is difficult when the time scale is millions of years.

To test these experiments, we have developed a detailed, free-radical kinetic mechanism for the thermal conversion of butane. Butane is large enough to have many of the chemical properties of oil but small enough to model in detail. We used this model to investigate the conversion rate at temperatures from 200 to 600 °C and at pressures from atmospheric to 100 MPa. For a typical geothermal gradient of 25 °C/km of depth, a depth of 7 km corresponds to about 200 °C and 70 MPa.

Our present butane conversion model consists of 209 species and

945 reactions, including initiation and termination, addition and decomposition, isomerization, cyclization, and hydrogen transfer reactions. We used the Chemkin II, Senkin, and HCT codes to solve the resulting differential equations for isothermal, constant pressure, and batch conditions. Results from the various codes compare favorably. Each code has advantages for specific diagnostic purposes, but the Senkin code is fastest. Equation-of-state computations were included in our final calculations with Senkin to explain deviations from ideal gas behavior at high pressures. Our calculations do not include activation volume or diffusion effects, which may further inhibit the conversion of oil to gas at high pressures.

We first validated our model calculations by comparing them with experiments at 550°C and 17 MPa (Hepp and Frey, 1953). The observed and calculated reaction rates for butane disappearance were within a factor of 2, and the product distributions were similar.

Model calculations for a wider range of conditions show that the effect of pressure depends on temperature. High pressures dramatically increase the overall butane conversion rate at 600°C but tend to decrease it at 200°C, as shown in Figure 1. This decrease occurs because pressure enhances the chain propagation reaction at high temperatures but normally enhances chain termination reactions at low temperatures.

Simulations for temperatures of 300 to 375 °C—the lowest at which the reaction is fast enough to study in laboratory experiments—show that pressure can either accelerate or

inhibit the reaction rate depending on the precise temperature and extent of reaction. Therefore, laboratory experiments assessing the importance of pressure often appear contradictory. Although our model calculations are too limited to predict oil stability in real situations, they do show that pressure may have a strong influence on stability. ■

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Figure 1. Butane cracking as function of time at (a) 600 °C and (b) 200 °C, where 10^{14} s = 3.2 million y. Pressure accelerates butane cracking at high laboratory temperatures but inhibits cracking relative to atmospheric pressure (0.1 MPa) at low geologic temperatures.

Fluid Flow through Fractures in Rock

W. B. Durham and B. P. Bonner

We have measured both the mechanical and hydrologic properties of a single fracture in a rock. We have also determined its topography and analyzed its detailed shape in terms of fractal geometry. These measurements should lead to a better understanding of fluid flow in the Earth's crust.

Fractures, or joints, in rocks control or influence the mechanical and transport properties of the Earth's crust. Many important processes depend on whether fractures effectively transport fluid at depth and remain open for the time periods of interest. These processes include enhanced oil recovery, exploitation of geothermal heat, isolation of toxic and radioactive wastes, and mineralization of ore bodies. An intimate coupling of mechanical and chemical processes acts to sustain open fractures or, in contrast, contributes to their closure. Tectonic activity, increasing fluid pressure, and net mineral dissolution tend to sustain open fractures, whereas increasing normal stress and net mineral precipitation tend to close fractures.

We have begun an experimental program to measure, interpret, and predict the transport properties of typical rock joints under realistic conditions. We think that the tools are now available to achieve this understanding for individual joints. If we can establish this basis, models involving interconnected joints will be on a much sounder footing, and we will be closer to understanding the effects on hydrology of

forces that open and close joints in real rock formations in the Earth's crust. The goal of this project has been to (1) measure the hydrologic and mechanical properties of joints, as well as their topography, using the tools now available, and (2) show whether the information obtained provides a complete set of constraints to the physically correct modeling of fluid flow in joints.

On the first point—demonstrating our ability to accurately measure the requisite properties of a joint—we have had much success. We have not yet completed the second goal, that of guiding the model builders with our measurements, but we are actively pursuing this phase and are encouraged because the DOE Office of Basic Energy Sciences will begin funding the project in FY91.

Most models of fluid flow in individual joints have been based on the so-called "cube law," which holds that the rate of flow increases as the cube of the joint opening, or aperture. A complexity arises because the aperture of a rough rock joint is not single-valued, so various models have been introduced wherein a variable aperture is characterized by some effective aperture that leaves the cube law intact. One of the more important questions about fluid flow—how it changes with normal force across a joint—can then be modeled by handling the question of how this effective aperture changes as a function of normal force.

Past efforts have tended to be joint-specific, requiring, for instance, that the elasticity of a joint be known before its hydrology can be modeled. Our strategy has been to combine recent progress in

understanding joint topography—in particular, the observation that most joint surfaces have a fractal geometry—with a recently developed technique for precise three-dimensional surface profiling, to develop a generally applicable model for joint elasticity and/or joint permeability. We seek a model whose input parameters are only related to the shape of the joint, and we think that the recent revelations concerning the fractal geometry of rough surfaces mean that the apparent complexity of a real fracture surface is in fact characterizable in terms of three or four easily extractable parameters.

We can characterize the entire problem for the first time by measuring the elasticity and hydraulic conductivity of the very joints whose shapes we measure and parametrize. Figures 1 and 2 show the results of our careful measurements on a sample of Westerly granite. We began with a whole cylinder that we split axially into two halves. We profiled a 65- × 13-mm area of each side of the fracture (Figure 1a) and calculated the power spectral density (PSD) function for the joint (Figure 1b). The fact that the PSD for the single surface is straight on a log-log plot indicates that the surface has a fractal geometry. The spatial frequency at which the PSD of the composite is twice that of the single surface (here, about 2 mm⁻¹) marks the finest scale at which the opposing surfaces are correlated. We then put the cylinder halves back together and encapsulated the rock in a pressure-tight flexible jacket, attaching cantilever displacement transducers to measure the flexure of the joint.

End caps on the rock allowed us to flow water through the joint one-dimensionally from one end to the other. We then measured the joint closure and the rate of water flow through the joint as a function of hydrostatic confining pressure from atmospheric pressure to 160 MPa (1600 atm, or about 23 000 psi). The results are shown in Figure 2.

Over the 160-MPa pressure change, permeability changed by about six orders of magnitude, and the joint closed almost completely. Curiously, permeability vs closure showed a cubic dependence initially, then a rapid drop at a pressure near 30 MPa, then a new cubic dependence up to 160 MPa.

Can this behavior be understood in terms of the statistical information about the joint? It turns out that for fractal surfaces, the PSD function (Figure 1b) contains sufficient information to enable us to determine the variances of three key characteristics of the surface: the elevations, slopes, and curvatures. A fourth parameter, which we call the mating scale, is the wavelength below which the opposite sides of the joint cease being correlated to one another. It corresponds to the spatial frequency in Figure 1b where the PSD of the composite topography is twice that of a single surface. Parameters closely related to these four (the three variances and the mating scale) appear repeatedly in fracture-specific models that have been developed. We see a strong possibility that in the near future we will be able to find and verify a model in which *only* these parameters appear. This model will set the stage for predicting the hydrology of a joint system whose physical shape and arrangement are known and may lead to further simplifications as we learn more about the general features of joint systems. ■

Figure 1. (a) Digitized image of one surface of our man-made joint in Westerly granite. A 13- × 65-mm area of the 135- × 135-mm joint is shown. (b) Power spectral densities for the single surface shown in (a) and for the composite that is the joint itself, formed by carefully aligning the digital images of opposite sides of the joint and then subtracting them.

Figure 2. (a) Elasticity and (b) hydraulic permeability of the joint shown in Figure 1. Permeability (k) is normalized by the permeability at 1 atm pressure (k_0). The curves are two different forms of the cube law.

Frictional Forces and Wear on Silicon-Based Microdynamic Structures

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We are studying frictional forces and wear in microdynamic structures made with silicon-based microfabrication techniques.

Small turbines, electrostatic motors, resonant structures, and microgrippers are examples of micromachinery now built with integrated circuit technology. Future micromachinery may include high-performance sensors and actuators for the automotive, chemical, biological, and medical industries and components for miniature scientific instruments. The functional parts of these devices have micrometer and submicrometer dimensions. Preliminary analyses indicate that as these functional parts become smaller, the frictional forces

between their surfaces dominate their performance (Pisano, 1989). Our goals are to understand how forces are transmitted between two surfaces in relative motion and how fast friction wears out micromachinery.

During FY90, we designed and built macrosample and microsample structures from the four materials most used for micromachinery: single-crystal silicon, polycrystalline silicon, silicon nitride, and silicon dioxide. We then performed static experiments to determine the forces needed to initiate movement.

For the inclined-plane friction experiment (Rabinowicz, 1965), we used macrosamples of sliders and ramps such as those shown in Figure 1. The ramps were made from 7.6-cm-diam disks. The sliders, which weighed only 44 mg, were made from square sections of silicon, 0.7 cm on a side and 0.038 cm thick. We tested these structures to determine what ramp angles started the sliders moving. If friction was very high, we needed to tilt the ramps to a greater angle before the sliders moved. The coefficient of friction was calculated by dividing the force on the slider parallel to the ramp surface by the force normal to the surface.

We used three types of sliders to simulate micromachinery operations. Each slider was coated with one of the four fabrication materials. The first slider was flat. The second had three large pedestals etched in its surface. Each pedestal was 50 μm high with a diameter of approximately 1 mm. The third slider also

had three pedestals 50 μm high, but the pedestal diameters were only 0.25 mm. We used the pedestals to control the area of contact between the slider and ramp. With the flat slider, the true area of contact was unknown, and the flat surfaces of the slider and ramp trapped a cushion of air between them, which lessened the friction.

