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**Laser Development for Laser
Fusion Applications
Research Progress Report
October 1977 - March 1978**

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LASER DEVELOPMENT FOR LASER FUSION APPLICATIONS
RESEARCH PROGRESS REPORT
October 1977 - March 1978

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**LASER DEVELOPMENT FOR LASER FUSION APPLICATIONS
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LASER DEVELOPMENT FOR LASER FUSION APPLICATIONS

INTRODUCTION

Advances in knowledge about laser-driven thermonuclear implosions have been and are being paced by the development of advanced laser drivers. The physical principles of this promising new source of energy are now quite clear, and commercialization of laser-fusion energy will be paced by the development of advanced high-average power laser drivers.

The laser research and development described herein was conducted for the USDOE, Office of Laser Fusion. The goal of this research is to develop the laser technology that is necessary for commercialization of laser-fusion energy.

Our program addresses three promising high-power laser systems: iodine, hydrogen fluoride, and Group VI atoms. Both the iodine laser and the HF laser have a demonstrated high-power capability, and experimental results indicate that there are probably no serious problems hindering development of either of these lasers for laser-fusion applications. In both these technology-development programs, we are making steady technical progress: we know where we are going, we understand the problems and know how to go about solving them, and we have a sufficient number of alternatives to give confidence for success. The Group VI atoms excited to the $1S$ energy level have characteristics that suggest that they could meet the requirements of laser fusion, but progress in the development of this class of lasers has been disappointing. However, recent kinetic studies suggest why early results were disappointing and through such studies we may be able to find ways to circumvent the problem areas.

HF-LASER PROGRAM

Overview

There were three major accomplishments in this present reporting period. The first demonstration that a high-optical-quality beam could be obtained from an HF oscillator-amplifier system. By pinhole transmission studies it was demonstrated that the beam from the present system (~ 200 -J extractable energy) is less than three-times diffraction limited, and the promise of even better optical quality is good. The second accomplishment was the development of a high-optical-quality, short pulse (5 to 25 ns), oscillator-preamplifier system. This system will be used to study the optical beam quality of amplified beams whose pulse widths are shorter than the chemical burn time. The third accomplishment is that better agreement between code and experiment was obtained. This was achieved by taking into account electron-beam nonuniformity in the medium. A cost per F atom of 5 electron volts now gives reasonable agreement between code and experiment for both pulse width and laser energy for the cases investigated to date.

Beam Quality Studies of Phoenix: A Near Diffraction-Limited HF Laser System

The principal question that had persisted about the use of HF lasers for laser-fusion applications was whether the amplifier medium, which consists of hot reacting gases and high-energy electrons, would maintain the high optical quality of the input beam. The experiments conducted during this report period emphasized studies of the quality of optical beams

amplified in an electron-beam-initiated HF amplifier. The optical quality of the incident and amplified laser beams was investigated by using a lateral shearing interferometer, a focal spot intensity distribution measurement technique, and pinhole energy transmission measurements.

The oscillator-amplifier chain and focusing system are shown schematically in Fig. 1. The electric-discharge-initiated oscillator-preamplifier system was fueled with a mixture of SF_6 and H_2 . SF_6 and H_2 were used in the present system because these gases yield HF molecules excited up to the sixth vibrational level, which is also the highest level populated in $\text{H}_2\text{-F}_2$ mixtures. A 20:1 pressure ratio of SF_6 to H_2 was used in these experiments because it yields the best efficiency and line overlap. The characteristics of the oscillator-preamplifier system were described in the last semiannual report.¹ A reflective two-times expanding telescope and spatial filter were used after the last preamplifier. The input beam to the amplifier was 7.0 cm in diameter and contained an energy of 1 joule in a pulse width of 100 ns (FWHM). The spectrum from the oscillator-preamplifier system consisted of 25 P-branch lines originating from vibrational levels up through $v = 5$.

The electron-beam-initiated amplifier studied here was also described in the previous semiannual report.¹ The pressure ratio of $\text{F}_2/\text{O}_2/\text{H}_2$ used in the amplifier for these experiments was 1.0/0.3/0.25 with a total pressure of 2340 torr. The O_2 is used to stabilize the mixture against spontaneous detonations. The amplifier, when configured as an oscillator under these conditions, produced an 180-J beam with a 50-ns (FWHM) pulse. The observed spectra contained 45 P-branch lines originating from the $v \leq 6$ levels of HF. The optical cavity consisted of a 12-m radius of curvature total reflector and a sapphire flat placed directly on the ends of the 91-cm-long amplifier cell.

For the oscillator-amplifier experiment gold-doped germanium detectors, which have risetimes of about 1 ns, were used to monitor the time history of the laser pulse into and out of the amplifier. Signals from these detectors were time correlated with the electron-beam-current pulse. The timing sequence between the input laser pulse and the electron-beam current was selected so that the maximum number of oscillator lines were present in the amplifier at the time of maximum inversion. In

Fig. 1. Experimental arrangement for pinhole transmission measurements.

setting this timing, it was assumed that peak inversion occurred at the time of the peak laser output that was obtained when the amplifier was configured as an oscillator. Time-resolved spectra from the oscillator-preamplifier were measured to determine the time interval containing the maximum number of lines.

The beam quality into and out of the amplifier was first investigated by using lateral shearing interferometry. In this method, reflections from the front and rear surface of a small-angle wedge interfere and form a fringe pattern. The shape and orientations of these fringes contain information about the radius of curvature and phase-front distortion of the beam.² In these experiments, the reflected interference patterns were recorded as burn patterns on unexposed developed Polaroid film.

