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STIMULATED VACUUM-ULTRAVIOLET EMISSION FROM HIGH-PRESSURE XENON EXCITED BY PULSED HIGH-ENERGY ELECTRON BEAMS

Most experimental studies of the continuum emission by the rare gases in the vacuum-ultraviolet have been done at gas pressures of less than 20 psi.¹⁴ The gases have usually been excited by electrical discharge or microwaves, although some recent studies have used high-energy charged particles. Current interest in the vacuum-UV emission from the rare gases stems from the possibility that these elements can undergo high-power vacuum-UV lasing.¹⁵⁻¹⁷ Since little information is available on the emission from high-pressure rare gases excited by high-current, high-energy electron beams, we have studied the emission characteristics of xenon under those conditions.

Experimental Apparatus

Our experimental apparatus, shown schematically in Fig. 9, consisted of a source of electrons, electron-beam monitors (not shown), a gas cell capable of holding pressures up to 1000 psi, and optical sensors for measuring the temporal, spatial, and spectral characteristics of the emission.

We used two electron-beam guns: one produces 2-MeV electrons in pulses having durations of 50 nsec FWHM (full width at half maximum), and the other produces 0.6-MeV electrons in pulses having durations of less than 2 nsec FWHM. Both generate electron currents up to 7000 A. The entrance window for the electron beams was a piece of thin (0.013 cm) stainless steel, which was supported by a perforated steel plate to minimize bulging due to the high gas pressure.

The central volume of the gas was viewed through 2-mm-thick LiF or sapphire windows. The UV windows were mounted at the outer ends of 10-cm-long line-of-sight pipes that extended from the central volume of the gas cell. Two of the windows were perpendicular to the axis of the electron beam, and a third faced directly along the axis. The line-of-sight pipes were filled with xenon gas at the same pressure as in the central volume of the gas cell. Collimators protected the UV windows from stray electrons and x-rays, which could cause the windows to become opaque to UV radiation.

Three different sensors were used for observing the vacuum-UV radiation. To determine the spatial characteristics of the emission, we used a lensless Nikonos II underwater camera with Kodak type 101-10 UV-sensitive film. To obtain spectral information we used, in addition to this camera, a

Fig. 9. Apparatus for studying the vacuum-ultraviolet emission from high-pressure xenon gas.

McPherson Model 235 vacuum spectrograph, which has a dispersion of 17 Å/mm. To measure the time dependence of the emission, we used an ITT 4115 planar photodiode having a 2.5-mm-thick MgF_2 window and a CsTe photocathode. A vacuum of less than 10^{-3} Torr was maintained in the line-of-sight pipe between the UV windows and these sensors.

The xenon gas was Reagent grade; the concentrations of impurities were 12 ppm H_2 , 3 ppm O_2 , 6 ppm N_2 , and about 100 ppm Kr. The gas pressure ranged from 15 to 450 psi. We made no attempt to maintain uniform gas temperature, but we found that our results were not reproducible unless we kept the entrance window cool by blowing precooled dry nitrogen on it.

Results

Measurements made with the vacuum spectrograph showed that xenon emits a nearly symmetrical band

of 150 Å FWHM (full width at half density) centered at 1700 Å. This spectrum was independent of pressure, although we observed some narrowing of the band when we decreased the current density of the electron beam (but kept the xenon pressure constant). No other emission was observed out to 7000 Å.

The 0.6-MeV electron pulse has a FWHM of less than 2 nsec. Except at very high pressures, this is so narrow that it approximates an impulse. Consequently, it can be used to determine the lifetimes of some of the species it creates. Figure 10 shows the variation with time of the 1700 Å radiation emitted by xenon at different pressures when the gas is excited by the short pulse of electrons. We found that the time dependence of the intensity I of the emission pulse could be approximated by the difference of two exponentials,

$$I(t) = I_0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right), \quad (1)$$

where λ_1 and λ_2 are the time constants for the decay and the buildup, respectively, of the emission pulse. We numerically fitted the observed pulses to Eq. (1), and determined that, for pressures less than 150 psi, λ_2 varied as the 1.7 power of the pressure P . Above 150 psi the pressure dependence could not be accurately determined because the buildup time, $1/\lambda_2$, approaches the width of the 0.6-MeV excitation pulse. We analyzed the decay for times up to 40 nsec after peak intensity and found it to be essentially independent of pressure: $1/\lambda_1 \approx 2 \times 10^{-8}$ sec.

When the xenon was excited by the wide (50 nsec FWHM) 2-MeV electron pulse, we found that the peak UV output intensity varied as $P^{1.5}$ and that the FWHM of the emission pulse varied as $P^{-0.5}$. Thus, the UV energy output, which is given by the product of the peak intensity and the FWHM of the pulse, varied linearly with pressure. This pressure dependence was expected, since the amount of electron energy deposited in the gas increases linearly with pressure.

We also found that the UV emission intensity of xenon at 150 psi varied linearly with the current of the electron beam between 130 and 3300 Å. The pulse shape did not change with the change in current.

To estimate the efficiency with which xenon converts the kinetic energy of the electrons into UV photons, we calculated the photon output from photodiode measurements and estimated the energy deposited in the gas from the characteristics of the electron beams. The energy W deposited in the gas was estimated from the approximation

$$W \approx I \tau \frac{dE}{dx} \rho x, \quad (2)$$

Fig. 10. The intensity of spontaneous 1700-Å emission from xenon plotted as a function of time at different pressures. The gas was excited by a 2-nsec (FWHM) burst of 0.6-MeV electrons.

where I is the electron beam current, τ is the FWHM of the electron beam, dE/dx is the rate of electron energy loss by collisions, ρ is the density of the gas, and x is the length along the beam axis that is observed through the defining collimators.

Using Eq. (2) we calculated that the efficiency of conversion of electron energy to UV radiation in xenon at 250 psi was about 20% in our experiments. This corresponds, for example, to a peak UV power output of about 300 MW and a UV photon density of about 1 J/cm^3 . Our results compare favorably with those of Hurst *et al.*,¹⁸ who measured the efficiency with which the rare gases at low pressures are excited by protons.

Our overall results are consistent with the kinetic model that predicts that excited xenon atoms and ions are de-excited through the formation of excited

diatomic molecules which subsequently decay through photon emission. The excited dimers are formed in a three-body reaction; the rate of this reaction should be proportional to P^2 . We observed approximately this pressure dependence in the buildup of UV emission.

Our results further show that the excited molecules decay radiatively with a mean lifetime of about 2×10^{-8} sec. In most of our measurements with the 0.6-MeV electron pulse at pressures greater than 200 psi, a very fast UV component — with a mean lifetime on the order of 2×10^{-9} sec — was also present. This component followed the electron excitation pulse. We concluded that it was evidence of stimulated emission.

A CYLINDRICAL, RING-ELECTRODE POCKELS CELL FOR ISOLATING FAST PULSES OF LASER LIGHT

Many laser applications require single, sharp laser-light pulses of very short (subnanosecond) duration. Such pulses can deliver their energy to a target before the energy can be dissipated by explosion or thermal conduction. Electro-optical devices such as ultrafast Pockels-cell shutters are normally used to extract these short pulses from laser beams. We have developed a Pockels cell that offers an order-of-magnitude improvement in pulse quality over previous designs.

In a Pockels cell, a voltage applied to the crystalline material that makes up the cell will cause the crystal to become birefringent — that is, the crystal will separate an incoming beam of polarized light into two components that propagate with different velocities. The two components recombine in such a manner as to rotate the plane of polarization of the beam as it exits the material. The amount of this rotation depends on the applied voltage. By using a calcite prism or Brewster-angle plate as an analyzer, the rotated beam can be extracted from the rest of the beam. (The analyzer is oriented to pass light whose plane of polarization has been rotated 90° ; the Pockels cell is most efficient at this rotation.) Thus, by applying very short, sharp electrical pulses of the proper voltage to a Pockels cell located between a polarizer and an analyzer, one can extract very short, sharp light pulses from a laser beam.

The properties of a Pockels cell that are critical to its performance are the voltage required to produce the 90° rotation; the uniformity (across the diameter of the light beam) with which the cell transmits and rotates the beam; the losses in the cell due to reflection from the faces, absorption in the crystal, and birefringence due to stress in the crystal; and the speed

To support this conclusion, we made additional measurements with the 2-MeV beam using mirrors to form an optical cavity (see Fig. 9). To make alignment less critical, the mirrors were slightly curved (1-m radius).

In our experiments with the cavity, the previously observed 150-Å-wide emission band centered at 1700 Å narrowed to a sharp emission line about 17 Å wide and centered at 1716 Å. This gain narrowing indicates that stimulated emission occurred. Pinhole-camera photographs showed a high-intensity, well-defined beam spot along the axis of the optical cavity (perpendicular to the electron beam) and the FWHM of the photodiode output narrowed from 50 nsec to about 3 nsec.

