LAWRENCE LIVERMORE LABORATORY
University of California/Livermore, California, 94550

UCRL-50022-1

**LLL LASER PROJECTS
REPORT NO. 1**

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Ms. date: May 1972

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Foreword

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LASER-PRODUCED PLASMAS FOR X-RAY STUDIES

Among the laser's most remarkable characteristics is its capability for converting energy into a uniquely controllable form. No other form of energy compares with coherent light in the extent to which it can, for example, be selectively and simultaneously localized in both time and space. This ability to localize radiant energy accounts for the laser's unique capability for producing exceedingly high levels of internal energy in matter. Lasers developed and operated at LRL are creating small, high-density plasmas with temperatures comparable to those found in nuclear explosions and the interiors of stars. Such laser-produced plasmas can provide laboratory-sized sources of both thermonuclear neutrons and soft x rays. Further-

more, these miniature stars can be reproduced dozens of times a day for controlled and repeatable studies. The following discussion describes the initial phases of an investigation of the yield and spectral character of the x rays emitted by dense, high-temperature, laser-produced plasmas.

The neodymium-glass laser system (called *Plasma X*) used for this study is shown in Fig. 12. Although it can produce subnanosecond pulses with peak powers of roughly 10^{11} W, it is relatively conventional compared to other laser systems under development at LRL. The heart of the *Plasma X* laser system is the mode-locked laser oscillator. It can produce a train of infrared pulses, each of which is a few

Fig. 12. The *Plasma X* laser system for producing high-density, high-temperature plasmas, and the apparatus for studying the x rays emitted from the plasmas. A mode-locked laser generates a train of very short pulses. One pulse is selected from this train, amplified, and focused to a point on a target foil to produce a plasma. The x-ray spectrum is analyzed with a crystal spectrometer and scintillation detectors. The first five lasers in the *Plasma X* system use rods of neodymium-doped glass that are 0.5 in. in diameter and 9.5 in. long. The final laser amplifier uses a rod 1 in. by 19 in.

picoseconds in duration — or a few millimeters in actual length. With auxiliary components, subpicosecond pulses could be produced or, as in the case of the oscillator shown in Fig. 12, the pulses can be lengthened to a range of several tens of picoseconds. Such ultrashort pulses can deliver a quantity of energy to a submillimeter-sized target area before the energy can be dissipated by explosion or by thermal conduction. Such a quick-delivery system is essential for the production of very high temperatures with relatively small energy capabilities.

The key to the operation of the mode-locked oscillator is a cell a few millimeters thick containing a solution that acts as a saturable absorber. This solution tends to absorb the light photons that are reflected back and forth between the mirrors of the laser oscillator — except when the photons arrive in bunches. Thus the short, intense light pulses, which bleach the absorbing liquid, are selectively amplified. Or, stated differently, the saturable absorber acts as a nonlinear element to organize and combine the optical frequencies or modes within the laser oscillator cavity into a very short light pulse. Each time this short pulse hits the partially-reflecting output mirror, part of the pulse is emitted from the oscillator. The total emission from the oscillator is, therefore, a limited sequence of very narrow pulses separated by the time it takes light to make one round trip within the oscillator. The pulses first grow in amplitude, due to the gain in the oscillator laser, and then decrease as the light energy pumped into the laser rod by the flash lamps is depleted.

Although the use of multiple pulses may prove to be of great importance for the production of high-temperature plasmas, current practice is to select a single pulse from the multipulse envelope generated by the mode-locked oscillator. A single pulse is selected by a fast-acting electrooptic shutter whose action depends on the polarized nature of the train of light pulses. The unwanted pulses are excluded by crossed Glan-Thompson prisms. Between these prisms are Pockels cells to which voltage can be momentarily applied so as to rotate the plane of polarization of the desired pulse and permit that pulse to pass through the shutter. A high-speed switch, in the form of a laser-triggered spark gap, turns the voltage on for the brief time required to separate a single pulse from neighboring pulses that are only 10 nsec away.¹⁹

The single ultrashort light pulse passed by the electrooptic switch contains only a few millijoules of energy, but it can be amplified to provide sufficient energy to produce a high-temperature plasma. With the Plasma X laser system a gain of about 1000 is employed after the electrooptic switch. Such amplifi-

cation is straightforward, except that the laser pulse soon grows intense enough to destroy the laser rods as it passes through them. For this reason we use a larger-diameter rod for the last amplifier stage. Another problem is the tendency for such a high-gain amplifier system to act as an oscillator. This problem sometimes requires the use of an optical isolator. A more challenging problem, involving the entire laser system, concerns the measurement and control of the details of pulse shape and beam uniformity. It appears that these subtle pulse characteristics may be of great importance for the reproducible creation of very-high-energy plasmas.

The final operation on the laser pulse is to reduce its cross-sectional area with a converging lens from a few square centimeters to about 10^{-4} cm². The final needle-sized light pulse delivers energy to the target region at a peak rate of approximately 10^{11} W, and provides an intensity of 10^{15} W/cm².

An important question arises at this point in the experimental arrangement. The laser system provides an extremely intense concentration of radiant energy, but it does not necessarily follow that these infrared photons will efficiently communicate all their energy to the elementary particles of the target material. In most laser-heating situations the vibratory nature of the light pulse can be thought of as simply rattling the electrons, which collide with ions and absorb energy. Expressed another way, the photons of the laser pulse add energy to the plasma electrons by inverse bremsstrahlung. However, this process is only efficient for plasma temperatures up to the kilovolt range ($\sim 10^7$ °C). The attainment of much higher temperatures with this heating mechanism might be very difficult. Fortunately, there are theoretical reasons for believing that auxiliary heating mechanisms, involving the excitation of plasma instabilities, may begin to occur at the very high laser light intensities now becoming available. We are searching for evidence of these auxiliary heating mechanisms.

The laser system of Fig. 12 has been used primarily for studies of plasma radiation. The focused laser pulse converts a small portion of the target — generally a foil of a high-Z metal — to a hot, dense plasma. Since plasmas produced with this system have peak temperatures that are believed to lie above 10^7 °C, a significant fraction of their total emission is in the keV x-ray region. Such plasmas represent reasonable approximations to natural and man-made plasmas of great importance.

A continuing computational study of the radiations to be expected from laser-produced plasmas provides guidance and understanding for the experimental program. The machine code used for these

computations includes the treatment of hydrodynamic motion and radiation transport, and does not require the usual restrictive assumptions concerning local thermodynamic equilibrium for the bound-state populations. A particularly interesting prediction of the computations is that nearly half of the total radiation from high-temperature, high-Z plasmas will consist of x-ray line emission. However, accurate calculations of the positions and intensities of x-ray lines emitted by laser-produced plasmas are very difficult. This is why the experimental program is required. If we can develop a verified computational approach, including the ability to predict characteristic line emissions, we will have the basis for a powerful tool for the exploration and understanding of ultrahigh-energy plasmas.

Several types of detectors have been used successfully in measuring the radiation from our laser-produced plasmas. Thermoluminescent crystals, scintillation detectors, and x-ray-sensitive film all appear to be useful. The scintillation detectors, for example, which employ photomultiplier tubes with gains above 10^7 , are extremely sensitive to x rays. They permit a selective examination of the very-high-energy x rays that pass through relatively thick absorber foils. More generally, the measurement of relative x-ray intensities transmitted through a wide range of foil thicknesses is beginning to provide evidence about the general shape of the x-ray spectrum,

Fig. 14. Closeup of the curved-crystal spectrometer. See Fig. 15 for details.

and hence an indication of plasma temperatures. Figures 13 and 14 show some of these detectors.

One of the most promising diagnostic techniques involves the use of a crystal that can sort out the energies of incident x rays by Bragg reflection. For each angle θ at which the x rays intercept such a crystal, the crystal selectively reflects only those whose wavelength λ satisfies the Bragg criterion $n\lambda = 2d \sin \theta$, where d is the crystal-lattice spacing and n is the order of the reflection. Differences in energy are thereby converted to differences in direction, permitting the x-ray energy spectrum to be recorded on film. This spectral record is characteristic of a particular plasma. However, the burst of x rays from a superhot plasma is extremely short, and only a small fraction of the x rays incident on the crystal is selectively reflected. Our first attempts at crystal spectrometry failed to yield a detectable film image. But a special high-reflectivity crystal, formed by compression-annealing of pyrolytic graphite, provided part of the answer, and a cylindrical version of the same crystal boosted the intensity even further. The curved crystal is arranged as shown in Fig. 15 to produce a line focus of the Bragg-reflected x rays. A range of x-ray energies corresponding to the range of Bragg angles subtended by the crystal is spread along the line focus. As shown in Figs. 13 through 15, the target foil and the x-ray film are placed in the same plane, at a distance from the crystal equal to its radius of curvature. This is about 4 cm for the crystal

Fig. 13. Target chamber used for studies of the x rays from laser-produced plasmas. The laser beam enters at "4 o'clock," i.e., right foreground. The tubes at 3 o'clock and 6 o'clock contain scintillation detectors. The curved-crystal spectrometer is in the center of the target chamber (see Figs. 14 and 15).

Fig. 15. Schematic of the curved-crystal spectrometer used to study x rays from laser-produced plasmas. The dashed lines indicate the x rays intercepted and Bragg-reflected by the crystal.

shown. Since the crystal we presently use is only 3/8-in. wide, the spectral range registered with each laser pulse is quite narrow. However, the crystal can be moved laterally (parallel to its reflecting surface) between shots so that an extended portion of the x-ray spectrum can be recorded in a sequence of separate laser pulses.

We have begun to use the bent-crystal spectrometer to explore the x-ray spectrum in the range from about 2 to 3.5 keV. For plasmas formed from lead and gold target foils, even this narrow spectral region appears to contain three reproducible and characteristic spectral features. The features are similar for the two target materials, but they are shifted to higher energies for the lead plasma, as expected. An important objective of future work is the association of such experimentally determined features of x-ray spectra with emission lines or groups of lines pre-

dicted by computations. Our efforts to accomplish this include an expansion of the computational approach to include more of the appropriate physical details, and a refinement and extension of the spectrometer's capabilities.

Future research with laser-produced plasmas promises to yield increased knowledge of high-energy plasmas that are, in some respects, comparable to those produced in nuclear explosions. Lasers with extended capacities for heating plasmas are nearing completion at LRL, and a number of promising plasma diagnostic techniques are being explored. Furthermore, we have access to uniquely appropriate computational capabilities. These considerations suggest that plasmas of even greater interest and diversity will soon be available and that fruitful studies of their characteristics will be possible.

DOUBLE-DISCHARGE CO₂ LASER

We have studied corona discharge as a method of initiating a uniform glow discharge in a transversely excited, atmospheric-pressure (TEA) carbon-dioxide laser at high efficiencies. With He/CO₂/N₂ mixtures, we have obtained typical laser output energies of 17 J/liter, indicating that we are converting electrical energy to infrared laser light at an efficiency of 24 percent. A typical pulse from this laser contains 100 J of energy and is 100 nsec long. Unique to our double-discharge TEA laser is the use of capacitor-bank discharge, which provides for simplicity, flexibility, and greater efficiency than earlier lasers of this type.¹⁶ We are continuing to study the characteristics of this laser.