Figure 2 shows interferograms taken at the input and output of the amplifier. The two patterns are shown at different magnifications and, therefore, are not to scale. The pattern shown on the left was obtained with lower intensity than the one on the right, which accounts for the absence of film burn outside the interference region. Analysis of the interferograms indicate that the phase-front distortion was less than $\lambda/2$ over the full diameter and that the divergence was approximately 220μ radians for both beams. This is about two times the diffraction limit value. Both interferograms exhibit the same fringe quality indicating that

the phase front distortion was not significantly increased by the amplifier medium. The results of the interferometry measurements indicate that the output from the amplifier should be focusable to a near-diffraction-limited spot size.

The focal spot radial intensity distribution was obtained by using a CaF_2 flat and a flat copper mirror to generate multiple reflections of the beam with various intensities.³ These images were then focused with a 5-m focal length mirror onto unexposed developed Polaroid film. The focal spot intensity distributions of the input and amplified beams is shown in Fig. 3. The intensity from the oscillator was not sufficient to determine whether the beam had a low intensity at large radius similar to that exhibited by the amplifier output. The integral of the intensity distribution of the amplified beam, shown in Fig. 4, indicated that the beam divergence obtained by this method was no more than five times diffraction-limited. Part of the increase in focal spot size can be attributed to double surface reflections from the CaF_2 flat which resulted in slightly displaced double images at the focal point. Therefore, this technique yielded an upper limit for the focal spot diameter. Attempts to use a wedge in place of the flat resulted in dispersion of the broad HF spectrum and an increased focal spot size. In obtaining the data for Fig. 3, it is assumed that there was not significant low-intensity energy at large radii.

Fig. 2. Lateral shearing interferograms of input and amplified beams.

For the pinhole energy transmission measurements, front surface reflections from combinations of infrasil and ZnSe wedges were used to reduce the energy routed to the focusing system in order to reduce the probability of pinhole closure caused by plasma formation at the edge of the pinhole. A calorimeter located behind the first wedge, calibrated against the output of the amplifier, provided a reference energy measurement to be compared with the second calorimeter located behind the pinhole in the focusing system. For pinhole diameters of $200 \mu\text{m}$ and greater, 1 percent of the amplifier energy was routed to the focusing system, and for pinhole diameters less than $200 \mu\text{m}$,

Fig. 3. Focal spot intensity distribution.

Fig. 4. Percentage of total energy contained vs radius in the focal spot.

0.16 percent was used. The focusing system consisted of an off-axis-parabolic mirror, a pinhole, and a calorimeter. These elements were placed in a chamber evacuated to 2×10^{-6} torr to prevent air breakdown. The mirror had a focal length of 45.72 cm and pinhole diameters between 57 and 400 μm were used. The pinholes were photo-etched in 80- μ -thick stainless steel foil. For these measurements the energy from the oscillator amplifier chain was ~ 20 J. Comparisons of the readings from the two calorimeters, with and without the pinhole in place, provided the percentage of transmission. Final axial positioning of the pinhole relative to the focusing mirror was accomplished by examining burn patterns on the surface around the pinhole and by using the amplified beam. The optimum pinhole location was slightly different when only the oscillator and amplified beams were used. All measurements were taken at the optimum focus for the amplified beam.

Figure 5 shows the percentage energy transmission vs pinhole diameter. Curve a gives the calculated energy transmission for a diffraction-limited beam of uniform intensity. This curve was obtained by integrating the Airy pattern and corresponds to a spot size (84-percent transmission) of 57 μm . Curve b, defined by the open circles, is the measured energy transmission from the oscillator-preamplifier system through the unpumped amplifier and associated optics. The spot size for the oscillator-preamplifier system was 97 μm , corresponding to 1.7 times the diffraction-limited spot size. Curve c in Fig. 5, defined by the closed circles, gives the measured energy transmission for the amplified beam. In this case, the spot size was 150 μm , corresponding to ~ 2.6 times the diffraction-limited spot size. The observed degradation in the amplified beam is apparently caused by the amplifier medium, although it is possible that plasma formed at the pinhole periphery could have been responsible for the reduction in transmission when the more intense amplified beam was investigated. Furthermore, thermally induced wavefront distortion at the surface of optics exposed to the high power beam cannot be ruled out in

these experiments. At any rate, these data demonstrate that near-diffraction-limited beams can be extracted from high-pressure H_2-F_2 amplifiers and that the output of the HF laser system can be focused to the small diameter required for laser fusion.

Fig. 5. Energy transmission as a function of pinhole diameter.

Energy extraction versus input power was measured with the SF_6 -HI oscillator-preamplifier system and compared with energy extraction obtained with an H_2-F_2 oscillator. Figure 6 shows the curve obtained with the H_2-F_2 oscillator¹ to which has been added data points obtained with the SF_6 -HI system. Within experimental error, the extraction efficiency was the same with the SF_6 -HI spectrum as was obtained when the full H_2-F_2 spectrum was used. This fact, in conjunction with the observation that the amplified SF_6 -HI spectra contained 25 lines whereas the amplified H_2-F_2 spectra consisted of 46 lines, suggests that rotational relaxation times in the amplifier are short compared to the pulse width. Plans are being made to add an additional amplifier to the system so that extraction experiments using the SF_6 -HI

produced spectrum can be extended to higher intensities.

Fig. 6. Amplifier output as a function of input intensity.

Short Pulse Option for Electric-Discharge-Initiated SF_6 -HI System

Previous reports have discussed the development of an electric-discharge-initiated oscillator-preamplifier system which uses SF_6 -HI gas mixtures and whose spectrum matches fairly well the spectra produced by electron-beam initiation of H_2-F_2 mixtures. Recent experiments have demonstrated that the HF spectrum produced by using an SF_6 -HI system is just as efficient as that from a H_2-F_2 system in extracting energy from the Phoenix amplifier system. One of our program milestones during this reporting period was to modify the discharge oscillator-preamplifier system so that short (5 ~ 25 ns) pulses could be produced. This modified system will be used to generate short pulses so that short-pulse generation and amplification can be realized in HF laser systems. In this section we will discuss the development of this short-pulse system. The basic system is the same as the one that produces 100-ns pulses, but a pulse cutting system and a 5.0-cm-diameter final amplifier were added. This larger amplifier is discussed elsewhere in this report.