Fig. 11. Cylindrical, ring-electrode (CRE) Pockels cell inside a Lucite sleeve. The cell is a crystal of KD_2PO_4 .

with which the optical signal can be affected. Our efforts to develop an improved Pockels cell were directed specifically toward maximizing the speed of response and minimizing transmission nonuniformities in the cell, while at the same time developing techniques for accurately monitoring the fast light pulses and determining the quality of the cell material.

Our new Pockels cell is made of potassium dideuterium phosphate (KD_2PO_4). This is the deuterated isomorph of potassium dihydrogen phosphate (KH_2PO_4), which has been produced commercially since the 1940's. KD_2PO_4 is a uniaxial crystal; i.e., it has only one axis of optical symmetry. When a wave of polarized light from an yttrium-aluminum-garnet (YAG) laser, which has a wavelength of $1.06\text{ }\mu\text{m}$, is propagating along this axis, about 10 kV must be applied to the crystal (also along the optical axis) to obtain maximum transmission through an analyzer oriented to pass light rotated 90° to that entering the crystal.

The crystal is cylindrical, 12 mm in diameter and 25 mm long; the axis of the cylinder approximately coincides with the optical axis of the crystal. The two electrodes through which the voltage is applied are 8-mm -wide brass rings placed around the ends of the cylinder (see Fig. 11). The assembly is enclosed in a Lucite sleeve to insulate the cell from air currents and

to protect experimenters against the high voltage. This cylindrical, ring-electrode (CRE) design represents a substantial advance in the technology of Pockels cells with respect to both switching speed and the uniformity of rotation and transmission of light.

In assembling such a cell, one must be careful to avoid straining the crystal, which would alter its optical properties by causing stress birefringence. The electrodes were machined to a tolerance of about 0.001 in. and carefully fitted over the ends of the crystal, and all soldering necessary to attach cables to the electrodes was done before the electrodes were placed on the crystal. Having taken these precautions, we found that, after the electrodes were attached, the transmission pattern of the cell between crossed polarizers was identical to that of the original crystal blank.

For the cell to have an optimum time response, the capacitance between the electrodes and the inductance of the cable connecting them must be minimized. The cylindrical electrode geometry represents a minimum-capacitance configuration, and, with the inductance of the connecting cable minimized by placing the exposed cable as close as possible to the surface of the cylinder, the switching speed of the cell is optimal.

The performance of a cell of this kind has been treated theoretically by Vitkov,¹⁹ who considered a

Fig. 12. Experimental arrangement for using a fast Pockels cell to obtain subnanosecond laser pulses. The polarizer and the analyzer are crossed 90° to each other, so that the analyzer passes light only when the Pockels cell is receiving a 10-kV pulse from the spark gap, briefly rotating a component of the beam by 90° . The spark gap is triggered by the pulse from the laser. The two oscilloscope traces show the pulse extracted by the Pockels cell and the remainder of the longer pulse generated by the laser.

rectangular cell with electrodes attached as bands around the ends of the cell. For our cell, conversion of his results to cylindrical coordinates yields a predicted nonuniformity in rotation of about 2.2% between the center and the edge of the crystal. We have checked this prediction experimentally by placing a CRE cell between polarizers and observing the variation in the voltage required to rotate a light beam 90°. The nonuniformity in rotation was about 1.5%, which agrees well with the theoretical prediction. (A rotational nonuniformity in this range implies a 0.02% nonuniformity in the amount of light transmitted across the aperture of the cell.)

With the same experimental setup, we have also checked the extinction ratio (the ratio of the intensity of light transmitted by the analyzer with the voltage applied to the Pockels cell to that transmitted with the voltage off) over the entire aperture for two different CRE cells. We found it to be greater than 1500:1.

These results represent order-of-magnitude improvements over commercially available KD_2PO_4 cells, which typically have rotational nonuniformities of about 13% and extinction ratios of about 200:1 over the same aperture.

Figure 12 illustrates the experimental use of a fast Pockels cell for obtaining subnanosecond laser pulses. A 30-nsec pulse is generated by a Q-switched YAG laser oscillator. When sufficiently intense light is focused onto the spark gap, it generates an electrical signal of 0.5-nsec halfwidth, and this signal is impressed across the Pockels cell. An optical signal, whose halfwidth is determined by the response of the Pockels cell to the electrical pulse, is then passed by the analyzer. After correcting the observed optical signal for the response time of the detecting system, we conservatively estimate that the halfwidth of the "chopped" light pulse is less than 0.7 nsec. We expect that this light pulse can be made even shorter by shortening the electrical driving pulse.

X-RAY CONVERSION EFFICIENCIES OF LASER-PRODUCED PLASMAS

A short, high-energy pulse of laser light focused on a target (usually of metal) can convert a small portion of the target to a hot, dense plasma that emits x rays. If the process by which laser energy is converted to x rays can be made efficient enough, laser-produced plasmas could be useful in applications that require a sharp burst of x rays — e.g., flash x-ray photography or simulation of the x-ray output of a nuclear explosive.

We have completed a survey of the x-ray yields and conversion efficiencies of plasmas produced when a pulse of light from the Plasma X neodymium-glass infrared laser²⁰ is focused on targets of various materials. As well as providing information on the possible use of laser-produced plasmas as a source of short-pulse x rays, the results of the survey also provide a test of theoretical calculations based on a hydrodynamic radiation code.

The experimental setup is shown schematically in Fig. 13. The energy output of the laser, which was measured by the photodiode, averaged about 2 J, and the pulse width was about 100 psec. The laser beam entered a chamber evacuated to a pressure of about 10^{-4} Torr and was focused on a target to a spot about 100 μ m in diameter.

We considered several different detectors for measuring the x radiation emitted by the resulting plasma, including calorimeters, solid-state detectors, and ionization chambers. An ionization chamber was judged to be the most suitable for our purposes because it is easy to calibrate and because its sensitivity

in its normal operating region is nearly independent of the energy of the x rays, so that the interpretation of the data obtained does not depend on the energy spectrum of the x rays.

The ionization chamber is a flowing-gas counter (90% argon, 10% methane). X rays enter the chamber through a 1/2-in.-diam window that is offset from the high-voltage electrode in the center of the chamber.

The lower limit of the x-ray energies that the counter can respond to is determined by the window material. We used two materials, 1-mil-thick Mylar and 0.6-mil-thick beryllium, and made measurements on the various targets with each window. The lowest energies at which these windows transmit at least half of the x radiation are 1.3 keV for the beryllium and 2.5 keV for the Mylar.

The upper limit of the counter's energy response is determined by its length. We arbitrarily selected a length (36 cm) that corresponds to two mean free paths for 10-keV x rays in argon; a detector of this length can capture 87% of the 10-keV x rays.

We calibrated the counter both with a known radiation source, ^{238}Pu (α particles), and with voltage pulses of known characteristics. Both methods lead us to conclude that the counter's accuracy in terms of absolute charge collection is at least $\pm 10\%$.

Figure 14 shows our results for the conversion efficiency (the ratio of x-ray energy to laser energy) as a function of the atomic number of the target material. The polyethylene data are plotted at atomic number 3, which is approximately the average atomic

Fig. 13. Experimental arrangement for measuring the x-ray yields and conversion efficiencies of laser-produced plasmas. The basic design of the ionization chamber is also shown.

number of CH_2 . With the high- Z targets (having an atomic number of at least 40) the conversion efficiency is about 10^{-4} for the $>2.5\text{-keV}$ x rays that pass through the Mylar window and about 10^{-3} for the $>1.3\text{-keV}$ x rays that pass through the beryllium window.

For sulfur ($Z = 16$), this order-of-magnitude difference in conversion efficiency is not observed. This indicates that the sulfur target is emitting relatively few x rays with energies between 1.3 and 2.5 keV. The K x rays of atomic sulfur have energies near 2.4 keV, and the transmissions of these x rays by the two windows actually differ by only about a factor of 2. Thus, the sulfur data suggest that most of the sulfur x rays above ~ 1.3 keV are emitted in K x-ray lines. The same conclusion can be drawn from the data on aluminum ($Z = 13$), since the ratio of the transmissions of the two window materials for the aluminum atomic K x rays (having energies of about 1.4 keV) is roughly the same as the ratio of the aluminum target yields — namely, about a factor of 10.

Since the x-ray line radiation from the carbon and CH_2 targets is of too low an energy to pass through either window material, the transmitted radiation must consist of continuum x radiation, which is due to recombination processes or bremsstrahlung or both.

Assuming that the electrons in the plasma have a well-defined temperature, we have inferred temperatures from the carbon and CH_2 data of about 0.4 and 0.2 keV, respectively.

We used the MONTE-WAZER code to try to theoretically reproduce some of the experimental results. In this code, the populations of the bound states of the target elements are not assumed to be in local thermodynamic equilibrium. This means that both the equation of state and the opacity of the plasma are more complicated than in equilibrium problems.