Figure 21 shows a schematic diagram of our double-discharge TEA laser, and Fig. 22 gives some details of its construction.

A CO₂ laser offers potential for supplying the short, high-energy laser pulses required for igniting fusion devices. A chief advantage of the CO₂ laser is that it appears capable of being scaled up to high outputs with present or near-future technology. LLL's present plan for achieving the short-pulse and high-energy requirements is to amplify the subnanosecond pulse

generated by a mode-locked CO₂ oscillator in a chain of CO₂ amplifiers. Irrespective of the future development of this plan, the fundamental question that must first be resolved is how to produce the necessary population inversion in the gas medium at atmospheric pressures with good uniformity and efficiency. The laser must be operated at relatively high (i.e., atmospheric) pressures and at large volume if we are to achieve laser pulses with sufficiently high total energy and high peak power.

In CO₂ lasers, population inversion (pumping) is accomplished by glow discharge. At low pressures uniform glow discharge is easily attained, but at higher pressures it becomes more difficult to achieve — arcing tends to occur, limiting the gas excitation to a small volume. Thus it is difficult to achieve uniform glow discharge at atmospheric pressures over a large discharge volume.

Conceivably, glow discharge can be accomplished in any gas between two electrodes by making the discharge time short compared to the arc-formation time, or by limiting the discharge-current density enough to prevent the formation of a constricted arc. Since the arc-formation times in gases at atmospheric

Fig. 21. Schematic diagram of our double-discharge TEA CO₂ laser. The use of a Marx bank, or capacitor-bank network, to trigger the discharge in this type of laser is believed unique to LLL.

Fig. 22. Some details of the construction of the double-discharge laser. A corona discharge between the cathode plates and nichrome wires initiates a uniform glow discharge between the cathode and the anode.

pressures are typically very short ($\leq 10^{-8}$ sec), it is difficult to make the discharge time shorter than the arc-formation time. Thus the alternative of limiting the discharge-current density must be applied.

The discharge-current density can be limited either by constraining the total current flow or by maximizing the discharge area. Since the total energy output of a gas laser is related directly to the total current flow in the discharge (within limits), the idea of limiting the total discharge current is unattractive. The only alternative, then, is to enlarge the discharge area. This is made possible by *triggering*, that is, by inserting electric charges between the two electrodes before the main discharge occurs.

When a single charge is inserted in the gas between two electrodes under tension, the charge causes a cascade or avalanche, which triggers the discharge between the electrodes. If a large number of charges are placed uniformly between these electrodes, then the avalanches triggered by these charges will initiate a uniform discharge over the entire volume between the electrodes. Ultraviolet illumination,¹⁷ electron-beam discharge,¹⁸ and corona discharge^{19,20} have been used to create the initial charges needed to trigger glow discharges in CO₂ lasers.

A CO₂ laser using the ultraviolet-illumination idea is simple and compact. The applicability of this technique to large systems, however, is questionable because of the difficulties involved in producing intense UV radiation for extended periods. A more attractive alternative is the use of an electron beam to initiate and stabilize the uniform glow discharge. This technique allows great flexibility in the density of the trigger electrons and in the discharge duration. These advantages must be balanced out with the complexity associated with the use of an electron-beam gun. LLL is pursuing an investigation of an electron-beam-initiated system. In addition we are studying the use of corona-discharge initiation. This latter method may have some of the advantages of the UV and electron-beam initiation techniques without the associated limitations and complexities.

Figure 22 shows details of our TEA CO₂ laser. The cathode is formed by a number of thin aluminum strips, and the trigger electrodes are made from nichrome wires inside glass capillary tubes. The anode

is a flat aluminum plate. Lucite sheets are used for supporting parts. When a negative high-voltage pulse is applied to the cathode, corona discharge occurs between the strips and the wires. Electrons are ejected from the edges of the strips; the result is a uniform layer of ionization near the cathode. This uniform electron layer initiates a uniform glow discharge between the cathode and the anode. Since two different discharges occur in this system, it is called a double-discharge TEA CO₂ device.

We have operated our double-discharge laser with a Marx bank (i.e., capacitor bank network) at high output energies and efficiencies over a large discharge volume (~5.5 liters). The best performance with the existing Marx bank was obtained with a He/CO₂/N₂ gas mixture of 5:1:1 at 56 kV for a 5-cm discharge gap. The mirrors that gave the best results (see Fig. 21) were a flat germanium output mirror (50-to-60-percent reflectivity) and a spherically curved mirror of gold (5-m radius of curvature, totally reflecting).

Measurements taken at the center of the laser beam gave a typical laser output energy of about 17 J/liter. This is equivalent to an efficiency of 24 percent. Our pulse-to-pulse variation in the output energy was less than 10 percent; this variation is due partly to the voltage variation in our unregulated power supply. Pulse length is typically 100 nsec. Pulses with durations between 50 nsec and 15 μ sec can be attained. The energy in a typical pulse is ~100 J; this corresponds (for a 100-nsec pulse) to a peak power of $\sim 1.7 \times 10^7$ W/cm². Our TEA laser can be pulsed about every 10 sec. Simple modifications would allow a higher repetition rate.

We have observed super radiance over a discharge length of 2 m; that is, lasing occurred when no end mirrors were in place. Self-mode-locking²¹ has also been achieved. The duration of the sub-pulses produced in this mode is less than 5 nsec.

We are making a detailed study of this double-discharge device. In the future we will attempt to operate the laser at greater than atmospheric pressures, and will double the active volume of the lasing medium by doubling the anode-to-cathode spacing (to 10 cm). Our present results indicate that this discharge scheme is an attractive alternative to the approach using electron-beam initiation.

PLASMA PRODUCTION WITH LLL'S LONG-PATH, NEODYMIUM-GLASS DISK LASER

We have constructed a unique neodymium-glass laser for studying the production of thermonuclear plasmas.²² This laser is designed to achieve outputs of up to 500 J in pulses 0.1 to several nanoseconds

long. Multiple passes through a 16-disk output stage permit high power amplification. To provide for multiple passes, the end mirrors in the output stage are placed 68 m apart; the total path length is 612 m.

Fig. 23. The long-path laser. A series of rod amplifiers provides input to the disk amplifier, which consists of 16 neodymium-glass disks 14 cm in diameter.

Major modifications have just been completed. Experiments prior to the modifications gave preliminary information on the behavior of laser-produced, neutron-emitting plasmas. Nanosecond pulses with energies as high as 200 J were focused on CD_2 and CH_2 targets, and the resulting neutrons (as many as 4×10^4), x rays, and reflected light were studied. The data from such experiments, together with computer calculations, should give us a better understanding of the complex processes taking place in laser-produced plasmas.

Figure 23 shows the laser, consisting of: (1) an oscillator/switchout arrangement that produces pulses of duration selectable from 1 to 15 nsec; (2) seven amplifiers, each built around a laser rod 13 mm in diameter and 24 cm long; (3) three amplifiers, each built around a laser rod 25 mm in diameter and 48 cm long; and (4) a 9-pass disk amplifier of 140-mm aperture (see Figs. 24 and 25). The assembly of disks is about 2 m long, but a spacing of about 33 m is required between the disk assembly and each of

Fig. 24. Multiple passes through the disk amplifier. To obtain maximum gain and stored-energy utilization from each disk, we allow a high-quality laser pulse of low intensity to pass repeatedly through the amplifier by a series of distinct paths. The arrangement of the mirrors that direct the laser pulse through this series is shown above. Most of the mirrors have been replaced with prisms, as called for in the original design.

Fig 25. Layout of the neodymium-glass disk laser amplifier. Each of the 16 neodymium-glass disks is 140 mm in diameter and 25 mm thick. When edge-clamped and positioned at Brewster's angle, each disk presents a surface area of 125 cm^2 to the beam, while the area of the emerging beam is 64 cm^2 . The overall length of the disk head is about 2 m. Twenty 5-tube flash-lamp modules pump the disk amplifier. They are powered by a system of capacitor banks that can deliver up to 360 kJ of electrical energy in a 1-msec pulse.

reflecting mirrors. The laser is intended to produce light pulses of up to 500 J with durations variable from 0.1 to several nanoseconds. To date it has delivered approximately 400 J in 5 nsec and 300 J in 2 nsec. These energies have not been obtained repeatedly, but recent system modifications are expected to improve this situation.

The rationale for this laser design follows from the vulnerability of glass components at high energy

densities and from the limited diameter possible with laser-rod amplifiers. Most high-quality optical glass can be damaged by pulses of 20 J/cm^2 of a few nanoseconds duration. Once a glass laser produces energy densities within a factor of 2 to 4 of this value it cannot be pushed farther without fear of damage. Hence if one wishes to produce 1000 J in a few nanoseconds, he must design the laser so that the beam intercepts a glass-surface area of at least 100 cm^2 .

Rod amplifiers cannot be scaled up to the required aperture because, at large diameter, the light from the flash lamps cannot be deposited uniformly across the diameter of the rod. The neodymium absorbs the light strongly, and, in a 76-mm-diam rod, most of the light is absorbed near the rod surface unless the absorber concentration is reduced to 0.3 to 0.1 of its normal value. If the absorber concentration is lowered the gain and energy storage in the rod are reduced drastically. The largest rod lasers in use have diameters of 63 mm.

We have used glass disks to build our output laser amplifier. The 16 disks in this output stage are face-pumped and oriented at Brewster's angle. By face pumping we uniformly deposit energy through the cross section of the amplifier, thereby getting around the absorption problem inherent in large-diameter rods. Putting the disks on an angle increases the glass-surface area intercepted by the beam. We orient the disks at Brewster's angle to minimize reflection losses from the surfaces of the disks.

The disks are pumped with 20 flash-lamp modules, each of which is supplied with a maximum of 18 kJ from a capacitor bank. The stored energy in the disks

Fig. 26. Some of the diagnostic equipment used in plasma-heating experiments with the long-path laser. A large plastic scintillator located above the target detects the neutrons emitted by the plasma. The scintillator is shielded by 2 cm of lead to exclude high-energy x rays; it is viewed by two photomultiplier tubes. Two scintillation detectors at angles of 45 deg from the incident beam monitor the x rays from the plasma.

Fig. 27. Some of the beam diagnostics used to measure characteristics of the pulse from the long-path laser. The reflected-pulse photodiode measures the laser light reflected by the plasma (see Fig. 29).

Fig. 28. Total neutron emission as a function of laser-pulse energy for a series of plasmas produced from CD, with the long-path laser.

is 800 to 1500 J. Because the single-pass gain for 1-to-5-nsec pulses is 3 to 6, the input pulse energy would have to be 200 to 400 J if the amplifier were to achieve 500 to 1000 J in a single pass. A 200-to-400-J input pulse energy can be achieved with a 63-mm-diam rod amplifier. By introducing a pulse of small input energy and passing it through the disk amplifier many times we hope to achieve pulse energies as great as 500 J without such a prior stage.