We performed our experiments in a Class 10 clean room to minimize exposure to dirt particles. Although we are still analyzing the results, we have concluded that eliminating all particulate contamination is difficult, and vibration and electrostatic charges must be considered in friction experiments with very light structures. In general, the coefficient of friction measured for the flat sliders was lower (0.2 to 0.7) than the coefficient of friction for the samples with the pedestals (0.5 to 3.0). The flat samples floated on an air cushion when first placed on the ramp. The pedestal sliders often had coefficients greater than unity and did not float on the ramp. In some cases, they moved a short distance along the ramp after it had been elevated, then hooked and adhered to it.

Our microfriction experiments are similar to recent experiments by Lim et al. (1990). Using the sacrificial-layer process shown in Figure 2, we fabricated a polysilicon cantilever beam with an end slider. This process allows us to anchor the beam to the substrate and measure friction in the lateral direction. Figure 3 shows a scanning electron micrograph of a typical cantilever beam with an end slider. The slider

Figure 1. Inclined-plane experiment for measuring the coefficient of static friction where the coefficient of static friction equals the tangent of angle ϕ . Experiment allows us to use the force of gravity, which can easily be calculated.

is $100 \times 100 \mu\text{m}$ and $2 \mu\text{m}$ thick. The cantilever beam is $5 \mu\text{m}$ wide and $2 \mu\text{m}$ thick. We varied the length of the cantilever beams from 200 to $1000 \mu\text{m}$. The mass of each slider was only $0.05 \mu\text{g}$, giving each a weight of $49 \mu\text{dyn}$. When we applied voltage to the cantilever beam, the slider electrostatically clamped to the substrate and made contact at two hemispherically shaped bushings.

In these microfriction experiments, we used a micromanipulator probe to deflect the slider horizontally so that the slope at the end of cantilever would be 45° . At this angle, the restoring spring force of the cantilever was 1.1 dyn (using a Young's modulus of $1.7 \times 10^{12} \text{ dyn/cm}^2$). We clamped the slider in this position by applying 50 V and removed the microprobe. Then we slowly reduced the voltage until the slider sprung back to its undeflected position, which occurred at 38 V . This voltage corresponds to an electrostatic clamping force of 5.9 dyn . The coefficient of static friction is the ratio of the tangential force to the normal force, $1.1/5.9$ equals about 0.2 . This ratio describes a polysilicon slider on a silicon-nitride surface.

During FY91, we expect to achieve a better understanding of the nature of friction forces. We will measure dynamic friction and wear to determine the force required to maintain motion, and we will continue to test microsamples using other combinations of the four structural materials as well as surface undulation. ■

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Figure 2. Sacrificial-layer process used to fabricate sliders for microfriction experiments. (a) The silicon wafer is coated with silicon nitride. (b) Then low-temperature glass is deposited onto the silicon-nitride-coated wafer for use as sacrificial layer. (c) Polysilicon cantilever beam and slider are patterned over the glass. (d) Low-temperature glass is dissolved to release cantilever beam and slider. This process creates a beam that is free and can be used to measure friction in the lateral direction.

Figure 3. Scanning electron micrograph of the polysilicon structure used in the microfriction experiments. The end slider is approximately $100 \times 100 \mu\text{m}$. The cantilever beam is $5 \mu\text{m}$ wide. Contact is made with the bottom wafer at two hemispherical bushings.

Induction-Linac-Driven, Millimeter-Wave CARM Oscillator

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We have constructed and operated a CARM oscillator driven by a 2-MeV, 1-kA electron beam generated by an induction linear accelerator. Data from initial tests indicate that about 50 MW of peak microwave power are generated at frequencies above 180 GHz.

The cyclotron autoresonant maser (CARM) is of considerable interest at LLNL because of its potential to provide high peak power at millimeter-wave microwave frequencies, which are required for plasma heating and high-resolution radar. The

CARM operates at a frequency that is Doppler upshifted by γ^2 from the cyclotron frequency for an electron rotating in an axial magnetic guide field. This is analogous to a free-electron laser (FEL) where the cyclotron wavelength plays the role of the wiggler period in determining the frequency. However, at lower electron energies (~ 2 MeV), the CARM can achieve much higher frequencies (smaller wavelengths) than the FEL because the high axial fields of conventional superconducting magnets result in small cyclotron wavelengths.

In collaboration with the University of California, Los Angeles, we constructed and tested a CARM oscillator that is designed to operate at 250 GHz (Caplan et al.,

1990). In FY90, we concluded a 3-y program to study the CARM theoretically and experimentally. We performed the experiment using the induction linear accelerator (linac) at the LLNL Accelerator Research Center. This accelerator consisted of a Pierce-type, 1.2-MeV injector followed by ten postacceleration cells that produced a 2-MeV, 1-kA, 30-ns solid electron beam pulsed at 1 Hz.

Using an axial guide field of 10 000 G, we focused this beam into a 12-mm pipe. Then we superimposed a transverse rotating magnetic field of 10 to 40 G onto the guide field using a bifilar wire. We wound the wire helically to impart rotational energy to the electrons. Finally, we compressed the beam into a 7-mm-diam circuit using guide fields of

Figure 1.
(a) Experimental setup for the induction-linac-driven CARM oscillator, showing the accelerator, transition magnets, superconducting magnet, and beam dump. (b) Microwave diffraction tank and waveguide diagnostics.

25 to 30 kG that were produced using a superconducting magnet.

After the beam passed through the circuit interaction region (40 cm), a transverse magnet expanded the beam and dumped it into the waveguide wall. The microwaves continued on into a 1.8-m absorbing diffraction tank. There, various calibrated waveguide horns measured power and frequency. Figure 1 shows the experimental setup, including the end of the accelerator (last six cells), transition coils, superconducting magnet, and beam dump. The diffraction tank is located in a tunnel section following the jump (Figure 1b). The microwave circuit was a Twystron configuration consisting of one 18-cm Bragg reflector section followed by a straight pipe amplification section. The Bragg reflector, which acts like an internal cavity oscillator, excites a transverse-electric 11 (TE_{11}) mode that is ten times above the cutoff at 250 GHz.

We designed the cavity with an external cavity quality factor (Q) of 5000 to ensure that the threshold for exciting oscillation was about 200 A. Thus, cavity fill times are limited to 20 to 30 ns. The desired TE_{11} mode is generated within the Bragg structure, which consists of 300 sinusoidal ripples spaced 0.6 mm apart and having 45- μ m amplitude.

Operationally, the experiment consisted of focusing the electron beam through the accelerator, wiggler, and CARM circuit, then optimizing the output power by adjusting cathode flux, wiggler current, and circuit axial field. We measured the electron-beam current using "beam bugs" and B-dot probes located at the end of the injector, the end of the accelerator, the entrance to the wiggler, and before the beam dump. Of the 1100 A emitted from the injector, we were able to transmit 700 A through the 7-mm CARM

circuit. We intercepted the current at the transition region from the accelerator (500-G axial field) to the wiggler section (10 000-G axial field). Then we measured microwaves using calibrated solid-state detectors in the range of 240 to 260 GHz and at the Ka band (25 to 48 GHz). A small waveguide horn inserted at the end of the diffraction tank sampled the far field of the high-frequency TE_{11} mode. A 2.5-cm iris in front of the horn filtered out other modes except those of the TE_{11} type.

We transmitted power through a WR4 waveguide and a 180-GHz filter and then to the detector. To monitor Ka-band power, we inserted another horn with a separate WR28 waveguide run on the side of the tank. Then we measured the frequency using a heterodyne system consisting of a 60- to 90-GHz, tunable backward wave oscillator (BWO), a mixer, and a 1-GHz bandwidth intermediate field (IF) amplifier. The high-frequency output of the CARM mixed with the third harmonic of the BWO to produce IF signals up to 1 GHz.

Both gyrotron-type modes near cutoff (Ka band) and high-frequency CARM operation occurred simultaneously. However, we minimized or

suppressed the low-frequency power by increasing cathode flux and lowering the circuit axial field (moving the low-frequency beam waveguide intersection away from cutoff). Figures 2a and b illustrate the detected power signals in both the low- and high-frequency channels and how they were calibrated when the best high-frequency result was achieved (above 180 GHz). We detected several frequencies in the range of 180 to 270 GHz. At this time, we can only be certain that the CARM produced approximately 50 MW of microwave power above 180 GHz and 3.5 MW at the Ka band. That power is the highest achieved by any microwave device in this frequency range.

Further testing using programmatic funds is now in progress to more clearly identify the exact operating frequency and optimize the output power. ■

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Figure 2. (a) Ka-band output power with a frequency range of 25 to 48 GHz. Detector sensitivity = 0.035 MW/mV, and actual output power = 3.5 MW. (b) High-frequency output power with a frequency range of 180 to 270 GHz. Detector sensitivity = 24.7 MW/mV, and actual output power = 53 MW.

Experimental Facility to Produce Dense Plasmas with X Rays

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In our work aimed at producing dense plasmas by irradiating solid targets with x rays, we have developed and demonstrated a facility that can generate broadband x rays and focus them to flux levels of about 10^{14} W/cm². We have also designed a Schwarzschild microscope to focus x rays for use in nonlinear optics experiments.

In conventional laser-matter interaction experiments, x-ray emission is primarily from the hot, low-density coronal plasma. This makes it difficult to study the higher-density, cooler regions with x-ray spectroscopy. To alleviate this problem, we have developed a facility capable of irradiating solid targets with x rays. By using x rays, we can heat thin foils uniformly throughout and generate plasmas at near solid densities, thus avoiding the steep density and temperature gradients that arise with laser-light absorption in underdense plasmas (Da Silva et al., 1989).