The system that will be used to deliver a 25-ns pulse to the PI-initiated HF amplifier is shown in Fig. 7. The beam from the oscillator, which is polarized, is put through another polarizer to clean up the beam. The beam is then run through a CdTe, EO crystal (8 in x 8-m x 40-mm). During a time when the pulse is on, the polarization can be rotated 90 degrees by applying the proper voltage. The beam is then expanded and spatially filtered. After the beam expander, the beam passes through the 2.5-cm amplifier and is then expanded 2X in a beam expander that is constructed from reflecting optics. The beam is then amplified by the 5.0-cm-diameter amplifier. The long pulse consisting of both polarizations has been amplified by both amplifiers. In the next set of planned experiments, a pulse whose width matches the burn time of the H₂-F₂ amplifier (~25 ns) will be cut out of this oscillator-amplifier system and will be amplified by the H₂-F₂ amplifier. For these experiments, the polarization switchout plate will be placed after the discharge system but before the H₂-F₂ amplifier.

Fig. 7. Schematic diagram of the oscillator-preamplifier system currently being used to deliver a short pulse to the PI-initiated amplifier.

The oscillator-amplifier system is currently in operation, and experiments to measure the contrast ratio have been performed by using a 10-ns pulse-forming line for the rotator. These experiments have concentrated on the generation of a short pulse, and very little time has been spent on the measurement of the spatial quality of the final beam. Contrast ratios from a quartz brewster-angle polarization rotator of 100 to 1 have been measured (peak power in short pulse/peak power in foot). A 10-ns-wide cutout pulse and the prepulse are compared in Fig. 8. This experiment has demonstrated that we can generate and amplify a short HF laser pulse having acceptable contrast ratios.

Fig. 8. A short pulse is compared to the foot from the long pulse.

Oscillator-Amplifier Studies Using the PI-Initiated IIF Amplifier

In order to better characterize the oscillator that is driven with the Physics-International Pulserad-110A electron-beam accelerator, radially resolved electron-beam energy deposition measurements were made. This system is configured in the π geometry. The electron-beam energy transmission through the gas was measured with a 31-element carbon calorimeter. This totally stopping calorimeter yields information about the radial distribution as well as the total transmitted energy of the electron-beam. Nitrogen at the same mass density as the $F_2/O_2/H_2$ densities was used in these measurements. Fluorine could not be used because it reacts with the carbon in the calorimeter.

The transmitted electron-beam energy was measured as a function of distance from the accelerator anode. Measurements were made in the laser cell at three different locations and at four different pressures. The amount of energy deposited in the gas is inferred from these data by subtracting the amount of electron-beam energy that is transmitted through the active region from the value of electron-beam energy that is transmitted into the active region. The results of these measurements are given in Table 1.

TABLE 1

Deposition Energy as a Function of Pressure

Equivalent $F_2/O_2/H_2$ Pressure (torr)	N_2 Pressure (torr)	Energy Deposited in Cell (joules)
300/90/75	537	42
400/120/100	716	60
500/150/125	895	84
700/210/175	1253	119

These results are in good agreement with previous results.

The calorimetry data have also been used to infer a radial deposition profile. This was done by dividing the calorimeter into a central zone with an area of 0.706 cm^2 and three annular zones, each with an area of 7.06 cm^2 . This left a zone near the wall of the laser tube with an area of 9.78 cm^2 in which no measurements were made. An attempt has been made to extrapolate the data into this region. Comparison of the results from elements comprising a zone (each zone except the first contained 10 calorimeter elements) indicated that angular variations were not large. Here again, the energy deposited in each zone was taken to be the difference between the electron-beam energy transmitted into the active region and the electron-beam energy transmitted out of the active region.

The resulting radial deposition profile is shown in Fig. 9. These results are for a nitrogen pressure equivalent to an $F_2/O_2/H_2$ mixture of 400/120/100 torr. The radius of the cathode used in the diode of the accelerator was 1.3 cm. Radial deposition profiles have been obtained for other equivalent pressures, and they are very similar to the results shown in Fig. 1. The zones beyond a radius of 2.2 cm show a negative deposition because scattering into and out of a zone cannot be accounted for in the transmission measurements. In fact, if there is more scattering out of a zone than there is scattering into a zone, as is the case for the first two zones, the energy deposition in these zones inferred from the transmission measurements is too high. Likewise in the outer zones, the transmission measurements imply a negative deposition because the scattering into these zones exceed the scattering out plus the energy deposited. This uncertainty is inherent in these type of measurements and cannot be corrected except by a two-dimensional Monte-Carlo calculation with superimposed magnetic field.

While this radial deposition profile is not very accurate because of the scattering problem, it does indicate that the excitation for this particular system is highly non-uniform. In fact, most of the electron-

beam energy is deposited in only about 25 percent of the active volume of the laser cell.

Fig. 9. PIFI radial deposition profile measurements made at an equivalent N_2 pressure equal to 400-torr F_2 , 120-torr O_2 and 100-torr H_2 .

The effect of the nonuniform deposition on calculations made with the computer model for the HF laser will be discussed in a later section of this report.