The decision to use multiple passes led to the long path of the disk amplifier. The passes had to be made in a nonresonant fashion to prevent premature and unwanted amplification of noise, which would deplete the stored energy. These passes had to be made with the full-sized beam, with each pass being at a small angle with respect to the others. Sending the beam through the disk amplifier at an angle to the axis resulted in cutting off a portion of the beam. Setting

Fig. 29. Input and reflected pulses for a plasma produced with the long-path laser. The double input pulse is altered in shape as reflected from the focusing lens and evolving plasma. A large reflected pulse, as shown, appears to accompany the effective production of neutrons and high-energy x rays. The input and reflected pulses are measured with separate photodiodes.

the maximum allowable cutoff at 10 percent, we calculated the required angle between the beams. The required interbeam spacing at the reflecting mirrors set the distance between disk amplifier and mirror at approximately 33 m.

The long-path laser has experienced several problems, the solution of which has required major modifications. Self-oscillations through the entire laser and damage caused by spontaneous-emission coupling between adjacent amplifiers led to our separating each rod amplifier by the Rayleigh distance. Damage to the turning mirrors led us to replace them with prisms. Amplified feedback of reflections from laser-produced plasmas forced us to install two Faraday-rotator isolators to prevent damage to the laser rods. Turbulence in the air between the disk amplifier and the turning mirrors was solved by enclosing the air columns of the lines of sight. Degrading nonlinear optical effects in the enclosed air column led to the use of a helium atmosphere in place of the air column. We are now studying the performance of the laser after the latest modifications.

In preliminary experiments prior to these modifications, we focused the beam (10 to 100 J in 1 to 5 nsec) on slabs of CH_2 and CD_2 and measured the resulting x-ray emission, neutron emission, and reflected-light intensity. The beam was focused with a f/8 lens to a spot about 50 μm in diameter. The power density at the focal point was a maximum of

$\sim 2 \times 10^{14}$ W/cm². X-ray detectors were set up to look at both soft and hard x rays (which were selected by thin and thick absorbers in front of the x-ray scintillators). Neutrons were detected by a plastic scintillator and photomultiplier tube (Fig. 26). Measurements of reflected light were made with photodiodes as shown in Fig. 27. The first neutron measurements yielded the energy dependence shown in Fig. 28. Later experiments did not reproduce this well-defined line but yielded a maximum neutron count of 4×10^4 neutrons. These more recent shots have a greater fluctuation about the earlier data points. In addition the later results at higher energies indicate a smaller slope. The x-ray measurements indicated an excessive number of hard x rays. This excess was also detected during neutron measurements when the x rays from CH₂ had to be attenuated by 1 cm of lead to shield the neutron detector. We now believe that both the neutron emission and the preponderance

of hard x rays are due to heating by plasma instabilities rather than to thermonuclear reactions.

We did not observe neutrons after pulses shorter than 2 nsec with energies up to 200 J, but we did observe neutrons for pulses longer than 2 nsec and for double pulses of total duration longer than 2 nsec. This may be explainable in terms of the time of buildup of the heating instability. In addition, we found a strong correlation between neutron yield, x-ray intensity, and reflected-light intensity (see Fig. 29). The reflected-light pulse had considerable intensity-time structure either on the second of two pulses or later in a longer pulse. This structure was not reproducible in intensity, but the period of the modulation was regular.

After the latest modifications are checked out, we plan to repeat these plasma experiments at higher input energy.

Details of our work appear in Ref. 22.

Initiation of thermonuclear fusion by laser energy is the primary goal of laser research at LLL. In experiments now planned, laser energy will be focused on CH₂ and CD₂ slabs and on very small D-T drops to produce heated plasmas.

Laser pulses of high energy and short duration are required in these experiments. A carbon dioxide laser has the potential for supplying such pulses, but the amount of energy required per pulse is so great that a multistage CO₂ system must be used. In the system planned, a CO₂ laser oscillator will generate a short, high-power infrared pulse, and the pulse will be directed through several CO₂ laser amplifiers and focused onto a thermonuclear target. We are developing a CO₂ laser oscillator and pulse selector that can supply an input pulse.

In a major step toward developing such a pulse source, we have mode-locked a transversely-excited, atmospheric-pressure CO₂ laser to provide a train of short, high-power pulses, and have demonstrated the first reported selection of a single pulse from such a train of high-power pulses.

Figure 13 shows the major components of the CO₂ oscillator. This oscillator produces pulse trains with individual pulses less than 2 nsec wide and with a peak power of about 4 MW. The laser cavity, which forms the resonant element of the oscillator, is bounded by two mirrors 3.75 m apart. A totally reflecting, spherical gold mirror constricts the beam to a narrow "waist" of 8 mm at the output end of the cavity. The output mirror is coated for 90-percent reflection, allowing 10 percent of the incident energy to pass through to the external optics. The discharge tube, aperture, and mode-locking crystal are inside the cavity. The aperture is normally set at 8 mm and keeps the laser oscillating "on axis" to limit oscillation to the TEM₀₀ mode. (In the TEM₀₀ oscillation mode the intensity across the beam is circularly symmetric and follows a Gaussian profile centered on the beam axis.)

CO₂ Gas Excitation

In carbon dioxide, laser action is a result of vibrational energy transitions of CO₂ molecules. Figure 14 details the various energy levels involved.

Fig. 13. The mode-locked CO₂ laser. The beam narrows from about 10-mm diameter at the spherical mirror to about 8-mm diameter at the aperture. (Components and distances are not drawn to scale.)

CO_2 molecules excited by an electrical glow discharge to the 00^0_1 state emit infrared radiation with a wavelength of $10.6\ \mu\text{m}$ when they make a transition to the 10^0_0 state. Fast down-transitions from the 10^0_0 state depopulate that level with respect to the 00^0_1 .

Although the CO_2 molecules may be directly excited, they may also be excited by energy transfer from excited nitrogen when that gas is present in the discharge tube. Figure 14 indicates the first vibrationally excited state of N_2 . This state is extremely close in energy to the upper CO_2 lasing level. Vibrational energy transfer, through collisions, between excited nitrogen and groundstate CO_2 is a statistically favored reaction. This leads to a significant increase in overall excitation efficiency and output energy since a large amount of excited nitrogen is produced by the electrical discharge.

Initially, CO_2 lasers were operated at very low gas pressures — typically 5 to 20 Torr. This was because at higher pressures strong arcs tend to replace the uniform glow discharge in the gas. These arcs excite a volume of gas too small to achieve laser action. While it was realized theoretically that increasing the gas pressure would increase peak power, existing electrode designs were unable to produce uniform discharges at higher pressures.

In mid-1970, the first CO_2 laser operating at atmospheric pressure was reported.²¹ This laser made

use of a unique electrode configuration — a solid metal anode opposing a cathode made up of more than a hundred resistively-loaded pins. The resistor in series with each pin acted to limit the discharge current through each pin. The result was a uniform discharge over a large gas volume. The electrodes were placed so that the discharge was perpendicular to the laser axis and the term TEA laser, for transversely excited, atmospheric-pressure, was coined.²²

The TEA Laser

The type of TEA laser we use for oscillator research is shown in Fig. 15. The cathode consists of some 700 pins ballasted by carbon resistors spaced on 6-mm centers. The pins cover an area approximately 2.5 cm by 100 cm. The solid aluminum anode is 2.5 cm from the pin tips, resulting in an active volume approaching 0.63 liter. A Lucite enclosure is separated into a gas compartment and a space for electrical components. All discharge-circuit electrical elements, with the exception of the power supply and the trigger generator, are housed directly beneath the anode to reduce circuit inductance.

Figure 16 shows the high-voltage circuit. The low-current supply charges the energy-storage capacitor through 1.4 M Ω and 50 k Ω resistors. The capacitor stores about 45 J until a high-voltage pulse from the trigger generator fires the spark gap, closing the

Fig. 14. Energy-level diagram for carbon dioxide and nitrogen. This simplified diagram includes only those energy levels involved in the production of $10.6\text{-}\mu\text{m}$ radiation.

Fig. 15. Simplified drawing of the discharge tube. The cathode consists of about 700 resistor-ballasted pins. The anode is a solid aluminum bar. The purpose of the resistors is to limit the discharge current below that required for arc discharge, and thus to assure a uniform glow-discharge along the length of the tube.

Fig. 16. The high-voltage circuit of the laser. The spark gap, when fired by the trigger generator, rapidly discharges the capacitor through the gas in the laser.

discharge path. The spark gap acts as an extremely fast high-voltage switch.

Nitrogen increases the energy in the laser output pulse by increasing the excitation efficiency. It does so, however, by increasing the pulse length without significantly affecting peak power. Peak power is the more important parameter for fusion applications as we will show later. For this reason, the laser described here is normally operated with a gas mixture of 10 percent CO_2 and 90 percent helium. Helium acts both to depopulate the lower lasing level and to reduce kinetic and rotational CO_2 temperatures. The mixed gas flows longitudinally through the discharge tube at 50 ft³/hr.

Mode-Locking

A CO_2 TEA laser normally produces pulses of infrared energy nearly 1 μsec long. For fusion experiments, however, theory predicts maximum interaction between laser energy and the plasma at pulsewidths in and below the 1-nsec range. Full utilization of the TEA laser's high output power for fusion research would be very difficult without some means of generating very short pulses of CO_2 energy. Short pulses are generated by "mode-locking."

The oscillation frequency spectrum of any laser has a relatively narrow, but finite, bandwidth. In the case of the CO_2 TEA laser, the bandwidth (or "linewidth") is about 3×10^9 Hz (at an oscillation center

frequency of approximately 3×10^{13} Hz). The laser is not permitted to oscillate continuously over that linewidth, however, because the optical cavity only allows oscillation at discrete resonant frequencies. These resonances, or oscillation "modes," occur at spacings equal to the speed of light divided by twice the cavity length. In our oscillator, which uses a 3.75-m cavity, the mode spacing is 40 MHz. This implies that approximately 75 discrete modes can exist in the cavity within the 3-GHz linewidth.

It can be shown theoretically that when a number of cavity modes are forced to oscillate at a fixed frequency-and-phase relationship, the composite output in the time domain is a train of extremely narrow pulses. Under these conditions, the laser is said to be "mode-locked." Typical output waveforms for our laser are compared in Fig. 17. The width of individual pulses is a function of the number of modes locked and is normally of the order of 1 nsec. The spacing of the pulses in the train is governed by the cavity length and is the reciprocal of the mode spacing.

In physical terms, mode-locking may be accomplished in several ways. Of the several, we have chosen the technique known as acousto-optic loss modulation. A single-crystal germanium block, about 1 cm on a side, is placed inside the cavity near the output mirror (see Fig. 13). Bonded to one side of the crystal is a thin piezoelectric transducer. The ceramic transducer is driven at its resonant frequency of 20 MHz, intentionally chosen to be one-half of the cavity mode spacing. The transducer creates acoustic standing waves in the germanium crystal which, in turn, lead to variations in refractive index across the crystal, forming a kind of diffraction grating. The standing-wave amplitude follows the radiofrequency sine-wave excitation and hence goes to zero twice in each cycle.