Figure 1a illustrates our experimental setup as it operates now at the Nova two-beam laser facility. In essence, we collect x rays produced in a laser-plasma interaction and focus them onto a remote but easily diagnosable "sample." By irradiating a gold target with one Nova laser beam (to generate a plasma that emits broadband x rays) and using a combination of a 100-nm-thick aluminum light shield and a nickel-coated ellipsoidal mirror operated at a grazing angle of 5 deg (to focus the x rays), we can produce

an x-ray beam at the sample with a spectrum centered at about 500 to 600 eV and with a spectral width of about 400 eV. We can change the photon energy spectrum by using different x-ray-absorption filters or by recoating the ellipsoidal mirror with a different material. Our current mirror is designed to produce a 100- μ m-diam focus.

Irradiating a gold target with 4 kJ of Nova laser light on a focal spot size of 100 μ m, as we are doing in our experiments, should produce an x-ray flux of about 5 kJ/cm² at the sample. To date, we have obtained only about 1 kJ/cm² in a 100- μ m spot; we think the shortfall is due to a degraded optical surface on the mirror and to operation at less than optimum irradiance for conversion into x rays. Optimizing the mirror reflectivity and using the Nova 10-beam laser should certainly allow us to reach our final design goal of 10 kJ/cm² of x-ray flux at the sample.

An important application of this facility is to perform absorption spectroscopy on plasmas that are only a little less dense than the sample material that was irradiated to produce them. In this case, the focused x rays can be used both to heat the sample and to penetrate it for diagnostic observations. A grating spectrometer placed behind the sample records the transmitted x rays with a resolving power of about 1000. In Figure 1b, we show the spectrum obtained with a 200-nm-thick carbon target under irradiation conditions sufficient to vaporize it. The carbon absorption edge is clearly observed. Absorption spectra of ionized plasmas will be possible in the future with this technique when the x-ray irradiance is increased.

To achieve higher fluxes, we studied various approaches for focusing an x-ray laser.

We ultimately chose a Schwarzschild microscope. Our initial designs considered a microscope of 100 $\times$ demagnification. However, detailed ray-tracing indicated that current multilayer-deposition techniques preclude such high-demagnification systems because of the wavelength mismatch between the primary and secondary mirrors. That is, the bandpass of each mirror varies across the surface because of the change in multilayer spacing, an effect that is enhanced for mirrors having a small radius of curvature such as used in high-demagnification systems.

As a result, we have designed 40 $\times$ and 20 $\times$ demagnification systems that should have a throughput of 25% at 15.5 nm, which is the wavelength of the yttrium x-ray laser (MacGowan et al., 1988). This laser has an output energy of about 1 mJ in a 200-ps full-width-at-half-maximum pulse. By focusing the pulse with the Schwarzschild microscope, we can achieve x-ray fluxes of about 10 kJ/cm² ($\sim 10^{14}$ W/cm²) in a 2- μ m focal spot.

This system produces x rays intense enough not only to irradiate targets and generate high-density plasmas but also to study nonlinear optical phenomena such as harmonic generation and four-wave mixing. Figure 2 illustrates the latter concept, in which an optical laser beam and an x-ray laser beam penetrate a plasma together. The optical laser beam is used to "preform" the plasma, i.e., form the plasma just before the x-ray laser beam is generated, and then to mix with the x-ray

laser beam in penetrating the plasma. Numerous experiments have demonstrated multiphoton processes in the near ultraviolet by focusing optical lasers through neutral gases (Reintjes, 1984).

To extend these experiments to the soft-x-ray region, we need a plasma rather than a neutral gas, both to reduce absorption and to allow for resonant enhancement of the nonlinear susceptibility (Muendel and Hagelstein, 1990). The isotropy of the plasma forces us to consider only odd-order processes, of which general four-wave mixing,

$$\omega_1 \pm \omega_2 \pm \omega_3 = \omega_4, \quad (1)$$

is the lowest order. For our applications with one x-ray laser beam (ω_1) and the possibility of an optical beam (ω_2), Equation (1) reduces to the cases of third-harmonic generation ($3\omega_1 = \omega_4$) when ω_2 is absent and to sum/difference frequency conversion ($2\omega_1 \pm \omega_2 = \omega_4$) when ω_2 is present. The prospect of mixing an x-ray laser with an optical laser is attractive because it offers the possibility of producing an x-ray laser that is tunable over a range of 1 to 2 eV. This greatly increases the possible applications of x-ray lasers. ■

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Figure 1. (a) Experimental setup for focusing broadband x rays. (b) Spectrum of x rays transmitted through 200-nm-thick carbon foil.

Figure 2. Experimental setup for x-ray laser focusing and four-wave mixing. The optical laser beam is used to produce plasma and to interact with the focused x-ray laser beam.

Harmonic Generation in Ferroelectric Liquid Crystals and Chiral Polymers

S. P. Velsko and L. M. Hair

We have investigated the optical properties of ferroelectric liquid crystals and chiral rigid-rod polymers in evaluating possible routes to inexpensive, designer materials for optical harmonic generation.

Nonlinear crystals have the ability to convert light passed through them into light of a higher (harmonic) frequency. They are widely used for frequency conversion of

near-infrared laser beams into coherent visible light. However, their high cost and limited availability remove them from consideration for many potential applications. In looking for less constraining materials to use for frequency conversion, we have examined the optical properties of chiral mesophases—specifically, ferroelectric liquid crystals (FLCs) and chiral rigid-rod polymers—to evaluate whether they could be developed into inexpensive, “engineered” frequency-conversion materials. The availability of such

materials could significantly reduce the cost of potential applications such as “throwaway” rocket-borne lidar systems (for strategic defense) and laboratory projects requiring large-aperture frequency conversion devices (for example, laser-driven inertial confinement fusion).

Ferroelectric liquid crystals and chiral polymers are noncrystalline ordered materials that can generate optical harmonics. In these materials, optical nonlinearity is guaranteed by the combination of chirality and liquid crystalline order.

Figure 1. Stereonet showing calculated phase-matching properties of prototype FLC harmonic generators and the structure of a typical FLC phase (from Liu et al., 1990). On the stereonet, each circle marked with letters is a locus of orientations for which phase matching is possible. Letters mark local maxima in the nonlinear coefficients; zeros mark orientations where nonlinear coefficients vanish. Orange dashed line on stereonet corresponds to the plane of smectic layers and cell walls.

Because the ordering is spontaneous at room temperature (a thermodynamically favorable feature), poling of the materials in very high electric fields is unnecessary, a major advantage over poled polymers, which are being studied elsewhere for nonlinear optics.

Chiral mesophases appear to offer several advantages over crystalline materials. For example, they could be manufactured by lamination or casting processes in a wide range of sizes, whereas for crystalline frequency converters, the size attainable and the cost depend on the vicissitudes of crystal growth technology and delicate fabrication procedures. Chiral mesophases also have large optical birefringence, which permits high-efficiency phase-matched operation. Most important, the birefringence, and hence the wavelength of highest conversion efficiency, can be "tuned" at will by changes in composition and processing.

Our goal was to understand the relationship between these substances' molecular and mesophase structures and their optical properties. This knowledge is a prerequisite for assessing the potential of the materials for producing useful harmonic generators. To this end, we developed a model that relates the structure of the FLC's molecules and their arrangement in space to the optical properties. Our model predicted that large nonlinear coefficients could be obtained with specific molecular structures. Figure 1 is a stereonet showing the results from our calculation of the

phase-matching properties of prototype FLC harmonic generators. Also shown is the structure of a typical FLC phase (adapted from Liu et al., 1990). In Figure 1, the z axis lies parallel to the molecular director, and the y axis is the polar axis of the FLC in the aligned state. These predictions have been verified by independent experiments at the University of Colorado, Boulder.

We have also demonstrated that variants of the techniques we previously developed for characterizing crystalline harmonic generators could be used to determine the nonlinear optical properties of FLCs. Figure 2 shows phase-matching data obtained in our laboratory for a prototype FLC cell containing a commercially available FLC compound. Because the investigators at Boulder have strong expertise in the area of FLC synthesis and cell fabrication, we are now collaborating with them and an allied company, Displaytech, to pursue funding through the Small Business Independent Research program for further development of these materials.

In addition to our work on FLCs, we have examined the harmonic-generating properties of chiral polyisocyanate polymers, which belong to the category of chiral rigid-rod polymers. Samples of these materials were obtained under contract with David Sohn of the University of California, Berkeley. Although small second-harmonic-generation (SHG) signals have been observed from cast solid films, preliminary experiments

with electric-field-induced harmonic generation imply that the structure of these polymers is too tangled to permit the formation of the large ordered domains needed for useful nonlinear response. Further progress in this area will depend on the availability of synthetic methods for controlling chain length and stiffness. ■

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Figure 2. We have observed second-harmonic generation in an FLC cell as it is rotated about the (polar) y axis. The face normal of the cell corresponds to 0 deg, and the large peak in intensity that occurs close to this direction is characteristic of phase-matched harmonic generation.

Microthin Lenses for the Correction of Aphakia, Myopia, and Hyperopia

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We have fabricated our first microthin lenses and demonstrated their imaging properties. These lenses make possible a radical new procedure to correct aphakia, myopia, and hyperopia.

In current intraocular lens (IOL) implant procedures, a cataract is removed through a large (5- to 11-mm) incision in the cornea and replaced by a thick (0.5-mm) plastic lens. Although a human cataract can be removed through a smaller (1-mm) sutureless incision by

ultrasonic emulsification, a simple and inexpensive procedure, the thick lens cannot be inserted through this incision. We have developed an ultraflexible microthin lens (MTL) that can be implanted through this small incision or placed directly on the cornea without surgery. Not only will this procedure be less costly (savings are estimated in the billions of dollars in the U.S. alone), but it also can restore sight to millions of Third World patients who go untreated because of limited finances and unavailable hospital facilities.