Our calculations for a gold target, in which it was assumed that all of the laser energy was absorbed by the target, indicated that about 30% of the energy is emitted from the front surface of the target in the form of x rays. The x-ray energy below 1 keV should account for 85% of the total x rays emitted. In addition, we predicted discrete line radiation at energies of 2.65 and 3.55 keV; the line at 2.65 keV should be about five times stronger than the one at 3.55 keV. (This agrees fairly well with observations reported previously, which show line radiation at an energy of about 2.4 keV. MONTE-WAZER uses an approximate procedure to compute line energies, so the difference between 2.4 and 2.65 keV is not significant.)

We calculated further that the total energy emitted from the gold target in the form of >2 -keV x rays should be 2.6% of the absorbed laser energy. This result is more than an order of magnitude greater than the value inferred from the x-ray yield measurements

(Fig. 14 shows that the conversion efficiency for a gold target with a beryllium window is about 6×10^{-4}). Additional investigations are being carried out in an attempt to understand the discrepancies between the computed and the experimental results.

Fig. 14. X-ray conversion efficiency of the plasma produced when the Plasma X laser beam strikes targets of various atomic numbers. Isotropic x-ray emission is assumed. The beryllium window transmits x rays with energies above ~ 1.3 keV; the Mylar window transmits x rays with energies above ~ 2.5 keV.

In high-powered, high-gain laser systems, the amplifier stages must be optically isolated from each other and from the target of the laser beam so that spontaneous emission from the amplifier stages and reflections from the target do not travel back through the amplifiers. Inadequate isolation results in depumping in the amplifier chain due to premature lasing (prelasing) and in damage to the laser oscillator and the other optical components due to high-intensity reflections from the target. As part of our continuing investigation of isolators for both gas and solid lasers, we have studied a potential bleachable isolator system based on sulfur hexafluoride (SF_6) as the saturable absorber for CO_2 laser systems, and are building several glass isolators based on the Faraday effect for solid lasers.

Bleachable Isolators for CO_2 Lasers

A bleachable isolator system for CO_2 lasers must absorb enough of the low-intensity spontaneous emission at all CO_2 lasing frequencies to prevent prelasing in the amplifier chain, and then it must become transparent, or bleach, with minimum consumption of laser energy when the desired high-intensity laser pulse passes through the system. This bleaching occurs when the populations of the upper and lower vibrational-rotational states of the absorber are equal. Absorption and stimulated emission occur at equal rates in such a system; it is transparent at the frequency of the transition between the states.¹³

We recently tested a bleachable isolator system that uses SF_6 as the saturable isolator and 1,1-dichloro-2,2-difluoroethylene ($\text{CF}_2 = \text{CCl}_2$) and dichlorodifluoromethane (CF_2Cl_2) to absorb those CO_2 frequencies not absorbed by the SF_6 . However, its transmission after bleaching was too low because large amounts of the two nonbleaching absorbers had to be used to sufficiently attenuate (by more than 95%) all of the CO_2 laser lines.

Figure 19 shows the frequencies of the molecular transitions $00^0_1 \rightarrow 10^0_0$ and $00^0_1 \rightarrow 02^0_0$ in CO_2 . These two transitions, at 10.4 and 9.4 μm , respectively, are the CO_2 laser transitions. The high-gain *P*-branch lines of the 10.4- μm band are on the left, with successively lower-gain branches to the right. The figure shows a rotational distribution that peaks near $P(20)$, which represents a system near room temperature. The intensity distribution is only

approximate; it is meant to show all the lines that should be attenuated by the isolator system.

With the exception of heated CO_2 ,¹⁴ no single gas can be expected to absorb strongly at all CO_2 laser frequencies and bleach at the frequencies expected from a mode-locked CO_2 laser: the $P(16)$ to $P(22)$ lines, usually $P(18)$ or $P(20)$, of the 10.4- μm band. Gases such as SF_6 , boron trichloride (BCl_3), phosphorus pentafluoride (PF_5), vinyl chloride, and propylene absorb and bleach at these frequencies, and many other gases absorb at CO_2 laser frequencies and will probably bleach.

We tested an isolator system based on SF_6 as the bleachable absorber and $\text{CF}_2 = \text{CCl}_2$ and CF_2Cl_2 as "sideband" absorbers. Figure 20 shows the absorptions of these gases in the CO_2 laser-line region. The SF_6 at 20 Torr-cm (pressure times path length) absorbs from $P(10)$ to $P(30)$ and nearly completely absorbs from $P(12)$ to $P(26)$ of the 10.4- μm band. CF_2Cl_2 at 300 Torr-cm absorbs the $P(30)$ and higher-*P* lines of the 10.4- μm band and the *R*-branch lines of the 9.4- μm band. The 2000 Torr-cm of $\text{CF}_2 = \text{CCl}_2$ absorbs the *P*-branch lines of the 9.4- μm band and most of the *R*-branch lines of the 10.4- μm band. To absorb the low *R*- and *P*-branch lines of the 10.4- μm band, we increased the amount of $\text{CF}_2 = \text{CCl}_2$ to 4000 Torr-cm in our test.

Fig. 19. Frequencies of the $00^0_1 \rightarrow 10^0_0$ and $00^0_1 \rightarrow 02^0_0$ transitions in CO_2 . These are vibration-rotation transitions, in which the rotational quantum number J changes by +1 or -1 in going from the initial vibration-rotation state to the final state. Transitions in which J changes by +1 are *P*-branch lines, and those in which J changes by -1 are *R*-branch lines.

Fig. 20. Absorption of three isolator-system gases in the CO_2 laser frequency region.

We tested this system of gases at two laser power levels. To test its transmission at low signal intensities we used a commercial continuous-wave CO_2 laser equipped with a grating so that it could be operated on most of the lines shown in Fig. 19. Its output power could be adjusted from 1 to 40 W for the high-gain lines of each branch, but we generally kept the power below 3 W when testing low-signal transmission. To test the high-signal transmission we used the LLL TEA laser,¹⁵ which has an output power of about 4×10^6 W/cm². We examined the output of the TEA laser with a monochromator and found that more than 90% of the laser output energy occurred at the $P(16)$, $P(18)$, $P(20)$, and $P(22)$ lines near $10.6 \mu\text{m}$.

The absorber cells are 20 cm long and have NaCl Brewster-angle windows and a side-arm trap for

freezing out the absorber gas. We determine the transmittance of the system with all gases removed and with any combination of the gases in the laser beam.

The test gas system at low signal intensity absorbed all of the CO_2 lines except about 2 to 3% of the $P(6)$ to $P(10)$ lines of the $10.4\text{-}\mu\text{m}$ band. With the high-intensity pulse, the transmission of this system was only 35%; with the SF_6 removed it was still only 37%. We obtained approximately the same transmission (with the SF_6 removed) at the $P(18)$ line with the low-power laser. The transmission of each of the individual gases for the high-intensity pulse was 94% for SF_6 , 94% for CF_2Cl_2 , and 40% for $\text{CF}_2 = \text{CCl}_2$. The accuracy of this transmission data is about $\pm 2\%$.

The transmission of the system for the high-power pulse could be increased to nearly the value of the SF_6 alone by reducing the amount of the other absorbers, especially the $\text{CF}_2 = \text{CCl}_2$. However, this would increase the low-power transmission of the low P - and R -branch lines of the $10.4\text{-}\mu\text{m}$ band to nearly 100%. Compounds with absorption bands centered near $10.4\text{-}\mu\text{m}$ could attenuate these lines; we are now evaluating CH_3CF_3 and $\text{CF}_3\text{CF}_2\text{Cl}$.

The narrow width of the SF_6 absorption band (about 10 cm^{-1} at 20 Torr-cm) is the major problem of the system. Most infrared bands do not have sharp absorption edges, and finding compounds to precisely match the areas of CO_2 emission not absorbed by SF_6 is difficult. A better solution would be to use a saturable absorber gas that absorbed all or nearly all of the P -branch lines of the $10.4\text{-}\mu\text{m}$ band and possibly some of the R -branch lines. Both BCl_3 and PF_5 have wider bandwidths than SF_6 . The PF_5 absorption maximum is centered near the $P(20)$ line, while BCl_3 has a band from $^{11}\text{BCl}_3$ (which constitutes about 81% of BCl_3) that absorbs the P -branch lines and a band from $^{10}\text{BCl}_3$ (about 19% of BCl_3) that absorbs the R -branch lines. We will study systems based on these two compounds and on other saturable absorbers with broad bands.

Faraday Isolators for Glass Lasers

Our efforts on isolators for solid lasers are concentrated on investigating and applying high-durability materials for Faraday-effect isolators. We then design the hardware for providing the magnetic field to complement this material.