Except when the standing-wave amplitude is zero, the grating introduces loss by scattering part of the laser energy out of the cavity. The crystal acts as an amplitude modulator operating inside the cavity at 40 MHz. This results in injection of modulation sidebands into the cavity modes, creating fixed frequency-and-phase relationships sufficient to mode-lock the laser.

Mode-locking through loss modulation may be understood by considering the timing of events in the operation of the laser. The acousto-optic germanium crystal operates as a fast shutter inside the cavity, opening briefly every 25 nsec (40 MHz period). Twenty-five nanoseconds is also the exact time necessary for laser energy to travel one round trip between the cavity mirrors. Hence, only a narrow pulse of energy traveling back and forth between the mirrors finds the shutter open at the appropriate time. The

Fig. 17. Effect of mode locking on the oscillator output. The maximum intensity with mode-locking is at least five times the intensity without mode-locking.

mode-locked pulse train is merely a 10-percent sample of this pulse as it strikes the output mirror. The change in pulse amplitude (see Fig. 17) is a result of the gain experienced by the energy pulse each time it traverses the discharge tube.

Using the configuration shown in Fig. 13 and the mode-locking crystal described here, we have achieved pulsewidths measured at less than 2 nsec (which is about the resolution of our detector scheme).

Single-Pulse Selection

For fusion experiments, the CO₂ oscillator must be capable of injecting a single narrow pulse into the amplifier system. Naturally it is desirable that the largest pulse in the train be selected. We separate a single pulse by using the selection optics detailed in Fig. 18. The gallium arsenide (GaAs) crystal is capable of electrically-induced birefringence. An electrical field introduces a phase decay between two components of laser polarization. For the appropriate input polarization and field strength this delay has the effect of rotating the beam's plane of polarization.

Fig. 18. The apparatus for selecting a single pulse from the oscillator output.

With reference to Fig. 18, the linearly polarized pulse train from the laser passes through the GaAs crystal in the (110) crystal direction (with the (001) direction parallel to the laser polarization). With no voltage applied to the crystal, the train continues on through the thin germanium (Ge) polarizer onto detector D1. The Ge polarizer, set at Brewster's angle (76 deg) to the laser beam, allows energy polarized in the plane of incidence to pass with nearly zero reflection. When the "half-wave" voltage is applied to the GaAs crystal in the (110) direction, the laser polarization is rotated 90 deg and is no longer in the proper orientation to satisfy the Brewster condition at the polarizer. In this case, it is nearly 90 percent reflected, along the dashed path in Fig. 18, toward detector D2.

The electronic circuitry is composed of two parts: a pulse-height discriminator and a Marx generator. The discriminator is a trigger generator that responds to a preset input-pulse amplitude. The Marx generator produces a short, high-voltage pulse when triggered by the discriminator.

To understand the operation of the selection apparatus it is simplest to follow one train of pulses completely through the system. After leaving the laser output mirror, the pulses begin passing through the GaAs crystal and Ge polarizer, and onto detector D1. The pulse-height discriminator monitors the detector

amplitude until it senses a pulse of the preset height. At that time it fires the Marx generator, which applies a high-voltage pulse of about 10 nsec duration to the GaAs crystal. If the electrical time delay and discriminator level are properly adjusted, high voltage will be applied to the GaAs crystal coincident with the passage of the largest laser pulse. The polarization of this pulse will be rotated 90 deg and it will be reflected by the germanium polarizer toward detector D2. When the next pulse arrives at the GaAs crystal 25 nsec later, its polarization will remain unchanged as it passes through. This pulse and the ones that follow it will continue through to detector D1 as earlier pulses did.

Detector D2 functions for diagnostic and monitoring purposes only. In fusion experiments, most of the single-pulse energy will be routed to the amplifier chain rather than to a detector.

Although the setup in Fig. 18 is technically feasible, we have had to modify it somewhat for the actual experiments. If the pulse train is made to travel twice through the GaAs crystal, only one-half the voltage is necessary to provide the required 90-deg polarization shift. This "quarter-wave" voltage is about 6000 V, presently the limit of our equipment. For that reason, a mirror has been incorporated into the arrangement that allows the pulse to traverse the GaAs crystal twice. One disadvantage of this system, however, is the additional precision required in optics alignment.

Using the mirror setup we have observed a single 2-nsec pulse at detector D2. We believe that this is the first time a single pulse has been selected from a mode-locked TEA CO₂ laser.

One of the most important considerations in the pulse-selection technique is the "extinction ratio" — the analog to signal-to-noise ratio. CO₂ amplifiers to be used in fusion experiments will store an extremely large amount of energy in the active medium. This energy will be released as the single pulse from the oscillator travels through the amplifier. If the oscillator pulse is preceded by a smaller, unwanted pulse, part of the amplifier energy will be wasted in amplifying this pulse. It is not clear at this time exactly which parameters most affect the extinction ratio and what the maximum attainable ratio will be. Ratios on the order of 1000:1 are near-term goals.

Future Work

For the near future, three areas are of primary concern. First, further experimentation is necessary on the oscillator itself to determine parameter relationships to allow optimization. Second, development work remains in the area of single-pulse selection. Reliable measurements of the extinction ratio will be required before we can optimize our selection technique. Investigation of optical and electronic components is continuing in an effort to find equipment that will best serve this purpose. Third, we are planning research into such areas as multiline oscillation, pulse shaping, and mode-locking of other gas lasers.

While considerable theoretical and experimental work remains, the system has shown much potential and should prove to be a most valuable tool for LLL laser-fusion research.

ULTRAFAST STREAKING CAMERA FOR LASER DIAGNOSTICS

We have constructed an ultrafast electro-optical streaking camera for detailed studies of the short pulses of radiation produced by Q-switched and mode-locked lasers. The camera transforms intensity-versus-time information in an incident pulse into blue-light intensity as a function of position along a fluorescent screen. The time resolution of the camera is ~ 10 psec (10^{-11} sec); it uses an image-converter tube and an image intensifier that are available commercially. Unique to LLL's ultrafast camera is the use of control-circuit transistors that are directly triggered by light from the laser pulse being photographed. The ultrafast streaking camera is proving to be an extremely valuable tool for diagnosing, understanding, and developing our laser systems. Additional cameras are planned, and development work on an even faster camera is in progress.

An ideal detector for diagnosing the performance of Q-switched and mode-locked lasers²³ would have a time resolution of ≤ 1 psec with good sensitivity. The techniques commonly used for pulse detection suffer various inadequacies from the standpoint of diagnosing picosecond pulses. Fast-photodiode detectors²⁴ are small, simple, and rugged, but the overall time response of such systems can range from 300 psec up to ~ 1 nsec, depending on input-pulse wavelength. One can determine pulse durations on a time scale of picoseconds by using the technique of two-photon fluorescence (TPF)²⁵; however, this technique cannot provide information about the detailed shapes of short pulses. In addition, since TPF is a nonlinear technique, low-intensity information (e.g., satellite pulses) may be lost. The use of an electro-optical streaking camera

appears to be a promising technique for detailed study of laser pulses on a picosecond²⁶⁻²⁸ time scale.

Figure 19 shows a diagram of the ultrafast streaking camera built and used at LLL, and Fig. 20 is a photograph of the apparatus. The camera operates as follows. An incident pulse of light is suitably attenuated and strikes a diffuser placed in front of a 25- μ m slit. An $f/1.3$ lens images the slit onto the photocathode of a commercially available gated image-converter tube that contains a set of deflecting plates. Electrons emitted from the photocathode (characteristic emission time ~ 0.01 psec) during irradiation by the light pulse are accelerated by the electric field between the photocathode and the first grid, and then by the field between the grid and the anode. Electrode structure in the tube provides electrostatic focusing as the electrons travel through the anode aperture. A suitable voltage pulse applied to the deflecting electrodes causes the beam, in the image of the slit, to be swept along the phosphor screen on the back face of the tube.

The intensity of the blue light emitted by the phosphor depends on the electron current striking it; this current in turn depends on the intensity of light incident on the photocathode. Intensity-versus-time information in the incident pulse is thus transformed into blue-light intensity versus position along the fluorescent screen.

A second lens images this faint streak onto the photocathode of a high-gain, magnetically focused image intensifier. This commercially available device amplifies the intensity of the streak by a factor of $\sim 10^5$ so that it can be conveniently photographed.

Fig. 19. The LLL ultrafast streaking camera. Not shown is the optical delay system used for trigger timing.

Time Resolution

Several factors in the design and operation of the image-converter tube combine to influence the total time resolution of the camera.

The camera's technical resolution, τ_t , is determined by the limiting spatial resolution of the system and the streaking speed.

An additional limitation to the time resolution arises because electrons emitted from the photocathode have a range of kinetic energies. Electrons with different energies (velocities) require different amounts of time to travel the length of the tube and strike the phosphor. This limitation to the time resolution, denoted by τ_e is essentially given by²⁹

$$\tau_e = 2.34 \times 10^{-8} \frac{\sqrt{\Delta\epsilon}}{E} \text{ sec}, \quad (1)$$

where $\Delta\epsilon$ is the photoelectron energy spread in eV and E is the accelerating electric field in V/cm. Equation (1) shows that in order to reduce τ_e it is necessary to have the field E , between the

photocathode and the first grid (see Fig. 19), as large as possible. This is the crucial factor for picosecond resolution with the image-converter tube.

The width of the slit also affects the time resolution. If the slit is too wide for a given streaking speed, the time for the image of the slit to move its width can be a significant factor. In practice, the slit-width time spread τ_s is small and can usually be neglected.

Finally, the illumination level on the photocathode must be considered. If the illumination level is too high, the electron current from the photocathode can be large enough to distort the focusing and accelerating fields in the tube and to cause spreading of the beam by electrostatic forces. For this reason it is necessary to use a high-gain image intensifier so that the lowest possible level of illumination on the image-converter tube will produce a recordable image.

If τ_q is the duration of a laser pulse, then the width of the image recorded by the streaking camera, τ , will be given by

$$\tau^2 = \tau_q^2 + \tau_t^2 + \tau_e^2 + \tau_s^2. \quad (2)$$

Fig. 20. The ultrafast streaking camera.

Operation

The fast voltage pulses required for the gating and sweeping functions of the image-converter tube are produced by avalanche transistor circuits. The voltages are generated by direct optical triggering of one of the series string of avalanche transistors; this system is unique to the LLL camera. The top of the cover of the optically triggered transistor is removed and replaced with a small glass disk to protect the active element. Figure 21 shows one such transistor.

A portion of the input laser pulse is focused onto the trigger transistor chip; the rest of the pulse is optically delayed by an appropriate time interval and is directed to the streaking-camera input. The time jitter of the entire system is less than 100 psec (time jitter means the uncertainty as to when the camera actually triggers). This triggering system is superior to techniques using laser-triggered spark gaps employed in other such ultrafast cameras.²⁶⁻²⁸ These more conventional systems typically have a time jitter of ~ 1 nsec, and require the use of complicated multi-element pulse-delay devices to ensure photographing the pulse within this interval.²⁸

We have measured the streaking speed by photographing the train of subpulses produced when the output of a short-pulse ruby or neodymium-glass laser is passed through an etalon (i.e., an interferometer). The spacing of the etalon determines

Fig. 21. A transistor modified for direct optical triggering. This efficient triggering scheme is believed unique to the LLL camera.

the time separation of the subpulses. We can find the streak speed and nonlinearity by measuring the positions along the film of the subpulses. Figure 22 shows position-versus-time curves for the two streak speeds at which the camera has been operated. The

Fig. 22. Position versus time for the two streaking speeds at which the LLL camera has been used. The streaking sweep is nearly linear over a sizeable range. The faster speed is about 4×10^9 cm/sec, and the slower is about 2×10^9 cm/sec.