Our original project goal was to study and develop a new type of IOL. Our MTL, a diffractive lens,

can either be an achromatic Fresnel phase plate or a blazed Fresnel phase plate etched into a thin (5- to 100- μ m) plastic membrane. The achromatic lens has a nearly constant focal length over a large wavelength, including the range of human imaging.

We have developed and tested two achromatic MTLs:

- A composite lens fabricated from sectors of different lenses, each sector designed and fabricated to image a particular narrow band of radiation.
- A stack of minus or notched filters, each patterned with a diffractive lens to focus radiation within its bandwidth.

MTL implant procedures mark the first time that single diffractive lenses have been used to produce "white-light" images. Traditional diffractive lenses are chromatic and can image only one color. White light is composed of all colors. The achromatic MTLs work with all colors.

Figure 1 shows a 5-mm-thick MTL mounted on a conventional IOL haptic. In white-light imaging experiments, the MTL showed better spatial resolution than conventional diffractive optics, and under monochromatic illumination, the MTL displayed nearly diffraction-limited resolution, outperforming conventional IOLs under similar illumination conditions. ■

Figure 1. A 5- μ m-diam MTL mounted on a conventional IOL haptic. Dime is shown for comparison.

Intense Lower-Hybrid Wave Penetration

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In FY89, we began a theoretical study of intense pulses to facilitate penetration of lower-hybrid (LH) waves into the core of reactor-grade tokamak plasmas. Such waves can efficiently create the currents needed to continuously operate a tokamak and heat it. Intense pulses can be used to saturate the Landau damping process, which excludes conventional LH waves from reactor-temperature plasmas. Focusing on short pulses (20 to 50 ns) and peak-power levels of 10 to 100 GW, we developed a theory of the collisionless absorption process, including the quantification of a nonlinear plateau in the electron distribution function, incorporated the results into ray-tracing calculations, and assessed the parametric stability of the waves. Adequate penetration required long launching structures that are feasible but unattractive.

In FY90, we refined the calculations to incorporate an arbitrary repetition rate, staggered pulses, a fuller relativistic treatment, and a more quantitative pump-depletion criterion for parametric instabilities. We also explored the influence of quasi-static potentials created by

space-charge effects. Our conclusions changed quantitatively but not qualitatively.

Thus, we explored alternative scenarios with pulses long enough that collisional dissipation is important; then absorption is insensitive to launcher shape. We developed a theory of absorption that incorporates transient collisional effects and compared it with Fokker-Planck code calculations. The results agree well, indicating that the pulse-averaged absorption coefficient decreases with pulse duration but levels off for durations longer than a small fraction of the collisional relaxation time of the nonlinear plateau. For plasma parameters proposed for the International Thermonuclear Experimental Reactor (ITER), the transition pulse duration is about 50 μ s; for LLNL's

Microwave Tokamak Experiment (MTX), it is about 20 μ s. Ray-tracing studies with the new absorption coefficient show that 9-GW, 80- μ s pulses launched from an 18-m² area in ITER penetrate beyond the plasma center (Figure 1). A development program is required to provide the radio-frequency sources for these relatively long durations; the gyrokystron is a promising candidate.

We have outlined an experimental test that could be done on the MTX. Power from existing 2-MW, 4.6-GHz sources launched with a parallel index of refraction of 5 in 20- μ s pulses from 640 cm² of launcher area should penetrate to the center, whereas pulses of much shorter duration or power, or pulses from a smaller launcher, should not. ■

Figure 1. Comparison of ray penetration in ITER for (a) 9-GW peak-power pulses launched from 18 m² of launcher and (b) 100-MW continuously applied power launched from 2 m² of launcher. Tic marks denote absorption of 10% of the incident power.

Chemical and Optical Characterization of Environmental Records in Coral Skeletons

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Coral is an environmental biosensor that can assist us in understanding global climate changes. We are using a time-resolved, laser-induced spectroscopic technique to demonstrate that the fluorescence emission intensity of coral is correlated with subannual rainfall.

Research in the 1970s resulted in a dramatic expansion of the use of corals as environmental biosensors and recorders. Following the discovery of annual and subannual growth records in coral skeleton, much work was done on evaluating environmental controls over growth patterns and the chemical and isotopic composition of the calcium carbonate exoskeleton of coral colonies.

A potentially important but, as yet, poorly exploited discovery is that patterns of variable fluorescence in the skeletal growth records of corals from the Great Barrier Reef are empirically correlated with runoff from the major drainage basins of northeastern Australia (Isdale, 1984). These patterns are also correlated with historical records of major regional fluctuations, such as

the El Niño southern oscillation cycle. Boto and Isdale (1985) showed that the fluorescence is associated with terrestrial fulvic acids incorporated into the coral skeleton. The obvious potential for climate reconstruction and studies of human impacts on marine and terrestrial ecosystems has stimulated a variety of surveys and trial studies (Scoffin et al., 1989). However, few additional contributions have been made to the scientific literature.

Several uncertainties have prevented the direct use of fluorescence spectroscopy for environmental assessment and climate characterization. These uncertainties stem largely from the highly empirical approach to the relations among environmental, physical, and chemical characteristics of a sample and the fluorescence signal. Boto and Isdale (1985) provide the only chemical data available, and optical characterization of the signals and samples has been limited to simple optimization of the fluorescence signal-to-noise ratio. Our goal is to resolve certain questions about the nature and utility of skeletal fluorescence records and to extend the range of applications.

LLNL has major ongoing efforts in climate modeling, studies on atmospheric impacts of carbon dioxide, and development of coupled

ocean-atmosphere models. A more far-reaching interest is the closely related areas of climate-change impacts and studies of model confirmation. Our research focuses on obtaining proxy climate-history records for oceanic areas that are inadequately instrumented. Such efforts could provide the basis for an analytical center of excellence at LLNL to exploit biorecorders as integrating environmental-monitoring systems that do not require installation, maintenance, or advance experimental design.

In FY90, we tested, adapted, and validated our analytical methods. We modified our laser-induced fluorescence spectrometer to allow reproducible positioning of coral cores in the optical path. Such modifications facilitate translation of the coral core along its growth axis. We established a collaboration with Michael Moore, a graduate student at the University of California, Berkeley, who provided a series of coral cores from Indonesia and Costa Rica.

We have demonstrated the feasibility of using time-resolved, laser-induced fluorescence to study changes in the nature and amount of humic material incorporated into the exoskeleton of coral as a function of climate changes. We measured the fluorescence intensity over a 20-y period of coral bands for coral cores

from Padang Bai, Bali, and from Cano Island, Costa Rica. Figure 1 shows that variations in the humic-acid emission intensity correlate well with the known rainfall data; thus, we have demonstrated the utility of the method for Bali data.

Using laser-induced fluorescence techniques, we have begun to elucidate the nature and structure of humic materials through spectral and lifetime studies. The temporal spectrum contains two components. The short-lived fluorescence signal has a lifetime shorter than we can measure with our current system (<10 ns). We also found a much weaker, longer-lived signal, which is most likely due to phosphorescence, that has not been reported previously in the literature. As shown in Figure 2, we also observed energy variations in the emission spectrum as a function of geographic location, a finding that is also previously unreported.

We attempted to dissolve coral samples and to study their solution emission spectra. The resulting spectra were qualitatively what we expected. Using chromatographic procedures from the literature, however, we were unable to isolate humic acid for nuclear magnetic resonance spectroscopic studies. Thus, we require further development of separation techniques for future studies.

We have collected coral samples from Indonesia to evaluate the rainfall history of that area for the last 500 to 700 y. The sensitivity of Indonesian rainfall to large-scale

Pacific Ocean and atmosphere circulation will enable us to reconstruct Pacific weather patterns during the global cooling and warming that occurred during the Little Ice Age (1550–1850). Soil and water samples from rainfall basins in the area are also being collected. Spectral information from these samples will assist us in linking information stored in coral skeletons to the terrestrial and drainage-basin rainfall-runoff sources. ■

Figure 1. (a) Precipitation data and (b) time-resolved, laser-induced fluorescence of coral bands for cores from Padang Bai, Bali, over a 20-y period. The variations in humic-acid emission intensity correlate well with known rainfall data, thus demonstrating the utility of this method.

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Figure 2. Energy variations in the emission spectrum of humic materials from two geographic locations: Papua, New Guinea, and Padang Bai, Bali. Such variations may provide a fingerprint for a given region.

A Library of Abundance-Calibration Standards for the Ion Microscope

D. L. Phinney, D. K. Smith, J. L. Wong, and K. D. McKeegan

We have built a library of standards for quantitative determinations of trace elements at interfaces with the ion microscope. Using these standards, one can determine the abundance of the elements in situ in a sample at a range of depths in the billion range for all elements of the periodic table in areas as small as a few square micrometers.

The ion microscope is an ultra-sensitive sputtering-source mass spectrometer with microscopic (about $1 \times 1\text{-}\mu\text{m}$) imaging capability for isotopes of all elements. When this instrument was introduced in the 1970s, many expected it to replace the electron microprobe for rapid, in situ quantitative chemical characterization of materials on a microscale. However, quantification for the ion microscope turns out to be a sample-by-sample process. This process entails the selection of a suite of standards similar to the sample's chemical composition; certification of the abundances of the elements of interest, usually by ion implantation; and cutting, polishing, and mounting the standards for the ion-microscope sample mechanism. Not only does this process lack the virtue of rapid turnaround, typically requiring several months per sample, but it also is costly, running about \$25,000/sample.

Our strategy for both speeding turnaround and cutting cost is as follows. We have selected 200 diverse, chemically well-characterized materials and have prepared standards batch-style for certification by ion implantation and for ion-microscope analysis. Through economy of scale, the cost per implanted sample with this approach is about \$20. With 200 certified standards on the shelf, we can start quantitative work on most microanalytical characterization problems within minutes after samples arrive in our laboratory.