With this nonuniform radial deposition, one would expect the laser output to also exhibit a radial nonuniformity. The optical cavity of the oscillator consisted of a CaF_2 flat and a 20-m radius of curvature copper mirror. The cavity mirror separation was 185 cm. The oscillator output energy density radial distribution was determined by using various size apertures and a calorimeter. The resulting oscillator output distribution is shown in Fig. 10. This distribution is in general agreement with the radial distribution of deposited electron-beam energy as is shown in Fig. 9. These results confirm that the electron-beam radial deposition distribution inferred from electron-beam transmission measurements are qualitatively correct.

Fig. 10. Radial energy density distribution from PIFI oscillator. Cavity consisted of a CaF_2 flat and a 20-m radius of curvature copper mirror. Length of the active medium is 92 cm.

As a consequence of this radially nonuniform energy deposition, the production rate of atomic fluorine and, hence, the production rate of excited HF molecules will also have a radial variation. If one assumes that the electron-beam deposition can be represented by a 3-cm-diameter region with large deposition and that the deposition outside this region is much smaller, then the temporal behavior of these regions should be different. Measurements of the temporal behavior of these two regions have been made. The results for a mixture of $\text{F}_2/\text{O}_2/\text{H}_2$ equal to 400/120/100 torr are shown in Fig. 11. The solid line indicates the temporal behavior of the 3-cm-diameter inner region of the oscillator. The dotted line indicates the temporal behavior of the region outside of 3-cm diameter. The line marked sum is the equivalent sum of these two signals, taking into account that the area in the outer region is larger. Note that the signal from the outer region does not indicate that an appreciable amount of energy is being produced from a slow production of excited HF.

Fig. 11. Temporal behavior of the oscillator for the inner and outer regions. The gas mixture was: $F_2/O_2/H_2 = 400/120/100$ torr.

In Fig. 12, the same results are shown for a mixture of $F_2/O_2/H_2$ equal to 400/120/20 torr. With this hydrogen lean mixture, the production rate of excited HF molecules should be slower than in the case above. In this case, the core region temporal behavior does not exhibit a radiation pulse much different from the previous case. But, in this case, the region outside 3 cm does show evidence of a slow production of excited HF molecules.

Results obtained from amplifying an input signal by this system should give a better understanding of the effects of radial non-uniform energy deposition. The results from the oscillator measurements and amplifier experiments, when available, will be compared with the results from the HF computer model.

Fig. 12. Temporal behavior of the oscillator for the inner and outer regions. The gas mixture was: $F_2/O_2/H_2 = 400/120/20$ torr.

Energy Extraction from PI-Initiated HF Amplifier

Preliminary energy extraction experiments using the Physics-International Pulserad 110A driven amplifier have been performed. The input pulse in these experiments was obtained from the electric-discharge-initiated oscillator-preamplifier system and was much longer ($\tau_{\text{pulse}} \approx 150$ ns) than the duration of positive gain in the amplifier. Measurements have been made as a function of pressure in the amplifier for both an SF_6 -HI and SF_6 - C_2H_6 input spectrum. For the SF_6 -HI spectrum, the input power was estimated to be about 7×10^5 watts/cm² and for the case of the SF_6 - C_2H_6 spectrum, the input power was about 1×10^6 watts/cm². The results of these preliminary experiments are shown in Fig. 13 as a function of F_2 pressure in the amplifier. All measurements were made with a mixture of $F_2/O_2/H_2$ equal to 1.0/0.30/0.25. The energy

output when the system is configured as an oscillator is shown for comparison. The data in Fig. 5 show that at these power densities, approximately 35 percent of the energy was extracted at a P_2 pressure of 300 torr and 63 percent was extracted at 600 torr. The results, using an SF₆-C₂H₂ input spectrum, are larger than expected and will require additional measurements to confirm and understand.

Fig. 13. Energy extracted as a function of P_2 pressure. All measurements were made with a gas mixture of $F_2/O_2/H_2 = 1.0/0.20/0.25$ torr. The energy extracted from the amplifier for two different input spectrums is shown. Also shown is the energy obtained when the system was configured as an oscillator.

Fast-Discharge-Initiated Preampifier for the SF₆-HI System

The electric-discharge-initiated oscillator-preampifier system that was described in the last semiannual report¹ made use of

amplifiers that were excited by an electric discharge between two Rogowski shaped electrodes. The maximum separation between these electrodes was 3.75 cm. The volume of this amplifier, which was the final amplifier in the chain, was 3.7 cm x 3.75 cm x 50 cm. A three-stage Marx was used to excite the discharge.

In order to obtain larger energies and higher powers, this discharge amplifier design was scaled to 5.0 cm x 5.0 cm x 50 cm. The electrode system to drive this scaled amplifier was a four-stage Marx with four 0.04- μ F capacitors which produce an initial voltage of 140 kV across the discharge region. Previous amplifiers were triggered on at least two of the spark gaps. In this amplifier, it was found necessary to trigger only the gap closest to ground. This electric circuit for this scaled amplifier is shown in Fig. 14.

Fig. 14. Discharge circuit for 5.0-cm amplifier.

The amplifier was tested as an oscillator in order to determine if the energy scaled as expected. The system was studied using the previously determined optimum mixes of SF₆-C₂H₂ (15:1) and SF₆-HI (20:1). The results of this study are shown in Fig. 15.

These results, along with the determination of the discharge uniformity, indicate that the design was successfully scaled to 5.0 cm. One point of interest is that with this amplifier, the energy obtained from $\text{SF}_6\text{-C}_2\text{H}_6$ and $\text{SF}_6\text{-H}_2$ mixtures were almost equal. Past experience with this type of amplifier would have indicated that one-third to one-half as much energy would be obtained with $\text{SF}_6\text{-H}_2$ as is obtained with $\text{SF}_6\text{-C}_2\text{H}_6$. When this amplifier is used in the total system, about 2.0 J of energy can be extracted in a high-quality beam. This amplifier is now currently in use with the short-pulse oscillator-amplifier system.