Table 4. Characteristics of the LLL ultrafast streaking camera

		Comment
Voltage on first grid (V)	2500	
Accelerating field (V/cm)	~3500	
Sweep speed (10^9 cm/sec)		
Slow sweep	2	Solid curve, Fig. 22
Fast sweep	4	Dashed curve, Fig. 22
Usable sweep length (mm)	40	Screen of image-converter tube
Spatial resolution (mm)	<0.24	On screen of image-converter tube, for fast sweep
Technical resolution, τ_t (psec)	6.0	For fast sweep
Electron-transit time spread, τ_e (psec)	7.0 ^a	Based on measurements in Ref. 28
Slit-width time spread, τ_s (psec)	1.2	For fast sweep
Overall camera resolution (psec)	~10.0	For fast sweep

^aEquation (1) predicts a value of 3.7 psec for τ_e . The discrepancy most probably results from non-uniformities in the electric field caused by the rather open grid structure in the present image-converter tube.

fast speed is approximately 4×10^9 cm/sec, and the slow speed is about 2×10^9 cm/sec.

The spatial resolution of the image-converter tube has been measured by streaking an image of the elements of a resolution chart. The chart was illuminated by 2-nsec pulses from a neodymium-glass laser. These data, together with the measured streaking speed, determine the technical resolution τ_t . Table 4 summarizes the characteristics of the camera.

Results

Figures 23 through 25 show representative streaking-camera photographs of the pulses from three different Laboratory lasers.

Figure 23 compares photographs of part of the pulse train from a mode-locked ruby oscillator, as obtained by the TPF technique²⁵ and by the streaking-camera method (at the slower streak speed — the solid curve in Fig. 22). The subpulses are 7 psec long

Fig. 23. Comparison of TPF and streaking-camera techniques for displaying a pulse train from a mode-locked ruby laser. Top: a TPF record of the pulse train. Bottom: a streak photograph of the same train. The subpulses are ~ 7 psec long and are separated by ~ 300 psec.

as measured from the TPF pattern, and are spaced 300 psec apart. The multiple pulses are clearly displayed on the streak record.

Figure 24 (top) shows the output of a mode-locked neodymium-glass oscillator as recorded by the streaking camera. Two pulses can be seen, a main pulse followed 25 psec later by a weaker, barely visible pulse. Figure 24 (bottom) shows another pulse train from this same oscillator; this time three equally intense subpulses separated by 25 psec can be seen. The duration τ of each pulse on the film is 15 psec. We can use Eq. (2) and Table 1 to find the true duration τ_g of each subpulse:

$$\tau_g^2 = \tau_t^2 - \tau_e^2 - \tau_s^2 = 225 - 36 - 49 = 144$$

$$\approx 139,$$

so $\tau_g \approx 12$ psec.

Finally, Fig. 25 is a streak photograph of a 2-nsec amplified pulse from a Q-switched neodymium-glass laser. Within the pulse envelope of ~ 2 nsec, structure is apparent with a period of 150 to 200 psec. In addition the entire sweep shows finer structure on a 10-to-20-psec time scale. A fast-photodiode record of the same pulse, recorded on an oscilloscope (resolution ~ 300 psec), shows simply a smooth pulse envelope without resolution of the structure.

Fig. 24. Streaking-camera records of two different pulse trains from a mode-locked neodymium-glass laser oscillator. In the top photo the second subpulse, following the main pulse by 25 psec, is very weak; in the bottom photo the two subpulses are of nearly equal strength. The duration of each subpulse is about 12 psec (see text).

Fig. 25. Streak photograph of a 2-nsec pulse from a neodymium-glass laser. Within the pulse envelope, structure with a 150-to-200 psec period can be seen; in addition, finer structure on a 10-to-20-psec time scale is apparent.

Our analysis of deactivation mechanisms and chemical reactions in HF lasers shows that the efficiency of present HF lasers should be in the range of a few percent. Our calculated efficiencies are lower than those predicted earlier and are in line with measured efficiencies. The calculations differ from those of other workers in that they treat molecular collisions as a strong competitor with photon emission in deactivating the vibrationally excited levels of HF. Our analysis of the reaction kinetics shows that it may be possible to achieve greater efficiencies by increasing the rate at which excited HF molecules are generated relative to the rate at which they are deactivated by collisions. We consider two ways of doing this: (1) raising the temperature to increase the reaction rates, and (2) exploiting a chain-branching reaction so that the free-radical intermediates necessary for producing excited HF will greatly increase in number in a very short time.

Introduction

Chemical lasers are expected to have great significance in both weapon applications and studies of the interaction of high-power laser pulses with matter, because large amounts of chemical energy (1 eV or more per molecule) are stored in vibrational modes and because a wide choice of reactants permits one to tune such lasers over a large spectral range. However, their potential has not been realized because of two obstacles. First, the stored energy is used inefficiently since collisional deactivation competes with photon emission in depopulating the excited states. Secondly, the gas density cannot be too high, otherwise the chemical processes that generate the excited species are slowed when excessive numbers of free-radical intermediates are lost in three-body collisions.

Unfortunately, some rather unrealistic predictions^{17,18} of chemical laser performance were made earlier. Experimental results have not fulfilled the expectations. Our studies of two- and three-level models of HF laser performance have provided realistic laser efficiencies that are much smaller than previous predictions because we properly treat collisional deactivation. Our analytic solutions of the chemical kinetics equations show that to achieve acceptable conversion efficiencies (photon energy out per total electrical energy expended in laser initiation) we must find a reaction in which chemical branching reactions can be exploited, so that for the short time required

for lasing, free-radical intermediates, and hence vibrationally excited HF molecules, will be generated at a much faster rate than they are consumed. If suitable reactions can be found, we should be able to achieve greatly enhanced laser output from HF chemical lasers.

The HF Laser

A chemical laser based on the $H_2 + F_2$ reaction operates as follows: (1) fluorine radicals are generated in the reactant gases by a flash of UV radiation or by an electric discharge, (2) these radicals react with H_2 to produce HF in an excited state, and (3) the excited HF molecules lose their energy by photon emission to produce lasing.

Excited HF molecules are produced chiefly in the second vibrational level, but some are produced in the first and third levels. (Some HF molecules are also produced in higher levels, but these do not contribute to lasing.) Lasing can occur by transitions from any of these levels to lower levels. In this work we treat the transitions from the second to the first vibrational level, i.e., the so-called *primary* transition.

The output pulse from an HF laser consists of contributions from all these transitions. The contributions can be separated using a monochromator. The contribution from the primary transition appears as a sharp peak at the beginning of the pulse. The contributions from the other (usually cascading) transitions (called the *secondary* transitions) make up the later portion of the pulse.

The Three-Level Model

A three-level model is the simplest analytical scheme that can describe the dynamics of those chemical lasers in which excitation occurs to levels higher than the first excited state. It enables us to determine how the shape of the output pulse is controlled by factors such as the rate of collisional deactivation, the rate of photon loss from the cavity, and the probabilities of chemically producing the lasing species in specific excited states.

Consider a population of excited molecules produced by ongoing chemical reactions. We will assume that the molecules can exist in three states: an upper level of excitation, a lower level of excitation, and a ground state. The population densities in these three levels will then be governed by the relative frequencies of collisional deactivation, probabilities of radiative transition, and rates of chemical formation.

Our solution of the equations describing the three-level molecule shows that the laser pulse shape and duration are significantly influenced by collisional deactivation. The upper and lower levels are strongly coupled by the radiation field, and upward and downward transitions are equally probable. However, molecular collisions favor the downward transition. Hence, for appreciable rates of collisional deactivation the population in the upper lasing level will be rapidly depleted by collisions, even during the short periods associated with lasing. (The population in the ground level builds up nearly linearly with time as some ground-state HF molecules are born in the chemical reaction and as excited molecules undergo collisional and radiative transitions. The power output of the laser, which is proportional to the product of the radiative-transition probability per unit time and the density of the population inversion, decreases as collisions populate the lower laser level at the expense of the upper until, at times of the order of the collisional deactivation times, the power output drops to zero. This time corresponds to the maximum energy output.)

Because collisional deactivation has so strong an effect, the lasing stops after only a small fraction of the excited molecules have formed. Although the chemical reactions continue to occur after the lasing, the depopulation of the excited state effectively takes place only collisionally and not by photon emission.

The solutions to our equations, when taken together with estimates of the rate at which the second excited state is populated by chemical reactions, suggest that the expected maximum output in the absence of branching is a few joules per liter (at room temperature) for HF lasers and that the small-signal gain is a few percent per centimeter.¹⁹

Chemical and Electrical Efficiencies

We define the chemical efficiency as the ratio of (1) the light energy emitted during the laser pulse to (2) the chemical energy produced and stored from discharge initiation to maximum laser output. If collisional effects were ignored, one would obtain predictions of chemical efficiencies of ≥ 10 percent for HF lasers. However, consideration of collisional effects reduces the calculated efficiencies to about a few percent, a result that is in agreement with experiments.²⁰

While our calculated chemical efficiency is the ratio of laser output from the primary transition to the chemical energy liberated during the interval between discharge initiation and the time of maximum laser output, the measured chemical efficiency in experiments is the ratio of the laser output from *primary and secondary* transitions to the *total chemical*

energy liberated in reactions producing HF. Since the reactions producing HF extend beyond the period of lasing, the liberated chemical energy in the experiments is larger than we consider in our calculations. However, since the experiment also measures a larger laser output than we consider, the net result is that our calculated chemical efficiency, though not the same as that measured, is in the same range.

A more pertinent measure of efficiency, from the standpoint of the economics of practical high-power lasers, is the *electrical-conversion efficiency*, i.e., the ratio of the laser output energy to the total electrical input energy provided for dissociation or gas heating. If chemical branching can be exploited, i.e., if free radicals can be produced rapidly and in excess by chemical reactions, the electrical energy required to produce an HF molecule can be made smaller than the chemical energy required to form that molecule, and hence electrical-conversion efficiencies will be greater than chemical efficiencies. Oravskii²¹ has shown that the output of such branching chemical lasers should be virtually independent of the amount of initiation energy expended.

In addition to branching, some improvement in both chemical and electrical efficiencies can be obtained by carefully choosing the reactants to minimize collisional deactivation rates. In HF lasers the rapid deactivation by HF-HF collisions limits this expected improvement to 1 to 10 times the performance of $H_2 + F_2$ systems. This situation can be improved by allowing collisional transfer of vibrational excitation to a nonreactive molecule that will then lase. (In fact, DF-CO₂ lasers are quite promising.)