The standards comprise 11 general categories: solid elements (44 items), single-crystal oxides (12), semiconductor materials (8), gem-quality minerals (42), optical materials (12), steels and ferrous alloys (15), nonferrous alloys (20), ceramics and sintered materials (13), glasses (12), crystalline fluorides (13), and miscellaneous (9).

Fifteen sets, as nearly identical as possible, of 25 ion-microscope mounts were made from polished pieces of these materials. Fourteen sets are reserved for ion implantation; the fifteenth is a blank.

Funds were available to make ion implants in eight sets. One set was implanted with ^2H . The other seven received two implants apiece: (^6Li , ^{67}Zn), (^9Be , $^{204,206,207,208}\text{Pb}$), (^{10}B , ^{19}F), (^{18}O , ^{109}Ag), (^{26}Mg , ^{93}Nb), (^{27}Al , ^{127}I), and (^{232}Th , ^{238}U).

Twelve other implants will be made later: (^{13}C , ^{71}Ga), (^{15}N , ^{76}Ge), (^{37}Cl , ^{133}Cs), (^{41}K , ^{134}Ba), (^{54}Fe , ^{142}Nd), and (^{65}Cu , ^{197}Au).

The ^{232}Th and ^{238}U implants are unprecedented in materials science,

having been achieved with 600-kV, monoenergetic ion beams.

We have two primary goals for this library of standards:

- *Rapid response to trace-element characterization problems.* The library is already bearing fruit in this regard. For example, we have determined the abundance of contaminant iron in Nova-laser phosphate glass; have traced actinide depth distributions in tuff rock from Yucca Mountain, Nevada; and are cross-calibrating uranium, thorium, and lead sensitivities in minerals with ion microscopes in other laboratories.
- *Sputtering-yield systematics.* We are analyzing all of the materials in our library for all implanted elements with a view to developing sputtering-yield systematics. These systematics will allow us to extrapolate sensitivity factors to materials and trace elements not in our library.

A byproduct of this project is the host of significant applications latent in the library as it now stands. These include trace-element partitioning in minerals, grain-boundary chemistry in metals, and geochemical problems related to the early history of the solar system. We will seek collaborators for such applications and do the work as time permits. ■

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p-n Bremsstrahlung at Medium Energies

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Cherenkov and ΔE e^+ , e^- identification elements; and two 40×40 -cm Nal calorimeters. Signals from all elements come via coupled 5-cm photomultipliers. All components are in-hand. Construction of light pipes and phototube contacts is proceeding for all of the elements except the Nal calorimeters, which are complete and have been tested. Testing of other components will begin in

FY91. Two identical detectors will be used in the experiment.

Computer modeling of the resolution of different detector configurations has begun. The detectors will be calibrated with photons of known energy at the University of Saskatchewan electron accelerator facility. Experimental n-p- γ measurements are expected to begin with the spring 1991 LAMPF running cycle. ■

We are a major part of an international collaboration whose goal is to measure the gamma-ray spectrum resulting from neutron-proton (n-p) scattering. Successful measurement of the n-p- γ spectrum has eluded experimenters for 40 y. It is a valuable probe of the n-p interaction because it is sensitive to the meson exchange currents between the neutron and proton.

Our experiment will use the white neutron source from the Weapons Neutron Reactor of the Los Alamos Meson Physics Facility (WNR/LAMPF), with a cryogenic liquid- H_2 target. We have designed and constructed a cryogenic target cell, and beam time has been allocated by the WNR Program Advisory Committee.

Design of the new detection system has been completed. This system consists of plastic charged-particle rejection elements; BaF_2 active gamma-ray converters;

Figure 1. (a) Configuration of the p-n bremsstrahlung experiment. (b) The elements of our new gamma-ray detector.

Search for Resonant States in Positron-Electron Scattering Using a Positron Gas Target

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In our search for resonant states in positron-electron scattering experiments, we are building a pure-positron plasma trap to form a cold, positron gas target.

Correlated narrow-peak structures in positron-electron spectra discovered in heavy nuclear collision experiments (Cowan et al., 1986) may reflect the existence of two-body decays of previously undetected neutral particle-like objects having masses of 1 to 2 MeV/ c^2 (Cowan and Greenberg, 1987; Salapura et al., 1990). We have designed an experiment to test the neutral particle hypothesis of the anomalous positron-electron peaks detected in these collisions. We will search for their resonant formation by scattering monoenergetic electrons incident on a gas of cold positrons confined in a Malmberg-type magnetic trap (Cowan et al., 1990).

Using a positron gas target for the experiment provides 100 times more sensitivity than previous positron-beam, thin-foil scattering experiments, which were limited by the momentum of the atomic

electrons in the solid targets (Tsertos et al., 1989; Judge et al., 1990). We will now be able to explore the neutral particle lifetime range of 10^{-13} to 10^{-8} s, a range that other methods have found impossible to probe directly.

To form a sufficiently dense and cold positron plasma for use as a scattering target, we will efficiently capture and magnetically confine the positrons in a Malmberg-type trap. As shown in Figure 1, the LLNL 100-MeV linear accelerator will drive positrons from the low-energy positron source into the solenoid's 60-kG magnetic field where they will be trapped by a time-of-flight technique. As each positron pulse arrives from the source, we will temporarily reduce the injection-end electrode voltage to allow the positrons to enter the trap and then increase it again to capture them. Confined in the trap, the positrons will form a plasma column that spins as a rigid rod about its own symmetry axis as a result of $E \times B$ forces from the radial space-charge electric field. Azimuthal nonuniformities in the trapping fields appear as a radiofrequency (rf) source to the rotating plasma column and exert a torque that expands and heats the plasma. To provide uniform trapping fields, we fabricated a structure in which the electrodes are mounted in a precision ceramic V-block to provide a mechanical concentricity of 2.5 μ m over a 60-cm length. Superconducting correction coils align the plasma with the magnetic field (B) axis. We anticipate an azimuthal uniformity

of $\Delta B/B \ll 10^{-4}$, with confinement times of up to 300 s. Table 1 lists the key operating parameters for the positron plasma trap.

Once a sufficiently dense positron plasma is formed (about 10^{10} e $^+$ /cm 3), an electron beam of about 2.5 MeV will be injected along the solenoid axis from a 3-MV electron accelerator. Coulombic heating of the positrons by the electron beam will be offset by cyclotron radiation cooling in the solenoid's strong magnetic field. We expect an equilibrium temperature of 100 K and a center-of-mass positron-electron energy resolution of about 200 eV (compared with 7.8 keV in solid beryllium target experiments). The temperature and radial density distribution of the pure-positron plasma will be measured using a two-dimensional imaging system (microchannel plate, phosphor, and charge-coupled device camera) developed for this experiment at the University of California, Berkeley.

Scattered positrons and electrons will spiral out of the trap into lithium-drifted silicon detectors where their measured energies, their identification by orbital helicity, and the coincident detection of the positron annihilation radiation in sodium iodide detectors will allow reconstruction of the collision kinematics. To search for resonant states, we will precisely measure the positron-electron scattering cross section as a function of the beam energy. A bias voltage on the trap will continuously scan and adjust the beam energy and thus minimize systematic uncertainties.

Table 1. Key operating parameters for positron-plasma trap.

Confinement time (s)	300
Fill time (s)	25
Cyclotron cooling time (s)	0.11
Beam heating rate (eV s $^{-1}$)	0.08
Equilibrium e $^+$ temperature (eV)	0.008
e $^+$ e $^-$ scattering rate (s $^{-1}$)	0.5

This technique's sensitivity to neutral resonant states can be compared with that of previous positron-beam, thin-foil scattering experiments. Our improved energy resolution and positron-electron identification will provide a hundredfold extension of the directly explored region from lifetimes of 3.5×10^{-13} to 10^{-11} s. Part of this range was recently explored indirectly using a recoil shadow technique and a solid beryllium target and may be excluded if the neutral object is assumed to penetrate the solid target. However, many recent theoretical models suggest that this is unlikely. A comparable recoil-shadow technique with our positron-gas target, which is 10^6 times less dense than the solid beryllium targets, provides a model-independent limit on the existence of such states over the lifetime range of about 5×10^{-12} to 10^{-8} s. Therefore, this experiment will constitute a definitive search for new neutral particle-like objects throughout the lifetime range of 10^{-13} to 10^{-8} s,

which other methods cannot reliably and unambiguously probe.

During FY90, we completed the procurement and assembly of most of the components for the positron trap including the precision containment electrode structure, Malmberg-trap computer control system, non-magnetic ultrahigh vacuum system, and plasma diagnostics detectors. We also completed preliminary layout of the trap and electron beam in the experimental area. The positron-electron system was tested in another ER&D project (Cowan et al., 1991). Final assembly and testing of the trap are planned for FY91, following delivery and installation of the solenoid and accelerator. Positron-electron scattering experiments should begin shortly thereafter. ■

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Figure 1. Schematic setup of the LLNL positron-electron scattering experiment. Positrons are injected into the 60-kG magnetic field (B) where they are captured and confined in a Malmberg-type positron plasma trap to form a positron gas target. A 2.5-MeV electron beam is injected incident to the target, and the scattered positrons and electrons are transported by the magnetic field to Si(Li) and NaI detectors.

Ab Initio Calculation of Electron Energy Spectra in Very Intense Laser Fields

K. J. Schafer and K. C. Kulander

Calculations of photoionization and electron energy spectra from atoms undergoing multiphoton ionization in short-pulsed, intense laser fields provide evidence for the proposed connection between above-threshold ionization and high-order harmonic generation in atomic systems.