In a Faraday-effect material, a magnetic field induces a rotation of the optical plane of polarization. This rotation is proportional to the Verdet constant (rotation per unit path length and field strength) of the material, the magnitude of the magnetic field, and the length of the material in the magnetic field. Also to be considered in the design of Faraday isolators are

the optical damage threshold and the optical distortion of the material.

Our present efforts are directed toward isolators of three aperture diameters: 16, 25, and 50 mm (70-mm commercial units are available but are very expensive). For the 16-mm size we have found that the presently used Shott SF6 glass has a limited life due to optical damage, so we are replacing it with Owens-Illinois EY1, a laser glass doped with 40% terbium. For both the 25-mm rotator, which is already built, and the 50-mm unit, which is now being fabricated, we are using Owens-Illinois ED-2 neodymium-doped laser glass because, in addition to its high optical quality, it has the same damage threshold as the laser amplifier rods. However, this material has a low Verdet constant (0.004 to 0.005 min/Oe-cm) and consequently requires magnetic fields in the tens-of-kilogauss range

for a length of 150 to 200 mm. Although this length of glass in the beam path could be detrimental (because nonlinear effects such as self-focusing can occur), we prefer the ED-2 glass to materials that have higher Verdet constants but lower durability.

Implicit in the design of high-quality Faraday isolators is the design or availability of high-quality polarizer-analyzer units. We have used commercially available calcite and quartz prisms, but calcite prisms are not available for aperture diameters larger than about 25 mm and quartz prisms give insufficient angular discrimination when the distance between the isolator and the amplifier is less than about 10 ft. Therefore, we are investigating alternative polarizing techniques such as stacked glass-plate polarizers, which have been used at other laboratories.

Experiments by Stamper et al.²⁷ have shown that large magnetic fields are generated in laser-heated plasmas by thermoelectric effects. Now we have demonstrated with computer calculations that inclusion of these magnetic fields is important in obtaining realistic predictions of neutron production from the plasma.

Using MOKU,²⁸ an Eulerian two-dimensional magnetohydrodynamic explosion code, we calculated the neutron production from a flat solid-deuterium target heated by a laser beam.²⁹ In one set of calculations the thermoelectric source of magnetic fields was not included, and in another set it was included. The calculations without magnetic field gave a neutron production some three orders of magnitude lower than experimental results, while the calculations with magnetic field agreed well with experiment.

For both sets of calculations we assumed a laser pulse with a total energy of 80 J, a pulse length of 4 nsec, a beam radius of 50 μm , and a wavelength of 1.06 μm . The target was a disk of solid deuterium 0.04 cm thick and 0.2 cm in diameter (see Fig. 9).

We assumed that the absorption of the laser light by the plasma occurred through inverse bremsstrahlung (collisional absorption), and we corrected for two-stream plasma instability, a type of anomalous (collisionless) absorption.³⁰ We also applied a similar correction to other plasma-transport coefficients, such

as those determining the rate of thermal conduction by electrons. Since the physics of the anomalous absorption is not well understood, we treated the magnitude of this correction as an adjustable parameter.

The production of neutrons in the absence of magnetic fields can be described as follows. As the laser pulse begins to propagate into the target, its energy is absorbed by the electrons near the surface. Because electrons are good thermal conductors, this energy is rapidly transported away from the region of absorption, resulting in rather low peak electron temperatures (less than 1 keV). Figure 10 shows the lines of constant temperature and density for a typical problem.

Without magnetic fields the peak temperatures occur in material of such low density that the rate at which energy is exchanged between electrons and ions is also low, leading to maximum ion temperatures of less than 0.5 keV. Consequently, the production of neutrons by the $D + D \rightarrow n + {}^3\text{He}$ reaction, which depends strongly on the ion temperature, is also very small. Depending on the size of the anomalous absorption correction, we obtained total neutron yields in the range of 100 to 500.

For problems in which the thermoelectric generation of magnetic fields is included we obtained the following results. Initially the behavior is similar to that of the nonmagnetic problem: the laser energy is absorbed near the target surface by electrons that are consequently heated and thus try to expand. This leads to density gradients along the z-axis (see Fig. 9) and temperature gradients in the radial (r) direction. Consequently, a magnetic field proportional to the cross product of the temperature and density gradients is created in the azimuthal direction (around the z-axis). Computed magnetic field strengths range from 1 to 10 MG.

In contrast to the nonmagnetic case, with the magnetic field the electrons cannot conduct heat across field lines. Thus, heating of the material occurs only in the region of the beam. The result is a flow of electrons out of the target along the z-axis, lowering the density at the target surface and allowing the laser beam to penetrate further into the target, boring a hole in it. The electrons and ions become very hot near the deep end of the hole, where the heat loss rate is low, resulting in a substantial production of neutrons (see Fig. 11).

In general, we found that the production of neutrons is a very sensitive function of the anomalous

Fig. 9. Assumed laser and target configuration.

Fig. 10. Density and temperature contours 2.13 nsec after a laser beam has struck the target - magnetic fields not included. The density contours indicate where the density equals the normal density of the target and where it equals the critical density (the density at which the plasma frequency equals the laser frequency). Lines of constant electron temperature (T_e) and ion temperature (T_i) are also shown; maximum values of T_e and T_i in the problem were 0.828 and 0.352 keV. The dense shock region created by the expanding plasma is indicated.

Fig. 11. Density and temperature contours 2.08 nsec after a laser beam has struck the target - magnetic fields included. Contours of density, electron and ion temperature, and magnetic field (B) are shown; maximum values of T_e , T_i and B in the problem were 15.0 keV, 1.9 keV, and 8.7 MG.

absorption correction and of the assumptions made concerning the behavior of laser light near the critical depth in the plasma (where the plasma and laser frequencies are equal). Depending on the values chosen

for these parameters, the neutron yields we obtained ranged from about 10^3 to 3×10^4 ; the upper end of this range is in good agreement with experimental results.³¹

In summary, our calculation without magnetic fields gave neutron yields on the order of a few hundred neutrons, and this yield was relatively insensitive to the details of the calculation. With magnetic fields the yield was increased by a factor ranging from 10 to 1000 and was very sensitive to the assumed problem

parameters. Although the good agreement between the experiment and some of the problems including the magnetic field may be partially fortuitous, it seems clear that the inclusion of such fields will be necessary in understanding the physics of laser-heated plasmas.

A NEW APPROACH FOR DEMONSTRATING THE FEASIBILITY OF EFFICIENT PULSED CHEMICAL LASERS

We have developed an experimental method that appears to be the simplest and fastest way to search for the branched-chain chemical reactions that may be essential for a high-efficiency pulsed chemical laser. With it we hope to demonstrate that such lasers are feasible. The method is based on a form of initiation designed to separate two important functions: (1) transformation of the stable premixed chemicals to a "critical" state where branched chain reactions

can occur, and (2) production of a suitably intense and appropriately timed uniform supply of radicals in the mixture to initiate the reactions. We produce the critical state by using a pulsed CO_2 laser to rapidly preheat the chemical mixture through vibrational-to-translational energy transfer, then we produce the supply of radicals to initiate the branched chain reactions by an electric discharge. The apparatus can also be used to search for chemical laser action

Fig. 12. Pressure-temperature diagram for a spontaneously exploding gas. In the region of strong branching, chemical reactions readily occur that multiply the number of free radicals and thus increase the number of molecules in excited states that are responsible for laser action.

induced solely by the CO_2 laser, and to study the temperature dependence of some chemical laser systems.

Background

The efficient pulsed chemical laser is normally envisioned by reference to an "explosion-peninsula" diagram for a spontaneously exploding gas mixture that exhibits branched chain reactions (see Fig. 12). Such a diagram indicates regions of stability and spontaneous explosion for chemical mixtures as a function of the pressure, temperature, and (implicitly) the relative concentration of reactants. Several Russian scientists³² have proposed that within the ignition region there may be an island or zone of branching chain reactions that produce laser inversion. The objective in the development of an efficient pulsed chemical laser is to be able to premix reaction gases in a stable pressure, temperature, and concentration region and then provide, at will, a weak uniform source of initiation radicals to drive the mixture into the desired zone of branched reaction. For the laser to be compact and yet be capable of high power output it is also desirable to be able to use pressures of at least 1 atm. Unfortunately, no one to our knowledge has yet achieved these objectives.

In our early work on the initiation of $\text{IF}_7\text{-H}_2$ mixtures with a 3-nsec electron beam we reported that the output and pulse width of the laser energy depended on the pressure.³³ The energy output did not steadily increase with pressure but instead reached a maximum at about 125 Torr with a maximum energy density of about 8 J/liter. This observation emphasized for the first time that simply increasing the pressure would not necessarily produce the desired high power and high energy density in pulsed HF systems. Furthermore, the output pulse widths decreased rapidly to about 200 nsec near and above the pressure for optimum output energy. Other HF-producing systems show similar behavior.³⁴ Because of the relatively long electronic-to-kinetic energy transfer times and also the low thermal equivalent of the initiation energy ($\sim 10^\circ\text{K}$) we do not believe the gas reached the spontaneous explosion region in Fig. 12 before termination of the laser pulse. We conclude that the laser output was primarily the result of the radicals produced by the electron beam initiation, which reacted to form excited HF. Thus our initiation method did not stimulate the fast, inversion-producing, branched chain reaction that is theoretically required to overcome the detrimental effects of collisional relaxation and three-body recombination.