Chemical Kinetics of HF Lasers

Table 3 shows important reactions in the production and destruction of H• and F• free-radical intermediates and excited HF molecules (HF*). Rate constants for these reactions are fairly well known. The first set of reactions in Table 3 represents laser initiation by either photodissociation or electron-collisional dissociation of F₂ molecules (e.g., by flash lamps or electrical discharge). A crude model of electrical discharge shows that the electron density at our experimental conditions saturates at about 10^{14} electrons per cm³, at which time the F• density will correspond to about 10^{16} F• free radicals per cm³. Thus there is a limit to the number of free radicals created in this initiation method.

The second set of reactions shows the production of excited HF from free radicals by two reactions: the cold reaction and the hot reaction, so named because the latter generates more heat. These reactions occur in chains, i.e., a free radical reacts to produce an HF* molecule and another free radical, and the process can

Table 3. Chemical reactions important to lasing in H_2 - F_2 reaction mixtures

Chain initiation ^a	$\begin{cases} h\nu_{uv} + F_2 \rightarrow F_2^* \rightarrow 2F\cdot \\ e^- + F_2 \rightarrow F_2^* \rightarrow 2F\cdot + e^- \\ \text{or} \\ e^- + F_2 \rightarrow F^- + F\cdot \end{cases}$ <p>(The kinetic energy of the e^- is less than the ionization potential of F_2.)</p>
Chain propagation ^b	$\begin{cases} F\cdot + H_2 \rightarrow HF^* + H\cdot \text{ "cold" reaction} \\ \Delta H = 32 \text{ kcal} \\ H\cdot + F_2 \rightarrow HF^{**} + F\cdot \text{ "hot" reaction} \\ \Delta H = 98 \text{ kcal} \end{cases}$
Chain termination by three-body recombination	$\begin{cases} 2F\cdot + M \rightarrow F_2 + M \\ 2H\cdot + M \rightarrow H_2 + M \\ H\cdot + F\cdot + M \rightarrow HF + M \end{cases}$
Chain branching	$\begin{cases} HF^{**} + F_2 \rightarrow HF + 2F\cdot \\ HF^* + H_2 \rightarrow H_2^* + HF \\ H_2^* + F_2 \rightarrow HF + H\cdot + F\cdot \end{cases}$

^aThe (*) denotes any state above the ground level.

^bThe (**) denotes any state above the fifth vibrational level.

be repeated time and again, like successive links in a chain, until the chain is terminated by the loss of a free radical. (A cold reaction followed by a hot reaction comprises a link in the chain.) The cold reaction, being the propagator of the lasing levels, is the important one for laser action.

The third set of equations are reactions that can terminate the chains, i.e., three-body recombination reactions that consume free radicals. (We specifically exclude termination at the surface of the laser cavity because diffusion times are longer than the duration of the lasing under most conditions of interest.

The last set of reactions are branching reactions, which, in producing two free radicals for each free radical that is consumed, end one chain and begin two more, hence giving the effect of chain branching.

Thermal explosions of H_2 - F_2 mixtures can be explained by reference only to the chain and chain-terminating reactions, and assuming thermal dissociation to generate the first free radicals needed to initiate the explosion. The energy generated by the hot reaction and three-body recombinations raises the temperature of the gas, causing an exponential increase in the reaction rates that leads to thermal runaway. However, because the hot reaction is slow and thermal explosions take a relatively long time, this mechanism is not of interest for producing short laser-light pulses.

Chain-branching reactions can take place rapidly, giving rise to a much faster increase in the number

of excited species than does thermal runaway. In a chain-branching explosion, free-radical densities increase exponentially without the temperature rising until times much later than are of concern for lasing.

One way to determine if chain branching is occurring in a chemical system is to measure the densities of $H\cdot$ and $F\cdot$ radicals after a brief reaction period. The ratio of these densities should be the same as the ratio of the rates of the cold and hot reactions. A deviation from this is taken to be evidence of branching.

The most optimistic estimates of the rate of chain branching for the $H_2 + F_2$ system turn out to be lower than the rates of collisional deactivation. Hence chain branching will not contribute effectively to lasing in this system.^{19,22} For other systems that produce HF there is hope of exploiting another form of branching — *material branching*, i.e., the chemical exchange of radicals not requiring excited-state participation. Such a process will likely occur with larger reaction cross sections than energetic branching, and hence may satisfy the criterion for fast branching.

Most chemical branching reactions are material branching reactions, e.g., the explosive oxidation of hydrogen to form water. In terms of HF lasers, we envision a reaction such as the following:

where the $R'H\cdot$ is generated in an initiation step. This equation displays the two processes required for a working chemical-branching laser system: the HF must be formed in an excited state, and one free radical must give rise to two or more free radicals. We have found that the $SiH_4 + N_2F_4$ system exhibits branching properties. However, we have not yet been able to show that the branching contributes to lasing.

Energetic branching reactions, another form of chemical branching reactions, are those that are mediated by excited molecules, e.g., the fourth set of equations in Table 3. In the $H_2 + F_2$ system, material branching does not occur, and chemical branching can only take place by energetic branching.

Using a crude model of the electrical discharge and recently measured $F\cdot$ recombination rates, we find that at room temperature three-body recombination overtakes the cold reaction at about 1/7 atm, corresponding to the experimentally observed "pressure turnover" in laser output at about this pressure.²⁰ Because the reaction rates increase exponentially with temperature while recombination rates decrease with temperature, one should be able to move the pressure turnover to higher pressure merely by operating at higher temperature. Such action should also lessen collisional-deactivation problems for HF lasers, provided that $T < 1000^\circ K$.

Of course fast branching will also circumvent the pressure-turnover problem by overwhelming the system with free radicals. One must be cautious, though, in making optimistic predictions on such a basis, because sufficient free-radical production will correspondingly

increase deactivation rates. ($F\cdot$ free radicals are more efficient than either H_2 or HF in deactivating excited HF .) As a consequence, one may enter a regime leading to branching explosion, but without any contribution to lasing.

EXPERIMENTS WITH HF CHEMICAL LASERS

We are surveying potential hydrogen donors and fluorine donors for use in HF lasers. In laser experiments, we have studied hydrogen donors including hydrogen gas, deuterium, inorganic hydrides, olefins, aromatics, and branched and straight-chain hydrocarbons. With NF_3 as the fluorine donor, we measured how lasing was affected by pressure, by mole ratio of hydrogen donor to fluorine donor, and by the addition of nitrogen and rare gases. Our results are in keeping with the theoretical treatment discussed in the preceding article (p. 23ff of this report and Ref. 19). They suggest that only with some form of branching chemical reaction will it be possible to develop chemical lasers with greater electrical-conversion efficiencies than CO or CO_2 lasers.

The first LLL experiments with the NF_3 - H_2 system showed that free-radical recombination sets an upper limit on the laser's efficiency and energy at pressures above 100 Torr. At lower pressures the laser's overall efficiency is low because the lasing HF molecules are produced only from fluorine radicals formed in the initiating electrical discharge and are not produced from chain reactions. The limit at high pressure can be removed if a chain-branching laser can be found that produces free radicals faster than the recombination reactions remove them. We are studying chemical systems in which chain branching might occur. We first made a relatively fast and easy survey of hydrogen donors with weakly bound hydrogen, but failed to find a suitable chain-branching lasing reaction. We concluded from this survey that branching can occur only if weakly bonded fluorine donors are used. We are about to begin this more difficult survey of fluorine donors with weakly bound fluorine.

One aim of these surveys is to find a reaction that "energetically" chain branches by producing HF or other products that are so strongly excited that they collide with reactant molecules, causing them to dissociate. A second aim is to find a "material" chain-branching reaction in which one free-radical reactant produces two free-radical products. We are developing experimental methods for the study of chain-branching chemical lasers.

Experimental Setup

At LLL, three basic systems have been used to initiate chemical reactions in lasers: flashlamp light in N_2F_4 - H_2 mixtures; electron beams in N_2F_4 - H_2 , NF_3 - H_2 , and IF_7 - H_2 mixtures^{23,24}; and electrical discharge. Flashlamp initiation is very inefficient, and is limited to chemical systems that absorb light of the wavelength emitted by the flashlamps. Electron-beam initiation has considerable promise, but equipment is expensive, cumbersome, and relatively fragile. We have used electric-discharge initiation in our survey because the system is simple to construct, durable, and readily controlled and monitored for energy input. Figures 8 and 9 show some of our equipment.

NF_3 - H_2 Experiments

Practically speaking, the power and energy of a HF chemical laser are limited by the efficiency and the operating pressure. To achieve sufficient output, chemical laser systems that have low efficiency must be driven by high-energy input pulses, and those that have to be operated at low pressure must be of large volume. We have examined in detail the effect of pressure on the performance of the NF_3 - H_2 system. This examination has indicated some of the physical processes that control laser performance and thereby limit laser efficiency and pressure.

For all mixtures of NF_3 and H_2 the total energy output of the laser increases with pressure at low pressures, reaches a maximum at a given pressure (identified as P_{max}), and then decreases with pressure at higher pressures. Figure 10 shows some typical results.²⁰ For systems with shorter initiation pulses, maximum laser energy is achieved at higher pressure. Maximum energy was achieved at a 5:1 mole ratio of NF_3 to H_2 . At this mole ratio the maximum chemical efficiency (based on the conversion of the energy from the $F\cdot + H_2 \rightarrow HF + H\cdot$ reaction into laser light) is about 1 percent and the maximum electrical efficiency (conversion of electrical energy input into laser light) is also about 1 percent.

We analyzed the laser output at 15.5 Torr and determined the placement in time, relative intensity, and duration of each lasing line. The transitions from

Fig. 8. Laser for studying HF lasing in different chemical systems (pulse length = 70 ± 10 nsec). The current-limiting resistor was varied to vary the discharge current. Values were 0, 25, 50, 100, and 235 Ω . The radius of curvature for the gold mirror is 10 m.

the second to first vibrational levels lase first and comprise more than 70 percent of the energy of lasing; they are followed by lasing in transitions from the first vibrational level to the ground level (20 percent of the energy), and by transitions from the third to second vibrational levels (10 percent of the energy).

The addition of krypton, nitrogen, and argon to a 50-Torr $\text{NF}_3\text{-H}_2$ mixture decreases the lase energy, whereas addition of helium increases the lase energy at low reactant pressures and decreases the lase energy at high reactant pressures.

Interestingly, for these $\text{NF}_3\text{-H}_2$ mixtures the slope of the plot of lase energy against pressure did not change as conditions were varied to take the system from the nonexploding to the exploding regimes. The amount of lase energy released seems not to be affected by whether or not a chemical explosion occurs. We also found that at low pressures (below P_{max}) the lase energy is proportional to the partial

Fig. 9. End window and part of the bank of pin electrodes at one end of the laser.

pressure of NF_3 (at low NF_3) and to the partial pressure of H_2 (at low H_2). The simplest set of reactions consistent with such results is:

Free-radical production:

Production of excited HF:

The low chemical efficiencies we measured are consistent with this set of reactions, since reaction (1) absorbs 59 kcal/mole, and reactions (2) and (3) should produce about 5 kcal/mole lase energy at best.