Current laser systems can develop field strengths comparable to or exceeding the binding energy of an electron to an atom or molecule. Experiments with such lasers have led to some unusual and potentially useful results. Two have been the focus of intense effort: (1) above-threshold ionization (ATI), whereby an electron absorbs many

more photons than the minimum needed for ionization; and (2) very-high-order optical harmonic generation (OHG), in which an electron absorbs many long-wavelength photons and emits one short-wavelength photon. Theorists propose that ATI and OHG are strongly connected (Javanainen et al., 1989), but it is very difficult to observe the two processes simultaneously in an experiment.

We have calculated ab initio ATI spectra by integrating the time-dependent Schrödinger equation for a short-pulsed, intense laser interacting with various atomic systems. At the highest intensities studied, more than one million spatial grid points were employed. This project has necessitated several advances in numerical technique, which have been incorporated in a set of computer codes developed at LLNL.

Figure 1 shows a typical spectrum of emitted electron energies. The peaks in energy above zero indicate the absorption of 1, 2, 3, ..., ATI photons. By integrating the area under each such peak, we can calculate the rate for each ATI process. In Figure 2, we plot these partial ionization rates against the number of excess absorbed photons for various laser intensities. The onset of a plateau in the rates at higher intensity is similar to the plateau behavior seen in OHG spectra. We have also found good agreement between the intensity dependence of the ATI rates and that of the corresponding OHG processes. ■

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Figure 1. Calculated photoelectron spectrum for a 532-nm laser at $2 \times 10^{13} \text{ W/cm}^2$ on hydrogen. Sharp peaks below zero energy are population remaining in excited states at end of laser pulse. Peaks above zero energy indicate absorption of 1, 2, 3, ..., ATI photons.

Figure 2. Partial rates for absorption of excess photons during multiphoton ionization of hydrogen by a 532-nm laser. These rates are proportional to the areas under peaks such as those in Figure 1. Intensities are in W/cm^2 : filled square, 1×10^{13} ; open circle, 2×10^{13} ; triangle, 4×10^{13} ; open square, 6×10^{13} ; filled circle, 1×10^{14} .

FY90-560,000

Inverse Square Law of Gravity: A View from on High

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A series of experiments done in Australian boreholes suggested that Newton's inverse square law of gravity may break down at a depth of 1 km. These experiments were confirmed at boreholes in Michigan, one in Greenland, and by several laboratory experiments that were designed to test the compositional dependence of the gravitational interaction. Our experiments refute the reported breakdown of Newtonian theory and, in fact, show that Newton's description is remarkably precise at ranges from 10 m to 2 km.

The basic principle of the experiment is discussed in most textbooks on mathematical physics. By measuring the potential energy on a closed surface, we can rigorously calculate the potential outside the surface if there are no external sources. The atmosphere is a negligible source of gravitational energy, so an accurate survey of the surface gravity field can yield a precise prediction of the gravitational field at various points on the tower.

In FY90, we completed a survey of 1000 locations on the ground within 10 km of the 465-m Bare Reactor Experiment Nevada

(BREN) tower at the Nevada Test Site. At each station, we knew the gravitational potential to within a few parts per billion (ppb) and the absolute location to within a few centimeters. We combined this information with the gravity values from 60 000 stations that were surveyed by other groups out to a distance of 600 km from the tower.

Figure 1 compares the results of our predictions to measurements on the tower. (Note that $1 \mu\text{gal}$ is $10^{-8} \text{ m} \cdot \text{s}^{-2}$, and the canonical value for gravitational acceleration is $9.8 \text{ m} \cdot \text{s}^{-2}$.) We subtracted the gravitational attraction of a theoretical model of the entire earth from both curves to accentuate the differences.

The agreement is remarkable and is never larger than 30 ppb of the total field. All measurements agree at the 1-sigma level. We are using these data to place constraints on various theoretical models that predict Yukawa-type interactions between massive bodies by the exchange of graviscalars and graviphotons.

We have shown that the Newtonian theory of gravity is an excellent description of the gravitational interaction at intermediate distances, a range that had not been tested previously. Our data will guide and constrain future theorists who want to create non-standard models of the gravitational interaction. ■

Figure 1. Tower data compared with Newtonian surface-gravity predictions. The points labeled Batman VI refer to the computer program that extrapolated the surface-gravity field (from ground measurements) up to the locations measured on the tower. Errors on the extrapolated points were always smaller than $15 \mu\text{gal}$, where $1 \mu\text{gal} = 10^{-8} \text{ m} \cdot \text{s}^{-2}$. Errors on the tower points were only $20 \mu\text{gal}$. Our tower measurements agree with the Newtonian extrapolation of the surface gravity field to within 30 ppb.

Nuclear Physics of Anomalous Pair Production in Collisions of Low-Energy Heavy Ions

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In FY90, we published results from calculations on the collision of $^{238}\text{U} + ^{238}\text{U}$, giving a theoretical explanation for the anomalous pair production in the heavy ion collision. We have applied the theory to same-same collisions between lead ions of three species and predicted similar pair production for two of them and no pair production for the third.

In FY89 (Weiss et al., 1989), we described our progress in seeking a theoretical explanation for the anomalous pair production observed

experimentally in collisions between low-energy heavy ions (Schweppe et al., 1987). We took an ab initio approach, using constrained Hartree-Fock-Bogolyubov (HFB) calculations to simulate the collision of two ^{238}U nuclei at low energies, about 5.5 MeV/nucleon. (Note that the two ^{238}U nuclei have 476 total nucleons—184 protons + 292 neutrons).

The calculations indicated that electromagnetic transitions having energies between 1.5 and 1.8 MeV could be expected in the center-of-mass system of the colliding nuclei; these transitions could be the source of the observed anomalous pair production. The calculations were also consistent with the observed laboratory energy

dependence and Z dependence of the pair production.

In FY90, we completed the calculations on the $^{238}\text{U}-^{238}\text{U}$ system (Berger et al., 1990). We have also produced computer-generated, three-dimensional pictures of the nuclear density at various stages in the $^{238}\text{U}-^{238}\text{U}$ collision (Figure 1).

We believe we have found a plausible theoretical explanation for the anomalous pair production in this collision. Briefly, the key is in the calculated behavior of the colliding ions as they approach each other. Figure 2 shows a plot of the calculated total energy of the $^{238}\text{U} + ^{238}\text{U}$ system as a function of the constrained mass quadrupole moment $\langle Q_{20} \rangle$ of the combined system and of the distance d between the centers of mass of the two nuclei when they are separated.

Two distinct curves are obtained. Along the right curve ($19.5 \text{ fm} < d < 25 \text{ fm}$), the HFB states are composed of two well-separated ^{238}U nuclei whose multipole deformations differ from their ground-state deformations because of the polarizing effect of their mutual Coulomb interaction. Along the other curve ($16 \text{ fm} < d < 21 \text{ fm}$), the HFB states describe a unique system representing two overlapping uranium nuclei. For $d < 19.5 \text{ fm}$, we cannot obtain HFB solutions in the form of two separated nuclei. For such distances, the nuclear short-range attraction dominates the mutual Coulomb repulsion of the two nuclei, and the $^{238}\text{U} + ^{238}\text{U}$ system becomes unstable against fusion.

In the fusion valley

Proton density: 95/20

102/15

115/10

At the top of the barrier between the fission and fusion valleys

Proton density: 95/12

102/10

115/5

In the fission valley

Proton density: 95/5

102/3

115/1

Figure 1. Nuclear density at various stages of the heavy ion collision and the configurations it would reach after emitting electromagnetic radiation. The proton densities listed are outside/inside the neck.

Similarly, for $d > 21$ fm, the composite system is unstable against fission. The dashed part of each curve indicates the regions where these instabilities begin to appear and HFB solutions of lower energy are obtained. Between 19.5 and 21 fm, the $^{238}\text{U} + ^{238}\text{U}$ system may exist under two configurations with different degrees of "necking" but relatively close total energies.

The coexistence of two stable configurations around 20.5 fm indicates that the interaction energy of the two ^{238}U nuclei depends not only on the distance between them but also on other degrees of freedom. One of them is clearly the necking of the overall system shape. In Figure 2, the neck is the portion of the overlapping nuclei common to both, and its thickness is represented numerically by the average number of nucleons it contains.

If we plot potential energy vs neck thickness in the region of d where two different stable configurations exist, we get curves with two energy minima (valleys) separated by a barrier whose maximum height (for $^{238}\text{U} + ^{238}\text{U}$) is about 3 MeV. This situation implies a mechanism for producing correlated electron-positron pairs, the origin of which is a fusion-like nuclear transition (Berger et al. 1990).

We next applied the technique on collisions between pairs of low-energy ions of several lead isotopes: ^{202}Pb , ^{208}Pb , and ^{214}Pb . Figure 3 shows a plot of total energy vs quadrupole moment for low-energy collisions between same-same pairs of these ions. In two plots, two distinct curves are obtained, corresponding to the uranium-uranium results. The definite transition regions in the ^{202}Pb - ^{202}Pb and ^{214}Pb - ^{214}Pb curves indicate that electromagnetic radiation (i.e., pair production) is expected. However, the curve for ^{208}Pb - ^{208}Pb shows no such transition. We interpret this as evidence of

the lack of an electromagnetic transition for this collision; hence, no pair production is expected.

In FY90, we began to calculate the electromagnetic lifetimes of our predicted transitions. This work is continuing under funding from the Physics Department.

The full implication of our research cannot be drawn until the lifetime calculations are complete. Nevertheless, we have shown that the most puzzling phenomenon in current nuclear physics may well have an exotic explanation within the realm of conventional science. Moreover, we have predicted a new

class of electromagnetic transitions in nuclear collisions, transitions that await further testing. ■

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Figure 2. Total HFB energy of the system as a function of the constrained mass quadrupole moment ($\langle Q_{20} \rangle$) of the combined system: d = distance between the centers of mass of the two nuclei when they are separated. Horizontal arrows on the right indicate total energy into the system for collisions at 5 and 5.5 MeV/nucleon.