Since many chemical systems and initial pressure-temperature-composition conditions had been tried using a variety of initiation methods (flashlamps,

electric discharges, electron beams) with no evidence of a contribution from branched chain reactions, we questioned the adequacy of conventional initiation techniques. Most such techniques utilize the heat of reaction to drive the mixture into the explosion region, and they start the formation of radicals in an environment which, by virtue of its stability, must inherently quench any chain reactions. Hence, the progress of the pressure-temperature coordinates toward the ignition region is probably slow (thermal) and entails undesirable side effects prior to the branched-chain part of the reaction. Such side effects include buildup of a thermal distribution of the vibrational states of HF (which reduces the population inversion required for lasing), buildup of additional collisional quenching species, and depletion of the reactants.

We considered several ways of shifting the pressure-temperature-concentration coordinates rapidly into the region of fast branched reaction prior to the formation of a sizable quantity of radicals for uniform initiation. These included rapid adiabatic compression, microwave heating, shock tube techniques, bleachable inhibitors, and preheating by laser stimulation of vibrational-to-translational or rotational-to-translational energy transfer. We chose to pursue the vibrational-to-translational preheating approach because the short (microsecond) vibrational-to-translational relaxation times and the availability of high-power CO_2 laser radiation offer the possibility of producing significant and rapid changes in the pressure-temperature coordinates prior to initiation by conventional discharge or flash photolysis. Furthermore, this technique is applicable directly to gases that absorb CO_2 laser wavelengths (10.6 μm) and can be extended to any gas mixture by doping it with small amounts (10-20%, say) of materials such as SF_6 , which strongly absorb near 10.6 μm . In this manner we can in microsecond risetimes achieve temperature jumps of at least a few hundred degrees Kelvin.

Experimental Apparatus

Figure 13 is a schematic diagram of our apparatus for preheating gas mixtures prior to discharge. Essentially, it consists of a double-discharge CO_2 laser³⁵ whose output is directed into the side of a chemical laser cell through a NaCl window. The CO_2 laser can produce up to 90 J in about 1 μsec over a 5-by-12-cm aperture. An electrical pin discharge for initiating the chemical laser can be fired at any time between 6 μsec before and 100 msec after the CO_2 laser is fired. This pin discharge provides 20-to-100-keV pulses with durations of less than 80 nsec.

Diagnostic equipment at present includes a photomultiplier for monitoring visible and near-uv

Fig. 13. Schematic diagram of the experimental chemical-laser apparatus in which a CO_2 laser is used for preheating the gas mixture before discharge.

chemiluminescence, calorimeters to monitor the chemical-laser output energy and the CO_2 -laser radiation transmitted through the chemical laser cell, Ge(Au) detectors to monitor the pulse shapes and timing of the two lasers, and current-voltage monitoring of the pin discharge. Within the next few months we hope to do time-resolved spectroscopy on the chemical laser output and the chemiluminescence. Also, we are designing a new chemical laser cell that can operate at pressures up to 1 atm. Shock-wave breakage of Brewster-angle windows now limits the cell to pressures of about 50 Torr.

Experimental Results

Using this equipment we studied $\text{NF}_3\text{-H}_2\text{-SF}_6$ mixtures at 25 Torr, which would explode on unfocused CO_2 irradiation alone. For the mixture ratios studied, the overall energy output always decreases as the delay between preheat and discharge

is increased (see Fig. 14). Despite this degradation of overall laser output, we have observed some HF laser pulse shapes that seem to indicate an enhancement in the power of the first transition to lase for preheat delays of about 10 μsec (see Fig. 15). We have not yet had a chance to do monochromator measurements to check the lasing transition sequence, so this inference is tentative.

Our data on $\text{N}_2\text{F}_4\text{-H}_2$ mixtures at 25 Torr, which would not explode on CO_2 laser irradiation alone, are shown in Fig. 16. These mixtures varied only in the small amount of *cis*-2-butene added to inhibit spontaneous explosion during mixing and preshot storage. The bumps and dips in Fig. 16 presumably indicate vibrational energy transfer processes and dissociation in N_2F_4 . The eventual approach to the discharge-only (no preheating) output level indicates that the system returns to the initial pressure-temperature conditions without explosion.

Fig. 14. Dependence of the HF laser energy output on the preheat-to-discharge delay time for various $\text{NF}_3\text{-H}_2\text{-SF}_6$ mixtures. The temperature of the gas mixture at the time of the discharge increases with the time delay (for delays up to a few hundred microseconds).

Fig. 15. A sequence of HF-laser pulse shapes showing the effect of CO_2 laser preheating for different CO_2 -to-discharge delay times. The laser gas mixture is 6:3:2 $\text{NF}_3\text{:H}_2\text{:SF}_6$ at 25 Torr. The discharge itself lasts about 80 nsec.

Fig. 16. HF laser output energy vs preheat-to-discharge delay time for a 75:25 $\text{N}_2\text{F}_4\text{:H}_2$ mixture at 25 Torr, for various amounts of *cis*-2-butene spontaneous-explosion inhibitor.

In addition to the preheating experiments just described, we also look for chemical laser action initiated by the CO_2 laser radiation alone. Such a discovery would be exciting because it might lead to improvement of overall chemical-laser efficiency and allow conversion to other wavelengths. In our investigations thus far we have not observed any initiation of HF chemical-laser *oscillation* by the CO_2 laser, although the CO_2 radiation is capable of inducing explosions in some of the reactive mixtures. This was true even for $\text{N}_2\text{F}_4\text{-H}_2$, for which there was an early unpublished claim³⁶ that laser action had been

observed. Subsequently we learned³⁷ that the observation had consisted of weak, and possibly questionable, *amplification* of a probe laser beam.

An important aspect of the CO_2 -initiated explosions is the time development of the chemiluminescence. By this measurement we hope to be able to observe the presence of a fast branched-chain reaction independent of any enhancement of the laser output; the more chain branching that is occurring, the sooner will chemiluminescence appear. Figure 17 shows the time of onset of chemiluminescence in various mixtures containing NF_3 or N_2F_4 . The chemiluminescence appears sooner for NF_3/H_2 ratios of 1 and 3.5 than for a ratio of 2.

Referring to Fig. 14, one can see that the faster the quenching speed, the earlier the chemiluminescence appears. This behavior would be expected for thermal explosions. It is also noteworthy that the laser output is quenched before the chemiluminescence appears; however, until we do some spectroscopy we cannot infer with certainty that the HF-producing reactions are taking place at the same rate or at the same time as the chemiluminescence is being emitted.

Conclusion

In summary, we have developed a versatile experimental system to (1) search for branched-chain reactions which could contribute importantly to the output of chemical lasers, (2) search for chemical-laser action initiated by a CO_2 laser, and (3) study the temperature dependence of pulsed chemical-laser systems.

Our initial results with $\text{NF}_3\text{-H}_2\text{-SF}_6$ and $\text{N}_2\text{F}_4\text{-H}_2$ mixtures at 25 Torr show that the overall energy output decreases as the temperature is raised above 20°C , which indicates that thermal reactions, rather than branched chain reactions, are occurring. Tentatively, it appears that the first transitions to lase can be somewhat enhanced by preheating. We have not yet seen any chemical laser action initiated purely by the CO_2 laser, which is not surprising in view of the slow buildup of chemiluminescence.

Our main conclusion is that the techniques we have developed are the simplest and fastest way to search for branched chain reactions contributing to laser output and to demonstrate that efficient pulsed chemical lasers are feasible. We will continue to investigate more promising chemical systems.

Fig. 17. Time-dependent chemiluminescence data for some CO_2 -laser-initiated explosions (no pin discharge) at 25 Torr.

The output of one of our lasers, the Short Path Laser, has been limited to unacceptably low intensities due to self-focusing damage in the Nd:glass amplifier rods. Experiments on this laser have demonstrated that the self-focusing in high-peak-power lasers is governed mainly by incidental and often unavoidable diffraction effects or transverse-mode perturbations. Thus, bulk rod damage occurs at power levels well below that at which large-scale self-focusing of the entire beam occurs. We have quantitatively corroborated these experimental observations by computer calculations.

Experimental

The Short Path Laser consists of a mode-locked Nd:glass oscillator, a single-pulse switchout, and a rod amplifier chain (see Fig. 17). The oscillator provides 15-psec pulses with bandwidths of about 50 Å. The detailed structure of these pulses is not known.