The decrease in efficiency and total lase energy at higher pressures could well be explained by free-radical recombination. The performance of the $\text{NF}_3\text{-H}_2\text{-He}$ laser is consistent with a reaction scheme in which radicals are destroyed by three-body recombinations at higher pressures.

Such a three-body-recombination explanation for the observed decrease in lase energy and efficiency at higher pressures is consistent with the explanation advanced by Semenov²⁵ to explain the explosion peninsula of certain gaseous explosive systems (see Fig. 11). Here, three-body recombinations give rise to the observed second explosion limit, generally at pressures of 10 to 100 Torr. As the pressure increases, the rate of reaction of a three-body process is proportional to the third power of the pressure, while the rate of a $\text{F}^\cdot + \text{RH}$ reaction (like reaction (2)) is proportional to the square of the pressure. Above the P_{max} the three-body process becomes dominant.

The results of our experiments on the effect of additives are also consistent with the explosion-peninsula explanation. Explosion limits are decreased by inert additives at high pressures because the additive provides additional third bodies for recombination. Explosion limits are increased by inert additives at low pressures because the additives slow the diffusion of free radicals to the wall. For the same reason addition of helium to $\text{NF}_3\text{-H}_2$ lase mixtures decreases the laser output energy at high pressures but enhances it at lower pressures.

An obvious way to overcome the problem of free-radical recombination is to attempt to achieve radical branching either by "energy branching" or by "material branching."

In energy branching, an excited HF molecule produced by one free radical (in a reaction of the same general type as reaction (2), above) subsequently reacts with another molecule to provide two free radicals, each of which can repeat the process. This

Fig. 10. Effect of pressure and mole ratio on lase energy for $\text{NF}_3\text{-H}_2$ mixtures. Maximum lase energy is attained at a mole ratio of about 5 to 1 and a pressure of about 65 Torr.

has the effect of ending one chain of reactions and beginning two more:

Generalized reaction (2):

Energy branching:

Here R and R' indicate inorganic or organic groups.

In material branching, free radicals produced in a chemical reaction without the participation of an excited state, participate in other reactions to produce excited HF and more free radicals (see the discussion on p. 25ff). The radical-branching step is as follows:

Fig. 11. Pressure-temperature diagram for an exploding gas.

Such radical branching should produce HF^* so rapidly that lasing would occur at high pressures in spite of the relatively slow three-body recombination reactions that inhibit HF formation.

Survey of Chemical Reactants

We are first varying the hydrogen donor while using the same fluorine donor (NF_3). Later we will use different fluorine donors. The survey of fluorine donors is being done last because many of the fluorine compounds are difficult to handle in the laboratory. The hydrogen donors studied include inorganic hydrides (SiH_4 , GeH_4 , AsH_3 , and B_2H_6), olefins and aromatics ($CH_3CH=CH_2$, $(CH_3)_2C=CH_2$, and xylene), cyclic aliphatic hydrocarbons (cyclohexane and cyclopropane), branched as well as straight-chain hydrocarbons ($(CH_3)_3CH$, $(CH_3)_4C$, *n*-butane, ethane, *n*-thane), and hydrogen and deuterium. The compounds were selected to include a wide range of

molecular structures and bond energies (104 kcal for hydrogen to 66.8 kcal for AsH_3).

Figure 12 shows the relationship of lase energy to pressure at the optimum mole ratio of NF_3 to hydrogen donor, for each of the materials studied. A P_{max} is attained in all cases where we carry the pressure sufficiently high, but the P_{max} is different for each of the compounds studied.

At 40 Torr, the maximum lase energy (achieved by varying the mole ratio) varies by a factor of about five for the materials investigated. Heavier aliphatic hydrocarbons yield the maximum lase energy, and inorganic hydrides produce the least lase energy. The optimum ratio of NF_3 to hydrogen donor is always greater than five and does not correlate with the stoichiometry of the complete reaction.

The lasing lines and pulse shapes show that when one uses hydrogen donors with weaker bonds, thus increasing the heat of reaction (4), some HF in higher vibrational levels is produced. However, such systems do not yield as much lase energy as do some of the

Fig. 12. The results of our survey of hydrogen donors for use in HF lasers. The numbers in parentheses give the $\text{NF}_3/\text{hydrogen-compound}$ mole ratio.

systems with lower heats of reaction. This shows that the lase energy is more strongly affected by the kinetics of the HF^* -producing reaction and deactivation processes than by the total chemical energy available.

Status of the Survey

We have surveyed hydrogen donors and are beginning to study fluorine donors. We are developing apparatus to study chain-branching lasers, should a

suitable chemical system be found. It is extremely difficult to handle chemical systems within a branching-chain-reaction regime since they explode or burn spontaneously during mixing. We are pursuing two approaches to overcoming this difficulty. One consists of using a consumable inhibitor in small quantities to delay the onset of the branching-chain explosion long enough to allow the reactants to be mixed and initiated. The other consists of mixing the reactants at a low temperature to prevent an explosion

and then rapidly heating the reactants into the branching-chain-explosion regime with a pulse from a CO_2 laser. The first approach could result in a

high-power chain-branching laser, while the second is being considered primarily as a means of studying chain-branching lasers.

SHORT-PULSE HF OSCILLATOR

We have developed a short-pulse HF oscillator and have sent the output of this oscillator through one stage of amplification.²⁶ SF_6 and CH_4 were the reactant gases used to produce the HF. The oscillator produces 5-nsec output pulses with a peak power of >0.1 MW (0.65 mJ). The amplifier, with a power gain of 15, was used to demonstrate how the oscillator pulse can be shaped by stages of amplification.

These techniques are being developed in anticipation that chain branching can be exploited to achieve high-efficiency HF lasers and thereby make practical the very powerful chemical-laser amplifier stages desirable for fusion research.

Subnanosecond pulses are desired for fusion experiments, but the input pulse to a multistage amplifier need not have so narrow a pulse width because sharpening will occur in sequential amplifier stages. We observed some sharpening to about a 3-nsec pulse width in the first stage of amplification. With additional stages of amplification some point will be reached where the pulse width will no longer be limited by the collisional bandwidth of the lines since nonlinear amplification will result in an effective line width being determined by the stimulated lifetime rather than by the time between collisions.²⁷

Figures 13 and 14 show the transverse-electric-discharge system used in our studies. It is similar to that used in the chemical survey mentioned in the preceding section (p. 19// of this report; see Figs. 8 and 9). In the case of the oscillator, however, the resistor-ballasted pins are arranged in a helix in two rows on opposite sides of the tube. The gold rear reflector of the oscillator cavity is interchangeable with a grating in order to permit scanning across the various lasing lines.

The SF_6 and CH_4 were mixed and allowed to flow through the laser tube at pressures ranging from 10 to 350 Torr. The ratio of SF_6 to CH_4 was varied from 1 to 8. At 125 Torr, the laser output energy increased as the mole ratio was increased from 1 to about 8, and then remained constant as that ratio was increased to about 18.

When the gold-plated rear reflector was used, the output from the laser was a double pulse similar to that observed earlier.²³

With a 75-kV input, a maximum laser output occurred at approximately 135 Torr. However, with a 90-kV input, the maximum occurred at about 160 Torr. This result indicates that a minimum

Fig. 13. Setup for the laser amplifier. See Fig. 8 for a circuit similar to the one used with this laser amplifier.

Fig. 14. Setup for the oscillator. The pins are arranged in two helices (anode and cathode, respectively) on opposite sides of the tube. This arrangement results in a symmetrical discharge, thus enhancing a single-mode laser pulse. The tube is about 30 cm long.

field-strength/pressure ratio of 700 V/cm per Torr is required for maximum energy output. The variation of the laser energy with increasing pressure showed a maximum, as observed in earlier work.²⁴ As the pressure was increased, the first peak continued to increase in energy and to narrow in time to a pulse width of 5 nsec, while the second peak decreased in amplitude until it had virtually disappeared at 200 Torr. We observed a pressure-related turnover in peak power at about 300 Torr.

The 5-nsec pulse from the oscillator was amplified, and the resultant 3-nsec pulse from the amplifier did not show the structure that was observed at the output of the oscillator. We are now optimizing the amplifier parameters.

Details of this work appear in Ref. 26.

SHORT-PULSE SOLID-STATE OSCILLATORS

An oscillator for laser-fusion experiments must produce a pulse that is single, with essentially no precursors, and that has diffraction-limited divergence and bandwidth-limited duration. These pulse characteristics are necessary to avoid damaging target pellets with precursor pulses and to reduce the likelihood of nonlinear optical effects, which can interfere with the simple exponential gain in the amplifiers and possibly cause optical damage – for example, via self-focusing.

Bandwidth-limited laser pulses, because of their lack of frequency or amplitude modulation, are highly desirable for laser-fusion research and several other applications. (A bandwidth-limited laser pulse may be generally defined as a pulse having a frequency bandwidth proportional to the inverse of its duration;

i.e., $\Delta\omega\Delta t \approx 1/2$.) The “smooth” unmodulated character of these well-defined pulses makes them particularly attractive for amplification to very high energies, which is one of the particular objectives of the LLL laser effort. The availability of such pulses may be essential for consistently and controllably achieving high energy densities.

We are evaluating several solid-state oscillator systems with respect to these required pulse characteristics. In the picosecond range, we are studying nonlinear optical effects and single-pulse generation with neodymium-glass, neodymium-YAG (yttrium-aluminum-garnet), and ruby oscillators. In the nanosecond range, we have developed a neodymium-YAG oscillator with pulse durations controllable from 0.5 to 2.5 nsec.

Fig. 14. Laser head with four linear flashlamps for the picosecond oscillator system.

Picosecond Oscillators

In one of the picosecond oscillator systems that we are evaluating, the laser head has four linear flashlamps arranged around the rod so that the cross section looks like a four-leaf clover. (This system was developed at Harvard University several years ago.) The lamps are enclosed in a cylindrical reflector (see Fig. 14). Pumping with the linear lamps is as uniform as with a helical system but two or three times more efficient. Also, the shape of the radial gain distribution is adjustable, as verified through both experiments and Monte Carlo calculations. A modification of this design has recently been applied to amplifiers operated up to saturation (0.2 J/cm^2 for neodymium-YAG, 2 J/cm^2 for ruby, and 3 J/cm^2 for neodymium-glass).

The oscillator cavity consists of two mirrors, a laser head, a saturable-absorber dye cell placed near the output reflector at Brewster's angle to the beam, and a pair of lenses of 40 cm focal length spaced about 80 cm apart in the center of the 1.5-m-long cavity (see Figs. 15 and 16). The saturable dye results in mode-locking, the production of a train of intense picosecond pulses separated by the time it takes light to make one round trip between the mirrors, which is about 6 nsec.³¹ The presence of the lenses is critical; without them this system produces spatially hot beams like other systems.