Figure 3. Total energy vs quadrupole moment for three isotopes of lead. The orange curves show the transition region between fusion and fission configurations, which we believe is characteristic of a collision that would cause pair production. The absence of such a transition region for the black curve indicates no pair production for this collision.

In Situ Characterization of High-Temperature Superconductors

H. B. Radousky, J. C. O'Brien, and R. H. Howell

We have performed temperature-dependent Raman and positron-annihilation lifetime spectroscopy on various high-temperature superconductors.

Understanding the phase diagrams and properties of high transition temperature (T_c) oxides requires knowledge of oxygen ordering, vacancy type and ordering, and underlying crystal structure. Over the past 2 y, we have been developing the capabilities to make in situ positron-annihilation and Raman spectroscopy measurements under a wide range of conditions, including low temperatures in the superconducting state and high temperatures in various atmospheres.

The $\text{Ba}_{1-x}\text{K}_x\text{BiO}_3$ (BKBO) system ($T_c = 30$ K) is of particular interest because it is cubic, does not contain copper, and is expected to play a

major role in future device applications. We obtained high-quality single crystals of BKBO in collaboration with researchers at the University of California, Davis, and we obtained melt-textured samples in collaboration with Argonne National Laboratory. From Raman spectroscopy on the melt-textured samples in collaborative work with Sandia National Laboratories, Livermore (McCarty et al., 1989) and our recent work on the single crystal samples, we have shown that at least one optical phonon is strongly coupled to the electronic continuum. This type of electron-phonon interaction is the coupling mechanism in conventional superconductors and is apparently a major contributor to the pairing of electrons in the BKBO system.

Positron annihilation spectroscopy has been widely used to study electronic properties and defect structures in solids. Positron

lifetimes gauge the overlap of the positron wave function and average electron charge density in defect-free material. Higher electron density results in a shorter positron lifetime. When defects are present that trap or localize the positron, the associated positron lifetime is a measure of the average electron density in the vicinity of the defect site. In bulk material, the electron density distribution does not change as a function of temperature, except by thermal expansion of the lattice. For the range of temperatures of interest to us, this effect is negligible. However, in the presence of defects, the measured positron lifetime can change with temperature.

The thermal energy of the positron and the depth of the potential well determine the likelihood that the positron will be trapped long enough to annihilate with an electron in the vicinity of the defect. At high temperatures, the positron can only be trapped by defects with relatively deep potential wells. Thus, a measured positron lifetime is an average of lifetimes associated with the deeper traps. At lower temperatures, the positron spends more time in shallower wells than it does at higher temperatures; hence, the measured lifetime will have a component contributed by the electron charge distribution in shallower wells.

Figure 1 shows our data on temperature dependence of positron annihilation lifetime on two compositions of melt-textured BKBO, $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ and $\text{Ba}_{0.5}\text{K}_{0.5}\text{BiO}_3$, first taken when the samples were freshly prepared and again 1 y later. The superconducting transition temperature of these samples was characterized before each

Figure 1. Comparison of the temperature dependence of the positron lifetime for aged and freshly prepared samples of $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ and $\text{Ba}_{0.5}\text{K}_{0.5}\text{BiO}_3$.

positron lifetime experiment, and in each case, the sample superconducted for temperatures below $T_c = 30$ K. In each plot, the positron annihilation lifetime in the vicinity of the superconducting T_c of 30 K does not change, indicating no difference in positron lifetime in material that is in the superconducting or the normal state. In both $\text{Ba}_{0.6}\text{K}_{0.4}\text{BiO}_3$ and $\text{Ba}_{0.5}\text{K}_{0.5}\text{BiO}_3$, we found a significant change in the magnitude of the positron lifetime due to aging of our samples, signifying a change in either the electronic or defect structure. We infer from these data that defect structures or populations that do not affect the superconducting properties of BKBO are the dominant

contribution to the measured positron lifetime, and these defect-related properties changed in our samples as the materials aged.

The range of lifetimes in our data fit well with the results of recent calculations (Sterne et al., in press) on the positron lifetime in BKBO in which various cation and oxygen vacancies are present. The correspondence between theoretical calculations and experimental results lends strong support to the identification of defects as the dominant contribution to the measured positron lifetime. Theoretical calculations indicate that the defect population of oxygen vacancies as well as that of barium and potassium

vacancies are likely candidates responsible for the observed changes in positron lifetime as material ages. ■

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Ion Collisions with Highest Charge-State Ions Extracted from an Electron-Beam Ion Trap

D. H. Schneider, M. W. Clark, J. W. McDonald, and D. R. DeWitt

We are producing high charge-state ions, such as Th^{80+} , through electron impact ionization in an electron-beam ion trap and efficiently extracting them from the trap to enable ion-matter interactions with the charged slow ions.

The development of new sources of very-high-charge, low-kinetic-energy ions is stimulating detailed experimental

studies of ion-matter-interaction phenomena. The electron-beam ion trap (EBIT) (Levine, 1988, 1989) is an extremely powerful new source for investigating atomic structure and electron-ion and ion-matter interactions. Highly charged ions are produced via successive electron-impact ionization of ions confined in EBIT by the dense electron beam, the magnetic field, and the electric potentials. Marrs et al. (1988) describe the operation and details of the production and loss mechanisms of the highly charged ions. EBIT can produce ions of very high charge states (up to U^{82+} so far) while keeping their kinetic energy very low (eV range). This ability makes possible high-resolution x-ray spectroscopy using the very-high-charge-state ions and efficient studies of processes such as dielectronic recombination within the trap.

Because EBIT is an ion trap, a distribution of ions with different charge states exists within it at all times and limits studies that are based on the observation of x rays. This charge-state distribution implies ambiguity in certain measurements such as collision cross sections. In addition, the only excited atomic states that EBIT produces are caused by the electron-beam impact; thus, ion-atom-excitation phenomena are not included.

In FY90, we improved EBIT's capability by adding a system that efficiently extracts up to about 10^3 Th^{80+} ions/s (Figure 1). This surprisingly high efficiency, i.e.,

the number of extracted ions per pulse available for experiments outside EBIT, opens unexpected avenues for new physics (Schneider et al., 1990). We have measured the ion-extraction performance, characterized the charge-state distributions, and studied the electronic interaction of multicharged ions with surfaces at low velocities. Electron-impact ionization-excitation studies and measurements of dielectronic recombination can be conducted most effectively by observing x-ray emissions from the trap and measuring the charge-state distributions of extracted ions. We can study collisions of highly charged, high-Z ions with surfaces and gas targets by using the extracted ions, which carry up to a few hundred keV potential energy but relatively little kinetic energy (about eV) if they are decelerated. Because the ion pulse has a narrow emittance, we should be able to efficiently retrap these ions in a secondary trap and use them for future applications without losing too many. We can then perform precision mass spectroscopy on the retrapped ions, study laser-ion interaction, and observe the retrapped ion phase transitions (crystallization).

For example, the first ion-collision studies measured ion-surface interactions using Xe^{44+} and Xe^{48+} ions that were produced in the trap with 9-keV electron impact. We bombarded a solid copper surface, oriented at 45 deg relative to the ion-beam axis, with the xenon ions.

Figure 1. Spectra of (a) charge-analyzed thorium ions (Th^{80+} and Th^{76+}) and (b) charge-analyzed uranium ions (U^{67+} and U^{64+}) following 9- and 24-keV electron impact, respectively, in EBIT. This experiment marks the first time that such high-charge-state ions have been extracted from an ion source.

The consequent xenon M- and L-shell x-ray emissions following the decay of the excited ions were observed with a Si(Li) detector perpendicular to the ion-beam axis. The xenon x-ray emission not only confirmed the charge state of the extracted xenon ions but also allowed us to estimate the number of ions extracted, which was about 10^4 s^{-1} and agreed with the directly measured rate of the charge-analyzed Xe^{44+} ions.

Figure 2 shows the M- and L-shell x-ray spectra following Xe^{44+} , Xe^{45+} , and Xe^{48+} ion impact on copper. Because no L-shell vacancy exists in the Xe^{44+} ion, its spectrum consists only of the M-shell x rays. The Xe^{45+} and Xe^{48+} spectra show additional intensities from L-shell x rays. The spectra indicate an energy shift toward higher energies with increasing charge states in the L-shell x-ray spectra caused by the increasing L-shell vacancies. The energy of the x-ray emission qualitatively implies that a "hollow" atom configuration for Xe^{48+} is formed during the neutralization process that occurs as the ion approaches the copper surface. The measured line positions demonstrate that electron capture into high n states occurs and is followed by fast radiative decay. ■

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Figure 2. M- and L-shell x-ray emission spectra obtained from (a) Xe^{44+} , (b) Xe^{45+} , and (c) Xe^{48+} ion impact on a copper surface (45 deg). The calculated x-ray transition from 3s to 2p is indicated by the central broken line. The x-ray transitions between different shells are described by L_{α} , $L_{\beta 1}$, $L_{\beta 2}$, L_{γ} , and $M_{\alpha,\beta}$. This experiment marks the first observation of x-ray emissions in ion-surface interaction using high-charge-state ions.

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FY90: \$200,000

Gamma-Ray Telescope for Arcminute Imaging of Cosmic Sources

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The gamma-ray arcminute telescope imaging system (GRATIS) represents an important new tool for understanding the nature of energetic cosmic sources. GRATIS consists of 800 cm² of gamma-ray image intensifiers coupled to a coded-aperture imaging optic. The 4-m focal length of this telescope is the longest ever flown on a balloon. The first flight of GRATIS will take place in the fall of 1991. During this flight from Texas, the system will observe several clusters of galaxies and the Crab Nebula. During the second flight from Australia, GRATIS will observe the galactic center.