Following a 1/2-in. "preamplifier," a lens-pinhole-lens spatial filter gives a diffraction-limited, spatially smooth beam that is expanded from the 4-mm oscillator output to about 1.3 cm. An aperture after the spatial filter may be used to restrict the beam entering the next 1/2-in. amplifier, which is 108 cm from the aperture. This amplifier is followed by two more — each separated by 19 cm. Each amplifier is pumped with up to 12 kJ and gives a small-signal gain of about 10. The system is capable of about 2 J out of the 1/2-in. chain, in a nearly diffraction-limited beam.

When we fire about 50 shots, all less than 0.5 J, using the 1/2-in. chain, the last 1/2-in. laser rod is severely damaged. The damage takes the form of a Fresnel diffraction pattern, which results from the

aperture following the spatial filter. Careful inspection reveals that the ring pattern is composed of straight trains of damage sites that begin 8 cm in from the exit face and become more numerous close to the face (see Fig. 18). Significantly, on the same shots that produced damage, the beam was nearly diffraction-limited (as indicated by far-field divergence measurements); only a small fraction of the beam energy was involved in the self-focusing.

Removal of the aperture after the spatial filter eliminated the diffraction ring pattern, but the diffraction from the edges of the rods then introduced slight (less than 20%) fluctuations in the intensity distribution out of the 1/2-in. rods. After many days of firing at low energy (less than 0.25 J), we observed new damage at about the same place as before, but with a less distinct pattern.

Theoretical

In our calculation of self-focusing we assumed the input pulse energy to be 2 mJ and we adjusted the rod gain so that the energy output of the final rod amplifier was 0.25 J. These energy values are representative of values measured for a number of firings of the laser. We used 1.555 for the normal index of refraction of the Nd:glass and 2×10^{13} esu for n_2 , the power-dependent term of the index of refraction.²⁸

Figure 19 shows the calculated evolution of the Fresnel diffraction and self-focusing pattern between the aperture and a point 8 cm before the exit end of the final amplifier stage, where damage to the glass was first observed. Each of the calculated

Fig. 17. Portion of the Short Path Laser involved in the self-focusing experiments.

Fig. 18. Damage observed at the end of the last 1/2-in.-diam Nd:glass amplifier rod.

radial-intensity profiles shown in Fig. 19 corresponds to the time in the pulse when the maximum on-axis intensity occurred.

The positions of the observed fringes (see Fig. 18) are noted at the top of Fig. 19 by dashed lines. The overall agreement between the observed and the calculated fringe positions is quite good. Only the calculated fringe at a radius of 0.075 cm is difficult to find in Fig. 18, although there are some spots in the lower half of the rod at the correct radial position for it. The peak intensity occurs at the center of the rod (zero radius). This central maximum, which focuses first, is off-scale in the uppermost profile. The fringes do not change their position between this point and the end of the rod, but they do sharpen considerably.

Conclusions

Both the experimental and the theoretical results indicate that, in any practical system, damage levels will be governed by the quality of the beam. This, in turn, is readily altered (i.e., degraded) by apertures.

Experimental elimination of diffraction effects will require underfilling the rods (i.e., not filling the entire diameter of the rod with the beam), so that the edges aperture the beam at a point of very low intensity. This will be accomplished by reducing the expansion of the spatial filter. However, the most promising solution to self-focusing problems is to use a longer pulse — perhaps 100 psec or more.²⁹ By narrowing the bandwidth of an yttrium-aluminum-garnet (YAG) oscillator with intracavity etalons, a longer pulse can be produced. Even if this effort does not give 100-psec pulses, at least we will obtain a bandwidth-limited pulse from the YAG. We are now using or preparing to use such oscillators on our other Nd:glass systems. We can then investigate self-focusing further using a well-understood pulse.

Fig. 19. Self-focusing occurs in rings determined by Fresnel diffraction patterns. Shown here is the evolution of the Fresnel pattern from the aperture to the point where evidence of damage is first observed (8 cm in from the exit end of the last rod). Intensity values shown at the left are at or near the maximum intensity calculated at each position. Intensity at radius zero (center of the rod) for the top curve is off-scale. Dashed lines at top mark the positions of the fringes in the damage pattern shown in Fig. 18.

Recent Laboratory studies of holography and its applications to nondestructive testing have resulted in a double-exposure holographic method for inspecting the welds of pressure vessels, a holographic technique for plasma diagnostics, and a method of implanting third-dimensional information in two-dimensional x-ray shadowgraphs. We have also developed a pulsed ruby laser system that operates in a single longitudinal and transverse mode and have studied the "chirping," or frequency shift with time, that occurs in its pulses. We are working on a holographic technique for analyzing picosecond infrared pulses.

A hologram is a record of interference patterns between light waves reflected or diffracted by an object and a suitable reference light wave. If the phase relation between the illuminating and reference waves remains constant in time and space (i.e., if the light is coherent), the hologram then records both the amplitude and the phase of the reflected light, whereas ordinary photographs record only light amplitude.

Since holograms contain this phase information, they can be used to reconstruct a replica of the original light wave that exhibits the parallax, perspective, and depth of field of the original three-dimensional object. This is achieved by illuminating the hologram with a coherent light beam similar to the original reference beam.

Double-Exposure Holography

One of the properties of holography is that several overlapping holograms can be stored on the same photographic plate. This is the basis of holographic interferometry, in which one makes two exposures, a "before" and an "after," on the same recording emulsion. Upon reconstruction, the two images will interfere, giving a measurable account of what happened during the time between the two exposures. Holographic interferometry is simpler than conventional optical interferometry, and the painstaking alignment of components is not required.

Our first use of double-exposure holography in nondestructive testing was to study the quality of the weld around the equator of metal hemishells welded together to make pressure vessels. By making one exposure of the vessel at ambient pressure and another at high pressure, we can determine the uniformity of the weld (see Fig. 19) and measure quantitatively the distortion of the vessel and the displacement of the seam at a variety of pressures. Promising results in

initial tests proved the feasibility of this technique, and we have now adopted it as one of our standard nondestructive-testing methods for a variety of applications.

The double-exposure technique is also useful for observing the electron density in plasmas as a function of time. Its simplicity, lack of critical alignment requirements, and modest optical-quality requirements relative to conventional interferometry make it a better choice in plasma diagnostics. This is of particular interest in the laser-fusion program, and we now have a holographic diagnostic effort under way as part of that program.

Pulsed Holography and Chirping

Pulsed holography refers to the technique of making holograms using short pulses of laser light. Among its advantages over continuous-wave holography are that the difficulty of stabilizing holographic equipment is eliminated and its stop-action capability extends the

Fig. 19. Image of a double-exposure hologram of the weld in an aluminum pressure vessel. The closely spaced fringes in the upper part of the picture mark considerable strain in the part of the weld that penetrated only 25% of the aluminum. The relatively strain-free weld in the lower part of the picture had a penetration of 75%.

Fig. 20. Photograph of the hologram-reconstructed image of a 20-cm-diam disk rotating at 9400 rpm. The rim speed of the disk is about 0.1 mm/ μ sec (98 m/sec). The resolution time of this technique is such that even at this fast rim speed there is no difference in the clarity of the holographic image and that of the actual object.

use of holographic techniques to many transient phenomena, such as reactions in plasmas.

The chief difficulty in pulsed holography is that the many modes of the output of a normal Q-switched laser limit the coherent length of the light. To overcome this problem we have developed a ruby laser system that operates in a single longitudinal and transverse mode.¹⁹ The system uses a saturable dye absorber for Q-switching, an etalon resonant reflector and intracavity pinhole for mode selection, and an amplifier for increasing the output energy to a usable level. The single-mode pulses have an FWHM of 5 to 10 nsec. Figure 20 is an example of a pulsed hologram obtained with this system.

Although our Q-switched laser puts out pulses of a single frequency, the frequency is not stable in time; it shifts during the pulse. This phenomenon is called frequency chirping, a term borrowed from radar technology. The quality of the pulsed holograms depends on the amount of chirp in the laser pulse. We have experimentally studied the correlation between the magnitude of the frequency shift, the relative brightness of holograms made with various path-length differences between the two beams, and the visibility of the interference fringes, which is related to the degree of coherence.²⁰ We found that

the frequency shift was approximately linear in time; the shift rate was 18 MHz/nsec for a pulse with a 12-nsec FWHM and 57 MHz/nsec for a 6-nsec pulse. (We shortened the pulse length by increasing the concentration of the Q-switching dye.)

We then used the same pulses to generate simple interference holograms. To do this we split the beam and allowed the two parts to superimpose on a photographic plate after having traversed paths of different lengths. After developing the plates we measured the light diffracted (into the first order) as a function of path-length difference. The amount of light diffracted will decrease as the path-length difference increases because the two pulses do not completely overlap in time and because the chirp causes a phase difference which can wash out the fringe pattern that is the source of the hologram.