Diagnostics with high spatial and temporal resolution have helped to elucidate the role that the lenses play in this laser. The index of refraction of the laser glass is intensity-dependent. As the picosecond pulse begins to build up, the radial index profile becomes convex as in a lens, leading to self-intensification or self-focusing of the light. We believe that as the equivalent focal length of the laser rod approaches 40 cm (corresponding to an intensity of about 200 MW/cm^2 for this cavity) the point of

focus between the pair of lenses moves from the center toward one of the lenses, causing the nonparaxial components of the light to be nonresonant with the cavity and to overfill the cavity optics. Since only the axial rays survive the cavity resonance conditions, the result is a smooth spatial beam profile at an intensity of more than 5 GW/cm^2 over the $\sim 0.3\text{-cm}^2$ cross section of the beam.

Concurrent with this spatial development is the well-known effect of self-phase-modulation. If the refractive index of the laser glass depends on intensity, and intensity is time-dependent, then the refractive index is also time-dependent. When an electromagnetic wave travels through a medium whose refractive index is time-dependent, it acquires frequency sidebands that are shifted at the modulation frequency. Since the modulation frequency varies continuously through the laser pulse, the net result is a spreading of the spectrum.

Time-resolved measurements of the duration and spectrum of each pulse in the train reveal that the initially bandwidth-limited 1-psec pulse (at about 100 MW/cm^2) evolves into a 5-psec pulse with a bandwidth greater than 200 cm^{-1} , as shown in Fig. 17. Since this is wider than the amplifier-gain bandwidth, the pulse at this point is no longer amplified, so that the pulse height stabilizes at an intracavity pulse energy of about 40 mJ/pulse. A pulse train of 75 to 100 pulses is then obtained before the energy per pulse begins to fall as the energy available in the amplifier (a high of about 3 J/cm^2 in the excited state) is depleted. The envelopes of such pulse trains have half-widths typically greater than 1 μsec for a spacing of 10 nsec between pulses (see Fig. 18).

These same techniques failed to produce spatially uniform beams over large lasing cross sections from

Fig. 15. Schematic of the picosecond oscillator cavity and the single-pulse selector.

Fig. 16. Experimental setup of the neodymium-glass picosecond oscillator system.

neodymium-YAG and ruby mode-locked oscillators. With these systems, the near-field structure was not affected by the lenses in the cavity. This was not surprising, since the index nonlinearity in these crystalline materials is estimated to be only about one-tenth of that in glass. With or without lenses in the 1.5-m cavity, 1-cm-diam beams were completely broken up into spatial hot spots. This result led us to suspect that self-focusing was occurring but that it was causing the lasing material to appear to be a lens of very much longer focal length than for glass. These studies implied that only small-diameter (about 1 mm) beams in oscillators would be stable against breakup. We have since built both a ruby and a neodymium-YAG oscillator that produce relatively smooth lasing across a 1-mm diameter with about 1 mJ/pulse at picosecond durations. The absence of a strong nonlinearity implies that the bandwidth of the pulses should not spread, and no spreading is observed.

In addition to being temporally bandwidth-limited and spatially diffraction-limited, the pulses generated by a system oscillator must also be single, or free from precursors. Single pulses are typically selected from the oscillator pulse train by a system consisting of crossed Glan polarizers with a Pockels cell (a fast-acting electro-optic shutter) between them, as shown in Fig. 15. The Pockels cell is "opened" by means of a laser-triggered spark gap to allow transmission of a single mode-locked pulse; the remaining pulses are rejected by the crossed polarizers.³¹ However, any optical noise (precursor pulses) occurring before the main picosecond pulse is likewise transmitted. Although the precursor will be weaker than the main pulse at first, it will undergo a higher effective gain through any amplifier in which the main pulse saturates. By the time the precursor and main pulse reach the target, there will be a significant amount of energy in the precursor. This can completely alter the target-heating properties of the pulse, most likely for the worse.

Fig. 17. Sudden onset of self-phase-modulation in a glass amplifying system, an effect that very rapidly reduces the high signal gain. Top: Streak spectrogram showing the spectral development of a mode-locked laser pulse in a high-power neodymium-glass laser oscillator. Bottom: Simultaneous diode energy monitor showing pulse-height buildup. Both photographs have the same time scale.

Fig. 18. Pulse train envelope with a half-width of about 1 μsec and an interpulse spacing of 10 nsec.

We have studied the sources of interpulse optical noise in the pulse train by using various techniques: our new ultrafast streaking camera³² and conventional streak spectroscopy; measurements of pulse duration by the two-photon absorption-fluorescence (TPF)

technique³³; and high-speed photodiode-oscilloscope analysis. There are at least three sources of precursor satellite pulses. One is *feedback* from optical elements in the beam, to the extent of about 1 part in 10^5 resonantly within the cavity or about 1 part in 10^4 resonantly from components outside the cavity. Possible sources of feedback are dust, damage sites, optically triggered spark gaps, and dye-cell windows (if they are parallel to the cavity mirrors), all of which can be identified by the temporal position of the feedback pulse in the pulse train and the spectrum of the entire train.

A second source of precursor satellites is related to the *position of the saturable-absorber dye cell*. The principal problems are 1) the finite relaxation time of the high transmission and 2) the enhancement of one-way transmission of a noise pulse by the main pulse traveling in the opposite direction. We have found several ways to produce pulses that are single and satellite-free as seen on the streaking camera: for example, putting one dye cell in the optical center of the linear cavity or using nonresonant (curved) dye cells in contact with the highly reflective mirrors. Typical results with both single and multiple pulses are shown in Fig. 19. (Note that it is necessary to rely on the streaking camera because small satellites often do not have enough contrast to be seen in TPF displays.)

The third source of precursor satellites is related to the *transverse mode structure*. The output of a large-aperture oscillator tends to break up into multiple laser beams that are almost independent spatially. Because this breakup occurs early in the evolution of the pulse train, and because the unequal division of energy leads to different relative evolutions of the nonlinear effects in each part of the broken-up beam, the net wavefront of the entire beam consists of several almost-independent wavefronts. Worst of all, these almost-independent high-intensity regions do not necessarily reach their maximum intensities simultaneously. The conclusion is that a single transverse mode is not only desirable but *essential* if one desires a satellite-free single pulse from a combination of an oscillator and a single-pulse switch-out device.

In the near future, we plan to begin studying techniques for producing oscillators that will generate pulses with durations adjustable from 5 to 500 psec. Satellite-free, bandwidth-limited single pulses are still mandatory, as is the Gaussian-like spatial distribution.

Bandwidth-Limited Nanosecond Oscillator

We have developed a neodymium-YAG oscillator that is capable of directly producing a train of Q-switched, mode-locked, bandwidth-limited pulses³¹

Fig. 19. Typical results of single-pulse generation with a picosecond mode-locked oscillator. Top: Streaking-camera and TPF recordings of the output resulting from improper positioning of the saturable-absorber dye cell. (Although the satellite pulses are visible in this TPF pattern, they often cannot be seen because of poor contrast.) Bottom: Streaking-camera and TPF recordings of true single-pulse operation obtained with a specially constructed dye cell and repositioned components.

with durations of 0.5 to 2.5 nsec. Plans for using this type of oscillator to generate the input for the long-path laser are now in progress.³⁴

The key to the performance of the bandwidth-limited nanosecond oscillator is that it operates as a self-driven optical resonator. In such operation, spontaneous emission noise in the cavity is gated out symmetrically in time to a high-Q (low-loss) cavity state for the desired pulse duration and then allowed to build up in a low-Q cavity state by means of controlled-bandwidth feedback.

The principal elements of the oscillator system are shown in Fig. 20. Two multiple-faced, resonantly

reflecting etalons define the optical cavity. Within this cavity are the following elements: a gain medium (neodymium-YAG in this case), a polarizer, a quarter-wave plate, and a Pockels cell. The combination of polarizer, quarter-wave plate, and Pockels cell (PQP) is situated between the gain medium and the highly reflecting rear etalon.

The PQP elements are arranged so that the cavity is in a low-Q (lossy) configuration when the Pockels cell is biased at either its half-wave or its zero-retardation voltage. With the Pockels cell biased initially at its half-wave value, the optical cavity is switched by shorting the bias voltage to ground while the gain medium is at its maximum inversion. As the Pockels-cell retardation passes through its quarter-wave value, the transmission of the PQP combination reaches a maximum, and a noise pulse is allowed to pass through the rear of the cavity with minimal attenuation.

Since the Pockels-cell voltage is switched to zero within a few nanoseconds, and since the transmission characteristic of the PQP-etalon combination is symmetric in time, a large-amplitude noise pulse, which is shorter than the cavity and whose envelope is symmetric about its intensity maximum, is initiated. This pulse is then permitted to oscillate by maintaining a cavity-round-trip gain that is greater than unity when the cavity is in its residual low-Q state (i.e., when the bias voltage of the Pockels cell is at zero). Gain occurs in this instance because the retardation of the Pockels cell does not drop to exactly zero as the bias voltage is rapidly switched but remains at a residual value because of strain effects on the crystal.

This residual retardation value remains for several microseconds and allows some light transmission through the PQP combination. For large-enough inversion levels, this transmission will permit a pulse to build up in the cavity. The frequency content of the oscillating pulse is constrained by the narrow-band feedback properties of the resonantly reflecting etalons, which will allow oscillation for only a few of the possible axial modes of the cavity.

The concept of the self-driven oscillator has been demonstrated in a neodymium-YAG system containing a PQP element in an optical cavity defined by a three-element resonantly reflecting (66%) front etalon. The total energy input to a helical lamp head containing a 1/4- by 3-in., flat-ended, antireflection-coated rod was about 100 J; the resulting output energies were nominally 0.25 mJ/pulse for a 5-mJ mode-locked pulse train, as determined by a calibrated radiometer system.

The spectral content of the pulses was measured with a Fabry-Perot interferometer. In this case the output pulses consisted of two or three adjacent axial

Fig. 20. Schematic of the self-driven bandwidth-limited nanosecond oscillator.

Fig. 21. A typical pulse train from the bandwidth-limited nanosecond oscillator.

modes (limited by interferometer resolution) of the optical cavity, which was about 1 m long. Correspondingly, the pulses were 2.5 nsec in duration (full width at half-maximum), in agreement with the inverse relationship between duration and frequency bandwidth by which bandwidth-limited pulses are

defined. By changing the mode-selecting components in the oscillator and adjusting the inversion, the frequency bandwidth of the cavity feedback could be varied to turn the duration of the output pulses. With various combinations of flat reflectors and etalons, bandwidth-limited pulses of 0.5 to 2.5 nsec duration have been obtained with the present system. A typical pulse train is shown in Fig. 21. We can use the usual single-pulse selection techniques to switch out one of these pulses.

In summary, the operation of the system relies on the combination of the fast, time-symmetrical PQP switch and the narrow-band frequency-selecting elements in the cavity. This combination, when incorporated with the control of inversion, provides for independent determination of the time and frequency content of the pulse, so that we can obtain bandwidth-limited pulses over a narrow range of pulse durations.

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33. The technique of two-photon absorption fluorescence is described in the August 1969 monthly report (UCRL-50000-69-8).
34. Plasma production with the long-path neodymium-glass disk laser is discussed in the November 1971 monthly report (UCRL-50000-71-11).