Fig. 15. Comparison of energy out of amplifier as a function of total pressure.

Numerical Modeling of HF Laser Performance

In the last semiannual report,¹ kinetic upgrading of Sandia's HF chemical laser computer model was described. The upgraded code was shown to be in reasonable agreement with other codes and experiment for two long-pulse cases in which the gas mixture contained a large amount of inert diluent. However, it did not appear that the upgrading remedied the problem of energy and pulse width overprediction for electron-beam-initiated cases of interest at Sandia.

Now, calculations made at Aerospace Corporation confirm that the overprediction is not an aberration of the Sandia model alone. The difficulty is summarized in Fig. 16: for the usually-quoted cost (in electron-beam-deposited energy) of 10 eV per F atom, the pulse width is overpredicted by more than a factor of six, and the output energy is overpredicted by nearly a factor of two. In order to predict the pulse width correctly, it is necessary to use an unrealistic F-cost, approaching the thermodynamic limit. Then, the energy overprediction increases to about a factor of three.

Fig. 16. Effect of cost-per-F assumption on REBA-experiment calculations.

There is now evidence that part of the difficulty may be related to deposition non-uniformity. In another section of this report, entitled Oscillator-Amplifier Studies Using the PI-Initiated HF Amplifier, electron-beam transmission measurements at various radii in the Physics International Pulserad-110A initiated laser are presented.

These measurements imply a much stronger-than-average initiation along the laser tube axis, and a weaker-than-average initiation in the outer annulus. Composite solutions have been constructed by suitably adding together the predictions of the computer model for such inner and outer regions. Figure 17 shows the overprediction problem for the PI-initiated laser and the significant improvement obtained from a composite solution. A narrower pulse width and smaller output energy are predicted, in fair agreement with experiment, at a reasonable ϵ -cost of 1.0. Investigation of deposition-nonuniformity effects in the PI-initiated laser is continuing.

Fig. 17. Effect of cost-per-F assumption and composite solution for PIP-initiated experiment calculations.

Another investigation of deposition-nonuniformity effects is under way for the Febetron-706 experiments that were reported in the last two progress reports.^{1,3} These experiments were designed to determine III laser pulse widths at pressures up to 10 ATM. A 2.5-ns (FWHM) electron-beam pulse was used to initiate III production in volumes on the order of a cubic centimeter, for $F_2/H_2/O_2$ mixtures with composition typically 1.0/0.25/0.2. For the comparisons reported here, a cavity was formed with a 95-percent reflecting mirror and a 60-percent reflecting coated ZnSe flat output coupler.

The results of the Febetron-706 experiments differ in three important respects from all of the other electron-beam-initiated $F_2/H_2/O_2$ experiments at Sandia. First, the output energy densities are on the order of 1.3 J/l-atm, more than a factor of 10 better than was obtained in the other experiments. Second, the output energy and pulse shape are sensitive to the O_2 partial pressure, with energy and pulse width decreasing as the O_2 fraction increases beyond some optimum. And finally, pulse shapes have more structure and, in many cases, much greater widths than in the other experiments.

In order to economically compute laser performance for active volumes as small as 1 cm³, it has been necessary to modify the IIF code. The implicit method of characteristics previously employed, in order to treat spatial variations along the optical axis of oscillators and amplifiers, takes time steps equal to the photon transit time between spatial grid points. For very short active-medium volumes, this becomes over restrictive. Therefore, a one-point option has been added, which calculates volume-averaged quantities, not unlike Aerospace's code SPIKE. This option has been used in all of the Febetron-706 comparisons reported here. Of course, this simplified model would not be applicable to amplifier modeling.

So far, the model predictions have been compared with a single set of Febetron-706 results, taken with 800 torr F_2 , 240 torr He , and 100 to 200 torr O_2 . From the last progress report, the initial F/F_2 ratio was expected to be 0.1 percent or more (0.05 percent dissociation). The model prediction is compared with experiment in Fig. 18. It is seen that the energy and pulse width are greatly overpredicted, and the pulse-onset is too early. A revised estimate of F_2 dissociation, which takes into account the particular electron-beam voltage and window used in these cases, is $\sim .01$ percent. Figure 19 shows the improved prediction of energy and onset time that occurs. However, the pulse width is still too short. Moreover, much of the calculated output energy is attributed to $v = 1$ P-branch radiation (P_1), which apparently is not observed. Because of the large uninitiated volume of gas within the laser cavity, trace quantities of HF formed during mixing can prevent oscillation on P_1 . A simple estimate shows that 0.04 percent will suffice, even neglecting production of HF ($v = 0$) during the electron-beam-initiated reaction. Therefore, P_1 radiation was suppressed in the remaining calculations. Figure 19 shows the predicted output pulse with P_1 suppression. Also shown is the result when the O_2 chain-terminating reactions

Fig. 18. Comparison of Febetron-706 experimental and calculated intracavity flux and output energy, assuming 0.05-percent F_2 dissociation.

are suppressed. The pulse remains too short. No other reasonable variation of kinetics data was found which could lengthen the pulse without causing a large energy overprediction.

Fig. 19. Calculated intracavity flux and output energy for the Febetron-706 experiment assuming 0.01-percent F_2 dissociation.