By measuring the pulse length and the frequency shift we can predict their effects on the diffraction power and the fringe visibility. The incomplete overlap of the pulses implies a decrease in diffraction power or fringe visibility for a characteristic length difference of many meters (essentially the length of the pulses), whereas the measured chirp value, assuming that the pulses are Gaussian in shape, lead to characteristic lengths of about 60 cm for the 6-nsec pulse and about 90 cm for the 12-nsec pulse. The experimental values were about 45 and 75 cm, respectively. The deviation of the pulse shape from Gaussian and the fact that the chirp is not linear at the beginning and end of the pulse probably account for the difference between the predicted and measured values.

Pseudoholographic Reproduction of Three-Dimensional X-Ray Images

Radiograms are two-dimensional x-ray shadowgrams of an object. Since the advent of holography it has been a hope to apply this technique to the x-ray region, but the hope has not yet been realized because the x-ray laser has yet to be developed. Nevertheless, we have developed a holographic technique by which phase information is implanted in radiograms,²¹ resulting in three-dimensional x-ray images.

The method involves two steps. The first is to take a sequence of positive transparency shadowgrams, each having a different perspective, using a set of (incoherent) point x-ray sources or using a single source and moving it in steps to different positions. The second step is the introduction of phase information by holography. The shadowgram transparency is placed in close contact with a high-resolution photographic plate. The incoherent x-ray point source is replaced by a coherent laser source, with extreme care being taken that the divergence of the beam corresponds exactly to that

of the incoherent source when the shadowgram was made. In addition, we introduce onto this recording plate a fixed, collimated, reference laser beam at an oblique angle θ . Then, by repeating the process as in the first step, we synthesize a sequence of holograms by placing the coherent source at the corresponding positions of the x-ray source of the shadowgraph. In this way, the phase information is restored through the correct interference between the coherent point source and the coherent reference beam. Consequently, the synthetic hologram possesses the intensity distribution from the contact point of the shadowgram and the phase information through the interference of the coherent light. The composite hologram can then produce a three-dimensional image of the object when the hologram is illuminated at an angle θ from the opposite side by a collimated laser beam. Precise measurements can be done on the three-dimensional image, making this technique a convenient way of studying the internal structure of a three-dimensional object.

Picosecond Holographic Diagnostics

We are working on a holographic diagnostic technique for picosecond infrared pulses for the laser-fusion program. We hope to be able to determine

holographically such parameters as the spatial coherence of the pulse and its frequency behavior as a function of time.

Our procedure is to switch out a single pulse from the mode-locked Nd:glass laser using a spark-gap-triggered Pockels cell.²² After amplifying the pulse to an energy of about 1 J, we split out a portion of the pulse and compress it to an even shorter duration using a pair of diffraction gratings. The compressed pulse is, in effect, a "known" pulse that can then be combined on a hologram with the uncompressed pulse; by choosing the proper timing we can make the compressed pulse interact with any part of the uncompressed pulse and thus gain information on the nature of the uncompressed pulse.

At present, we are developing the laser amplifiers for the mode-locked Nd:glass laser. The next step, pulse compression, should be achieved in the near future.

Since high-resolution photographic film is not available for this region of the spectrum (1.06 μm), we have to look for other recording means. Thin films of low thermoconductivity and a combination photoconductor/thermoplastic film are good candidates for this purpose, and we are now investigating their suitability.

A LARGE-APERTURE TEA CO₂ AMPLIFIER

We have built a large-aperture amplifier for 1-nsec laser pulses that uses a uv-preionized TEA CO₂-N₂-He discharge. This amplifier has produced peak gains as high as 0.032 cm⁻¹ (i.e., per cm of amplifier length) and reliable gains as high as 0.028 cm⁻¹. Flat and reproducible gains can be obtained over apertures as large as 10.7 cm in diameter.

Enlarging the aperture size is the easiest solution to several problems that arise in the design of CO₂ oscillator-amplifier chains for high-energy pulsed operation. The large beam areas help prevent optical breakdown of the gain medium in the amplifiers for 1-nsec pulses of the 10.6- μ m CO₂ light. Also, the saturation energy for nanosecond pulses has recently been found to be low,¹⁷ which is of concern for at least two reasons. First, to reach a given output pulse energy, one must either maintain the energy per unit area in the propagating pulse at only a few times the saturation energy and increase the beam cross-sectional area accordingly, or one must obtain a large small-signal gain in a constant aperture. But to obtain

large small-signal gains one needs absorbers saturable at 10.6 μ m, and satisfactory performance of such absorbers with nanosecond pulses has not yet been demonstrated. Second, if spatially Gaussian input pulses are assumed and diffraction is ignored, the area of the propagating pulse between its half-maximum points increases linearly with distance once the energy of the propagating pulse is larger than the saturation energy. If such a pulse is to be propagated without having significant portions of the beam become too large in diameter for the amplifiers, large-cross-section amplifiers are needed. For these reasons, the amplification of 1-nsec pulses to energies of hundreds of joules will require the excitation of large volumes of gas mixtures with beam apertures of 100 cm² or more.

One proved way to achieve large-aperture performance in an amplifier is by electron-beam discharges. However, because electron-beam devices are expensive to build and operate, we looked for an alternative among the several uv-preionized-and-sustained

Fig. 11. Schematic drawing of the 50-cm TEA amplifier module.

devices recently reported.^{18,19} In a test chamber similar to that of Seguin and Tulip¹⁹ we were able to deposit 525 J/liter at an electrode spacing of 10 cm into a mixture of CO₂, nitrogen, and helium. Less-uniform but still arc-free glow discharges could be produced even when the electrodes were separated by 22.5 cm (the limit of our apparatus).

Thus encouraged, we fabricated the 50-cm-long amplifier module shown schematically in Fig. 11. Both the anode and the cathode were cut accurately to a Rogowski equipotential profile calculated for an 8-cm electrode spacing. A uv-preionization pulse was obtained by discharging the main capacitor bank through 30 1-cm arcs between tungsten pins. The discharge between the main electrodes is adequately delayed to allow uv preionization by choosing a timing capacitor with a value 10 to 20% of that of the main bank.

This structure has provided extremely uniform and reproducible plasmas in a variety of CO₂-N₂-He

Fig. 12. Typical gain-vs-time trace for the 50-cm amplifier module.

mixtures. We measured the gain on the P18 transition with a 3-W cw probe laser having an internal grating and operating in the TEM₀₀ mode. Figure 12 compares a typical gain-vs-time trace with the

Fig. 13. Gain of the 50-cm amplifier module measured along the midplane between the two electrodes.

prediction of a five-temperature model of CO₂ molecular dynamics.²⁰

Fig. 14. Gain of the 50-cm amplifier module measured along the centerline (normal to the electrodes). See measurement zone sketch in previous figure.

Figures 13 and 14 show the remarkable uniformity of the gain provided by this amplifier structure. Within the accuracy of the measurement technique, flat and reproducible gain can be obtained over a usable aperture 10.7 cm in diameter. Uniform discharges have been obtained for electrode separations as large as 12.5 cm; however, the reliable performance range corresponded to separations of 10 cm or less. Still, this is one of the largest-aperture laser amplifiers ever successfully operated.

We have obtained peak gains of 0.032 cm⁻¹, but at the expense of intermittent arcing. The reliable performance range corresponded to gas mixtures and energy depositions yielding gains of 0.028 cm⁻¹ or less. Table 1 gives the electrode separation (beam aperture), gas composition, and applied voltage for several reliable operating points.

When operated as an oscillator with a 9.0-cm aperture and an uncoated germanium output mirror, this device yielded 55 J per pulse. We measured this energy with a 20-cm-diam bulk-absorbing thermopile consisting of Teflon sheets on copper. Energy measurements using cone-shaped ballistic thermopiles gave erratic but substantially higher estimates.