Optical and x-ray astronomers have powerful telescopes that can take extremely high-resolution images of cosmic sources. Knowing the distribution of radiation around such a source, rather than merely the amount of emitted radiation, can reveal crucial information about the inner workings of a source. To date, however, gamma-ray astronomers have had no means by which to obtain high-resolution

images. Indeed, it has proven difficult in many cases to even accurately locate gamma-ray sources.

The current impasse in gamma-ray astronomy largely arises from technical circumstances. Imaging any radiation requires a position-sensitive detector and an imaging optic. A simple example is a camera with its lens and film. However, it is difficult to construct good position-sensitive detectors and imaging optics in the gamma-ray region. This difficulty has hampered the development of high-performance imaging systems.

During FY89, we concentrated on developing advanced gamma-ray detectors and imaging techniques that could overcome some of the limitations of currently available hardware. During FY90, we proceeded with fabrication, assembly, testing, and calibration of the balloon payload. We have now integrated a variety of components into the payload. These components include detectors, optics, data-acquisition electronics, active and passive shields for background rejection, hardware for precision pointing and stabilization of the gondola, and telemetry for transmitting data to the ground and providing a command link to the balloon.

During our first flight, scheduled for the fall of 1991, the balloon payload will be launched from the National Scientific Balloon Facility in Texas. Our primary targets on this flight are objects called clusters of galaxies, which are vast agglomerations of hundreds of galaxies.

Because some of these clusters are known to be powerful gamma-ray emitters, we hope to gain detailed information about where in the cluster the gamma-ray emission originates. Possible sources are the nuclei of individual galaxies, halos around the galaxies, or the space between galaxies. We also plan to investigate the extent of gamma-ray emission and how emission depends on the energy of gamma rays. Answers to such questions should provide important insight into the workings of clusters, their energy sources, how they heat and cool, and how they interact with their environment.

After the initial flight in the northern hemisphere, we plan a second flight from Australia to observe the galactic center. The galactic center is a region filled with many sources of intense, time-variable gamma radiation. At least one of these sources may be associated with the presence of a massive black hole at the center of the galaxy. With its high angular resolution, GRATIS should be a powerful tool for probing the high-energy phenomena that occur in these objects and, thus, lead to a better understanding of this complicated region.

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Gamma Rays from the Galactic Center

E. P. Liang and C. D. Dermer

We have modeled the 100- to 400-keV MeV gamma-ray emission from the center of the Milky Way Galaxy with a hot, optically thin, pair-dominated plasma. Our results indicate that the central accreting black hole is not supermassive.

The episodic appearance of 511-keV electron-positron annihilation line and enhanced MeV gamma-ray continuum emission (Riegler et al., 1985) from our galactic center is an intriguing astrophysical phenomenon. This appearance has been interpreted as a manifestation of a massive accreting black hole (Lingenfelter and Ramaty, 1982). Because the spectrum resembles that from Cygnus X-1, we modeled the galactic center with the thermal pair-cloud model applied to that candidate (Liang and Dermer, 1988). The MeV gamma rays are thought to originate from a hot, optically thin plasma where photoproduced pairs outnumber hydrogen atoms.

Assuming that the dominant processes are bremsstrahlung, Compton scattering, and pair creation and annihilation, we used Monte Carlo simulations to compute the output spectra. By fitting these spectra to the observed MeV continuum, we obtained a best-fit temperature, Thomson depth, and cloud radius. Then we derived the mass from first principles using the theory of relativistic accretion disks around black holes.

Figure 1a compares the galactic-center x- and gamma-ray data obtained by the High-Energy Astronomical Observatory-3 with model fits. Our results give a best-fit pair-cloud temperature of about 550 keV. For a source at a distance of

8.5 kpc, the gamma-ray luminosity above 400 keV is 2.2×10^{38} erg/s. The 3- to 400-keV x-ray luminosity is estimated (Matteson, 1982) to be 8.5×10^{37} erg/s. Hence, the gamma-ray-to-total-luminosity ratio is about 0.72 with < 20% uncertainty.

This ratio plus the cloud radius (derived from the temperature to be about 7.25×10^8 cm) leads to an estimate of the black-hole mass as a function of the accretion disk "Keplerianess" parameter b . If $b = 1$, the accreted material retains Keplerian angular momentum at $6 \text{ GM}/c^2$. If $b = 0$, accreted material falls in radially at $6 \text{ GM}/c^2$. Thus the black-hole mass ranges from

about 100 to $250 M_\odot$ (Figure 2). Further work is needed to determine whether this model also can explain the observed 511-keV-line intensity. ■

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Figure 1. High-Energy Astronomical Observatory-3 galactic-center data (Riegler et al., 1985) compared with model pair-cloud spectra. The best-fit temperature of 550 keV helps define the size of the pair cloud.

Figure 2. Black-hole mass (M) in units of solar mass ($M_\odot$) as a function of accretion parameter b . Dashed boundary corresponds to 20% uncertainty. Figure indicates that the black hole is not supermassive.

Recovery System for CFC Refrigerant

S. D. Hayes

We have designed and built a recovery system for quickly and safely removing chlorofluorocarbon refrigerant from refrigeration systems for reuse or disposal.

Our goal was to build a recovery system that would quickly and safely remove chlorofluorocarbon (CFC) refrigerant from a refrigeration system for reuse or disposal. We designed and built a recovery system that uses a portable refrigeration compressor to maintain a pressure difference

between the recovery vessel and the refrigeration system whose refrigerant is being removed (Figure 1). The recovery compressor draws vapor from the recovery vessel and discharges it into the refrigeration system, thus pressurizing the system. This applied vapor pressure forces the liquid refrigerant out of the system and into the recovery vessel through a separate liquid hose. When the liquid is completely evacuated from the system, the valves are reversed and the remaining vapor is pumped out of the system and into the recovery vessel.

The recovery system uses an external-drive compressor with an oil-return system and a valving manifold, all mounted on one hand truck. For easy portability, the recovery vessels are mounted on separate hand trucks. Each vessel has a filter drier and a solenoid-actuated check valve on the incoming refrigerant line. A float switch in each vessel is wired to stop the compressor and close the check valve when the vessel is full.

The rate of liquid transfer varies from 2 to 4 lb/min, which is satisfactory for most situations. A limiting factor is the size of the liquid outlet on the refrigeration system being evacuated, usually only 1/4 in. The pressure difference between the recovery vessel and the refrigeration system is usually between 5 and 25 psi, which is smaller than we expected but adequate for liquid transfer with the 3/8- and 5/8-in. hoses used. The small pressure difference means that the vapor pumped from the recovery vessel is denser than it would be otherwise, so a larger, heavier motor is required. The compressor can pump the remaining vapor from the refrigeration system after the liquid is evacuated, provided no large pocket of liquid is left to be evaporated.

The float switches in the recovery vessels work very well. They do not open prematurely even though the refrigerant liquid is boiling. The valve manifold for reversing the flow is sturdy and allows maximum flow. An electrically operated reversing valve may be more convenient.

No one recovery method will work for all refrigeration systems. Ours method will evacuate large systems such as air conditioners. Its best feature is the safe recovery-vessel design. ■

Figure 1. Schematic diagram of our recovery system for removing CFC refrigerant from a refrigeration system. The compressor draws CFC vapor from the recovery vessel, compresses it, and forces it into the refrigeration system, where it drives the liquid CFC out and into the recovery vessel. When all of the liquid is removed, the valves are reversed to pump the remaining CFC vapor out of the system. A float switch in the recovery vessel stops the compressor and closes the check valve when the vessel is full.

ER&D Budget, Publications, and Author Index

Exploratory Research and Development FY90

FY90 Budget

Exploratory Research and Development Program

	FY90 actual cost (thousands of dollars)
Exploratory Research	\$7138
<i>Biotechnology</i>	300
<i>Chemistry and Materials Science</i>	1600
<i>Computation</i>	475
<i>Earth Sciences</i>	200
<i>Energy</i>	1200
<i>Engineering</i>	691
<i>Nuclear Chemistry</i>	402
<i>Physics</i>	2270 ^a
Director's Initiatives	9119
<i>Accelerator Mass Spectrometry</i>	1000 ^b
<i>Compact Torus Accelerator</i>	3200 ^c
<i>Human Genome</i>	700 ^d
<i>Massively Parallel Computing</i>	1342 ^e
<i>Nuclear Polarization</i>	645
<i>Semiconductor Lasers</i>	850 ^f
<i>Shape Isomers</i>	882 ^g
<i>X-Ray Holography</i>	500 ^h
Laboratory-Wide Competition	3252ⁱ
27 projects funded in FY90	
26 grants projected for FY91	
Totals	
Operating costs	\$19,509
Capital equipment budget	756

^aReceived an additional \$100,000 for capital equipment (Super-EBIT project) for a total of \$2.37 million.

^bReceived an additional \$180,000 for capital equipment for a total of \$1.18 million.

^cReceived an additional \$70,000 for capital equipment for a total of \$3.27 million.

^dReceived an additional \$73,000 for capital equipment for a total of \$773,000.

^eReceived an additional \$107,000 for capital equipment for a total of \$1.449 million.

^fReceived an additional \$60,000 for capital equipment for a total of \$910,000.

^gReceived an additional \$120,000 for capital equipment for a total of \$1.002 million.

^hReceived an additional \$6,000 for capital equipment for a total of \$506,000.

ⁱProjects in the Laboratory-Wide Competition received an additional \$40,000 for capital equipment for a total of \$3.292 million.

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