Finally, account was taken of electron-beam nonuniform deposition. In the Febetron-706 experiments, the optical axis of the cavity is transverse to the electron-beam axis. Because of the electron-beam spreading (as determined by cellophane dosimetry), the deposited energy density decreases in the direction of electron-beam propagation. Composite solutions have been constructed which crudely account for this nonuniformity. The most important result of this treatment is that the observed long tail on

the output pulse is now predicted and originates in the region of lower initiation. Solutions for O_2 partial pressures of 100 and 200 torr are shown in Fig. 20. For these cases, the HO_2 and FO_2 formation rates were reduced by a factor of four from the values supplied to Physical Sciences, Inc. Such reductions are not unreasonable since the FO_2 formation model is not complete and some third-body efficiencies for HO_2 formation are only approximations. The output pulses can be partially characterized by the five quantities plotted versus O_2 pressure in Figs. 21a-21e. The agreement between experiment and prediction is satisfactory, in view of the many remaining uncertainties. Comparisons at the other experimental pressures remain to be made. Tentatively, it appears that the explanation of results in terms of O_2 chain-terminating reactions, made by G. N. Hays in previous progress reports, is borne out by the composite solutions from the present HI computer model.

Fig. 20. Composite solutions (accounting for nonuniform deposition) for the Febetron-706 experiments.

Fig. 21a. Comparison of measured and calculated delay times for the Febetron-706 experiments.

Fig. 21b. Comparison of measured and calculated peak-to-peak times for the Febetron-706 experiments.

Fig. 21c. Comparison of measured and calculated pulse-tail lengths for the Febetron-706 experiments.

Fig. 21d. Comparison of measured and calculated peak power ratios for the Febetron-706 experiments.

Fig. 21e. Comparison of measured and calculated output energy for the Febetron-706 experiments.

In conclusion, deposition nonuniformity has been shown to be an important factor in modeling two electron-beam-initiated experiments. When taken into account, previously noted discrepancies between experiment and prediction are significantly reduced.

Pyroelectric Vidicon System for Beam Diagnostics at 2.8 μm

A number of past and proposed diagnostics for the HF laser require the determination of intensity spatial profiles. Among these are focal spot intensity distribution, beam uniformity before focusing, and beam profiles at various points in an oscillator-amplifier chain. A pyroelectric vidicon system has been acquired which will greatly simplify many of these determinations. Past operation of this particular system has been confined to the wavelength region around 10.6 μm ; therefore, some preliminary experiments were performed to check the operating characteristics at 2.8 μm .

A block diagram of the infrared imaging system is shown in Fig. 22. The vidicon tube surface material is etch-thinned triglycine sulfate (ETTGS) with a spectral range from 2 to 400 μm (Amperex Electronic Corporation). As seen in the diagram, information is recorded on a single-shot basis in two ways (simultaneously): (1) line selected data (i.e., up to 9 separate scan lines out of the 525 lines of information) can be recorded as analog signals on an oscilloscope trace, and (2) all 525 lines are stored as a full frame on a disc-storage device. Both of these mechanisms are triggered synchronously with the laser pulse. In addition, a chopped cw-beam can be viewed in real time on the TV monitor. In the pulsed mode, and subsequent to the laser shot, the full frame data can be analyzed in detail as analog signals obtained through the video analyzer unit or displayed continuously from memory on the TV monitor. This image can then be photographed to provide an approximate view of the stored intensity profile. The overall system is also adaptable to computer storage and subsequent analysis, although this technique has not yet been pursued.

Fig. 22. Block diagram of infrared imaging system.

Measurements were made by using the electric-discharge-initiated HF oscillator-preamplifier system described in our last semiannual report.¹ This system has been characterized and provides a near diffraction-limited TEM₀₀ pulse. Figure 23 is a profile of the oscillator pulse taken using method (2). The data points were taken from the recorded oscilloscope trace. The input energy density for this measurement was 2 mJ/cm². The near Gaussian pulse shape agrees well with earlier determinations obtained by scanning the near- and far-field profile of the beam at the rate of one data point per shot. Further experiments with a beam attenuation of approximately 20 gave identical results, indicating a linear operating regime from 2 mJ/cm² to at least 100 $\mu\text{J}/\text{cm}^2$ in this mode.

In a second experiment, an estimate of the spatial resolution of the vidicon was obtained by imaging the expanded and recollimated oscillator pulse on an 8.5 mm aperture placed 93 cm from the camera. A photograph of the stored intensity profile is shown in Fig. 24. The calculated Fresnel number for this configuration is 7. The spacing of the first two maxima is about 700 μm and the width of the first minima is about 200 μm . Other recordings taken to date include a shear plate interferogram picture of the oscillator-preamplifier pulse.

Fig. 23. Single-shot profile of HF laser oscillator pulse obtained with pyroelectric vidicon.

Fig. 24. Fresnel diffraction pattern of an 8.5 mm-diameter aperture using the expanded and recollimated III oscillator pulse. This is a photograph of the image stored on the disc (Fig. 1).

Although the disc storage-video analyzer system has not yet been tested for linearity over its stated operating range, the complete system is clearly a valuable diagnostic device for future III applications.

IODINE LASER PROGRAM

Overview

The iodine laser is the only proven laser that can satisfy the high-average-power high-optical-quality requirements of a laser-fusion power generator. However, its efficiency needs to be improved. Sandia is responsible for evaluating whether this laser can be developed to meet the requirements for a laser-fusion power-generation plant. In this report we summarize studies that show that the optical characteristics of gain-saturated iodine amplifiers are acceptable for laser-

fusion applications. Also, we now have available a code that can be used to project large-system performance. Our program has turned to studies that will lead to increased system efficiency. We are making steady progress in devising technology for efficient chemical recovery of laser-medium starting materials and for improved electrical efficiency; the large number of alternatives gives us confidence that a greater than 1-percent overall system efficiency can be realized.