Table 1. Parameters of several reliable operating points of the 50-cm module

Applied voltage (kV)	Electrode separation (cm)	E/P ^a (V/cm-Torr)	Input energy (J)	Input energy density (J/liter)	Gas composition (%)			Reliable gain (cm ⁻¹)
					He	CO ₂	N ₂	
50	7.02	9.33	280.0	111.7	84.4	11.2	4.4	0.0280
50	7.02	9.33	280.0	111.7	88.0	8.6	3.4	0.0130
55	7.78	9.31	338.8	85.8	85.8	10.8	3.4	0.0140
70	7.78	11.91	548.8	138.9	85.8	10.8	3.4	0.0131
55	7.78	9.31	338.8	85.8	82.0	11.2	6.8	0.0195
55	7.78	9.31	453.5	115.0	86.4	8.7	4.9	0.0191
55	10.66	6.79	338.8	66.2	88.0	8.6	3.4	0.0121

^aE/P is a measure of the electron mobility:

$$E/P = \frac{(\text{voltage})}{(\text{electrode separation}) \times (\text{gas pressure})}$$

SHORT-PULSE GENERATION AND AMPLIFICATION IN A CO₂ TEA LASER OSCILLATOR-AMPLIFIER

We have amplified a single 1-nsec CO₂ laser pulse to an energy of 2 J in a simple system consisting of an LLL-built mode-locked externally switched TEA oscillator and two commercially built TEA amplifiers,

with an effective aperture of 25 mm.²¹ This energy represents a flux of about 400 mJ/cm² in the output beam, which is about 10 times the available stored energy in a single CO₂ rotational transition in the

Fig. 15. The LLL-built TEA oscillator, actively mode-locked and externally switched.

amplifier chain. This high flux can be explained in terms of conventional single-line output theory if the rotational cross-relaxation times between the various transitions available in the $10.6\text{-}\mu\text{m}$ P-branch of CO_2 are much less than 1 nsec, as the data of Cheo and Abrams suggest.²² However, spectral measurements show that our system is not lasing on a single line but rather on a number of lines simultaneously; as many as 12 have been observed. Thus we do not have single-line output but rather multiline output. Some cross-relaxation must also be occurring, for the P20 line contains about half the total intensity of all the lines, a disproportionately large share according to our theoretical studies. Hence the explanation for the high flux level appears to be a combination of multiline output and some cross-relaxation of the rotational transitions. We have a theoretical model under development.

Figure 15 shows the LLL TEA oscillator.²³ The principal difference between this and other oscillators is the use of a three-section Lamberton-Pearson gain medium²⁴ whose active length is 150 cm. The single-pass small-signal gain of this device is of the order of 50.

A typical mode-locked train of light pulses sampled before switchoff contains 70 mJ in a train of about 15 1-nsec pulses. After switchoff, aperturing and attenuation reduce the energy of the switched-out peak pulse to 1.0 mJ in a 5-mm-diam beam. The magnitudes of pre- and post-pulses are reduced by a factor of at least 200 relative to that of the switched-out pulse. In the far field (5 m or more beyond the oscillator)

the output is nearly diffraction limited. To measure the far-field pattern we used a 128-element linear array of pyroelectric detectors. Figure 16 shows a typical trace of such a measurement. Inside the gain medium the circulating pulse has a peak energy of about 30 mJ, corresponding to a flux of about 60 mJ/cm^2 in the lasing volume.

The amplifiers used in this study were manufactured by Lumonics Research Ltd., Ottawa, Ont. Each amplifier has an effective aperture of 25 mm and a gain length of 1.35 m. The gas mixture was 86% helium, 7% nitrogen, and 7% CO_2 . From the oscillator exit it is 3 m to the entrance of the first amplifier and 8 m to the exit of the second amplifier.

We measured the small-signal gain of each amplifier to be 0.025 cm^{-1} on the device centerline, or 50 per

Fig. 16. Far-field intensity profile of the output of the LLL oscillator.

Fig. 17. Output spectrum of the LLL oscillator obtained with a pyroelectric array detector.

pass. Within the effective aperture the small-signal gain drops off as much as 20% on a vertical scan away from the centerline, but no more than 10% in a horizontal scan. From this small-signal gain and the saturation energy flux (9.8 mJ/cm^2), we calculate that the maximum extractable energy flux of each amplifier is 33 mJ/cm^2 per pass when it is operating on a single transition, neglecting cross-relaxation effects. If, on the other hand, cross-relaxation is effective or if the amplifier input spectrum contains an equal amount of energy in all rotational lines, the saturation flux is about 160 mJ/cm^2 , and a considerably larger amount of extractable energy is available.

In fact, we have repeatedly propagated a 1-mJ, 1-nsec pulse through our amplifier chain at peak gain and obtained an energy output of $2.0 \pm 0.5 \text{ J}$. This corresponds to an energy density of about 400 mJ/cm^2 , extracted mainly from the second amplifier. We believe there is little possibility that any significant part of this amplified output arises from amplification of a long, low-level signal, since we have removed the nitrogen-caused tail from our oscillator output pulses by judicious selection of the gas mixture, and since the switchout device attenuates the oscillator output by a factor of 200 or more except during a single 5-nsec window. Also, we have directly observed that the amplified signal has no perceptible amplitude after the single pulse.

In an attempt to resolve this disparity between the measured extracted energy flux and that projected (neglecting cross-relaxation effects), we scanned the spectrum of the oscillator using a small spectrograph with liquid-crystal and graphite targets at the exit focal plane of the oscillator. We routinely saw at least four lines, occasionally six.

We then set up a 5-m spectrometer designed for an f-number of 50, which gives an expected diffraction-limited spot size at the exit plane of 0.6 mm. With the focal lengths and diffraction grating we used, the dispersion is about 0.6 mm/nm, giving a spread of about 12 mm between adjacent rotational transitions of CO_2 (the line spacing is 20 nm at the wavelength of the P20 transition). This dispersion spreads the linewidth of each transition over about 0.55 mm at the exit plane, which is necessary to ensure that no line falls between the elements of our pyroelectric linear-array detector, which has a dead space of 0.1 mm between elements.

With the detector array at the spectrometer exit plane we obtained results such as those in Fig. 17 for the oscillator output. Unfortunately, for quantitative work this approach was not accurate enough. The peak intensities include a possible error of $\pm 50\%$ due to the chance that one line illuminated two detector elements rather than one. However, the array measurements established beyond a doubt that the oscillator lases on several rotational lines simultaneously.

To quantitatively measure the spectral content on each transition, we substituted for the pyroelectric array a Ge:Cu liquid-helium-cooled detector. To correct for the nonlinearity of this detector, we maintained the measured signal between 0.5 and 5 V by inserting calibrated attenuators in the measured beam. In this way the actual measurement was kept accurate to within 10%. Figure 18 gives the results of these measurements, which show that at least 50% of the oscillator output is on the P20 line.

We then measured the output spectrum of the entire oscillator-amplifier chain (actively mode-locked) by attenuating the amplified oscillator output and sending it to the spectrometer. We deliberately tried to saturate both amplifiers by placing a slightly focused 2-power beam-expanding telescope between the oscillator and the amplifiers; this expanded the beam diameter to $11 \pm 1 \text{ mm}$ in both amplifiers in the absence of pumping. With the amplifiers pumped, the exit spot size is difficult to restrict, due to saturation effects. The apparent core of the amplified beam (the burn spot on Polaroid film) was about 20 mm in diameter.

Figure 18 shows the spectrum resulting from saturated amplification of the oscillator output. In this spectrum the intensity of the P20 line is still about half the total intensity of all the lines. In fact, the spectrum is somewhat narrower after amplification than before. In all the spectra we obtained, the intensity decreases exponentially as the P-number increases or decreases from 20, and each spectrum is skewed toward the lower P-numbers. Since we made spectral measurements over a period of several weeks, the agreement between spectra measured under similar

conditions is impressive evidence that the observed behavior is repeatable.

In a classical laser oscillator, single-line operation is achieved by cross-relaxation of the rotational levels into the transition having the highest gain. Measurement of the spectrum of the spiked output of a mode-locked oscillator with variable spike duration is one way of estimating the *effective* cross-relaxation time independent of theoretical prediction. If the spike is much shorter than the cross-relaxation time between rotational levels, then cross-relaxation will not be able to effect single-line output.

Let us assume that the duration of the spike is shorter than the cross-relaxation time and shorter than the time it takes the light to make a round trip through the cavity. Under these conditions relaxation effects will not have time to prevent multiline lasing during a spike, but there will still be enough time to equilibrate between rotational levels during a cavity round trip. Then, using the simple theory of a saturating amplifier and making a number of simplifying assumptions, we can derive what the oscillator output spectrum should be, based on the Boltzmann population of the rotational states of CO_2 . Figure 18 shows the calculated result.

Quantitatively, this calculated curve is a very poor predictor of the observed spectra except that it does predict vanishing intensity at the correct pair of P -values and it gives the correct peak P -value. However, it should be recalled that, if the rotational lines equilibrated in a time very short compared with the mode-locked pulse width, the output spectrum of our oscillator would have consisted of a single line at $P20$. The calculated curve in Fig. 18 represents the opposite extreme — the rotational lines do not equilibrate at all during a mode-locked pulse. Our experimental data suggest that the truth lies between these extremes — namely, that the spike duration is only somewhat longer than the cross-relaxation time. We are now doing a computer analysis that retains both rotational and vibrational equilibration during the mode-locked pulse.

Fig. 18. Quantitative output spectra of the LLL oscillator and oscillator-amplifier chain. The observed spectra were obtained using a Ge:Cu detector. The calculated spectrum is based on the Boltzmann population of the rotational states of CO_2 , assuming that the spike duration is shorter than the rotational cross-relaxation time.

Notes and References

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