SAIL-1 Output Beam I asefront for Un-saturated and for Satu ated Output Beams

Lateral shearing interferograms of both unsaturated and saturated output beams from the SAIL-1 atomic iodine laser have been taken at a distance of 10.4 m beyond the main amplifier. These are shown in

Fig. 50, Ref. 1. The image is recorded with an ITT proximity-focused photodiode followed by an image intensifier. The response of the S-1 photocathode at $1.315 \mu\text{m}$ has been maximized. While an absolute calibration is not necessary to analyze the lateral shearing interferograms, the entire photodiode-intensifier system has been carefully calibrated for use with Polaroid Type 57 and Royal X Pan Type 4166 photographic film.⁴ This is essential for the focal spot photographs discussed below, since S-1 photocathodes are nonlinear at $1.315 \mu\text{m}$.

The radius of curvature of the wavefront can be determined² by measuring the rotation angle of the fringes with respect to a reference line (which can be seen in both photographs in Fig. 50, Ref. 1). The radius of curvature inferred for the unsaturated output beam is $22 \pm 1 \text{ m}$, which is the same as the radius of curvature of the oscillator wavefront at this distance. The radius of curvature in the wings of the saturated output beam is also $22 \pm 1 \text{ m}$, but the radius of curvature on-axis, where the saturation is greater than in the wings, has grown to $27 \pm 1 \text{ m}$. This indicates that under these conditions, there is a small amount of whole beam self-focusing, since the wavefront is now less convex. The self-focusing is a direct result of a gradient in the index of refraction caused by gain-saturated anomalous dispersion.⁵⁻⁸ The on-axis peak power is $\sim 4 \text{ GW/cm}^2$ for the saturated output beam. While no quantitative measurements of the intensity profile can be made from Fig. 50, Ref. 1, it can be seen that the saturated output beam diameter has grown by about 50 percent compared to that of the unsaturated output beam. This is simply due to the fact that there is more gain in the wings of the saturated beam than on-axis.

Figures 25 and 26 show the radius of curvature and intensity of the wavefront as a function of radius calculated with the ISTAR beam pro-

pagation code⁹ for an unsaturated and a saturated output beam, respectively. The unsaturated output beam has a radius of curvature that is almost independent of radius and has a value of 22 m , in agreement with experiment. The radius of curvature of the saturated output beam is 26 m on-axis and 21 m in the wings of the beam, also in good agreement with the experiment. We believe that the rise in the radius of curvature in the far wings of the saturated output beam is real, but unimportant, due to the lack of flux in this region. One additional point is that ISTAR also predicts the 50-percent increase in beam size.

Fig. 25. Calculated radius of curvature (R_c) and energy flux (F) of an unsaturated output beam at a distance of 10.4 m beyond the main amplifier of SAIL-1.

Fig. 26. Calculated radius of curvature (R_c) and energy flux (I) of a saturated output beam at a distance of 10.4 m beyond the main amplifier of SAIL-1.

Focal Spot Determination -- The question arises as to how well the above unsaturated and saturated output beam can be focused. We have compared photographs of the beam intensity in the actual focal plane of a 1.81-m-focal-length spherical lens to calculations of the beam size from ISTAR. The radii of curvature of the two antireflection coated lens surfaces have been selected to minimize spherical aberrations for the conditions of the present experiment.

Figure 27 shows the propagation through the actual focal plane of a 1.81-m spherical lens of the electric waist radius, w , of an unsaturated and saturated output beam as calculated by ISTAR. For a Gaussian beam, the electric waist radius, w is $\sqrt{2}$ times the intensity radius (e^{-1} and e^{-2} widths, respectively). The lens is included by adding a spherical phase shift to the wave front at the position of the lens. Spherical aberration effects are

completely negligible since the effective F/λ number is greater than 100.

Fig. 27. Calculated electric field waist radius w for an unsaturated and a saturated output beam from SAIL-1 as a function of distance z from a 1.81 m focal length lens.

it can be seen in Fig. 27 that the saturated output beam is predicted to focus to a smaller spot size than the unsaturated output beam. The reason for this is that the saturated output beam grows in size, thus reducing the diffraction limit. The degrading effects of the phase distortion in the saturated output beam are insufficient to overcome the smaller diffraction limit. ISTAR predicts that the focal spot intensity diameter (FWHM) for the unsaturated and saturated output beams should be 170 μm and 130 μm , respectively.

Figure 28 shows photographs of the intensity profile of an unsaturated and a saturated output beam at a distance of 2.0 m from the lens, where both output beams have a minimum focal spot size. The ITT proximity-focused photodiode has been used with Royal X Pan Type 4166 film to extend the dynamic range of the entire imaging system to ~ 50 . Photographs have been taken at numerous distances, z , from the lens in

order to determine the plane of minimum spot size. The photographs in Fig. 28 were taken in this plane. The image is magnified 26.1 times by a 32-mm microscope objective placed appropriately between the focal plane and the photocathode of the photodiode. It is interesting to note that the focal spot size of the saturated output beam is smaller than that of the unsaturated output beam (as predicted above by ISTAR). Both output beams have their respective minimum in the same plane to within the resolution of the imaging system, which is ~ 25 μm .

Fig. 28. Photographs and contour maps of an unsaturated and a saturated output beam from SAIL-1 in the true focal plane of a 1.81 m focusing lens. The beam diameter (FWHM) calculated by using ISTAR is shown for comparison for both cases.

Also shown in Fig. 28 are contour maps of the photographs. The contours are relative iso-intensity curves of the laser flux itself; the photodiode, image intensifier, and film nonlinearities have been removed. The six rings indicate increments of one-seventh of the peak intensity. The seventh ring, below the detection system threshold, is not shown. The intensity diameter (FWHM) is between the second and third outermost rings. Below each photograph, the intensity diameter (FWHM) predicted by ISTAR is shown for comparison. It can be seen that the agreement with the observed diameters is good.

Fabry-Perot Study of Iodine Oscillator Spectral Bandwidth

Comparisons between theoretical models and experimental data for beam propagation in an iodine laser have been made throughout the course of the laser physics experiments that use SAIL-1.¹ Agreement between theory and experiments has been good, which lends confidence in the use of theory to predict the performance of new iodine laser systems. Two basic assumptions of the theoretical model are that only one of the several possible laser transitions actually contributes to the oscillator output and that the oscillator pulse has a bandwidth limited Gaussian nature. Because these assumptions are important in system modeling, it was decided to verify them by investigating experimentally the frequency content of the output pulse.

Figure 29 is a schematic representation of the experimental setup used for determining the spectral bandwidth of the oscillator output. A pulse train from the oscillator was focused by the input lens and then allowed to expand and fill the mirrors of a Fabry-Perot interferometer. The Fabry-Perot output was relayed with a lens to the photocathode of a recording system consisting of an image converter having enhanced response at 1.315 μm , followed by an image intensifier and a camera. The Fabry-Perot plates had (manufacturer quoted) inside reflectivities of 0.985 and outside reflectivities of 0.005. Data were obtained for two

cavity lengths, i. e. 0.0396 cm and 0.155 cm. The recording system, including the film, has been calibrated for relative response at 1.315 μm in a manner similar to the calibra-

mate, as will be discussed in detail later. The coefficient can be obtained from the geometry of the experimental setup or from the separation of successive fringes obtained by recording Fabry-Perot output when the

Fig. 29. Schematic representation of experimental setup used for determining spectral bandwidth of iodine oscillator output pulse. All distances are in centimeters.

tion of our EPI-512C streak camera, as described in Ref. 4.

Figure 30 shows typical experimental data: the upper portion of the figure shows a photograph of the ring pattern from the interferometer; the lower portion shows a relative intensity contour map obtained by scanning the photograph with a densitometer and converting reflectivities to relative intensities. For the usual Fabry-Perot experimental configuration, the dependence of frequency on position is given by

$$d\nu/dr = \frac{cr}{A\lambda} \quad (1)$$

where ν is frequency, r is the distance of a point on the photograph from the center of the pattern, c is the speed of light, λ is the wavelength for light of frequency ν , and A is a calibration constant for the system. Because the experimental configuration used here is not the standard one, Eq (1) is only approxi-

interferometer is illuminated with a cw He-Ne laser. Both techniques were used here, and the agreement was good. The result for $\lambda = 1.315 \mu\text{m}$, is

$$\frac{d\nu}{dr} = 2.33 \times 10^{11} \text{ sec}^{-1} \text{ cm}^{-1} \quad (2)$$

All data were obtained in configurations for which (1) the fringe width on film was less than the frequency separation of the $F = 3 \rightarrow F = 4$ and $F = 2 \rightarrow F = 2$ transitions, and (2) the frequency range on film was appreciably larger than this separation. In no case was there any indication of contributions from more than one transition, confirming that only the $F = 3 \rightarrow F = 4$ transition lases.

Detailed interpretation of fringe widths is potentially complicated by the fact that the oscillator pulse is short, $\tau_p = 250 \text{ ps}$, and by the fact that the experimental configuration that was used does not allow for mirror illumination from all incident angles at all mirror points. Reference 10 and sources

cited therein consider the response of a Fabry-Perot to a short pulse. We have shown that for a plane wave pulse of arbitrary duration incident onto a Fabry-Perot, the transmitted energy $\mathcal{E}$ is given by

$$\mathcal{E} = \int d\nu \left(1 + \frac{4R}{(1-R)^2 \sin^2 k l} \right)^{-1} \rho_{in}(\nu) \quad (3)$$

where $k = 2\pi\nu/c$ and $\rho_{in}(\nu)$ is the spectral energy density of the input signal. From Eq. (3) it may be shown that

$$\Delta\nu_{film}^2 = \Delta\nu_{FP}^2 + \Delta\nu_{osc}^2 \quad (4)$$

where $\Delta\nu_{film}$ is the full width at half maximum intensity of the pattern on film as determined by using Eqs. (1) and (2), $\Delta\nu_{FP}$ is the instrument FWHM, and $\Delta\nu_{osc}$ is the FWHM spectral width of the oscillator pulse. The approximation, Eq. (4), is essentially an equality if, as for all cases reported here, $\Delta\nu_{osc}$ is much less than the free spectral range of the interferometer, $c/2l$.

Fabry-Perot interferometers are ordinarily used in such a way that light from all possible incident angles illuminates each point on the instrument mirrors. This can be accomplished, for example, by inserting a diffuser plate between source and the interferometer. Intensity considerations precluded this manner of operation here, and this fact complicates the interpretation of ring widths in terms of spectral breadths of the input pulse. It can be shown that if a diverging Gaussian beam is incident onto a Fabry-Perot, the output amplitude V is given by

$$V = \sum_{n=0}^{\infty} R^n \exp \left(i k (Z + 2 l n) + \frac{i k r'^2}{2(Z + 2 l n)} \right) \quad (5)$$

where Z is the distance of the observation point from the waist of the input beam and r'

is the radius of the fringe at the Fabry-Perot mirror. It has not been possible to obtain a closed form for the summation in Eq. (5), but it is clear that only as $r' \rightarrow 0$ does this term reduce that appropriate for the usual experimental configuration.

Analysis of experimental results has been accomplished by determining $\Delta\nu_{film}$ as a function of r and extrapolating to $r = 0$. Figure 31 shows typical results. Linear extrapolation is shown, but other extrapolations were found to result in essentially the same final value of $\Delta\nu_{osc}$ as that reported later in this section. The extrapolated values of $\Delta\nu_{film}$ obtained are $\Delta\nu_{film} = 2.0$ GHz for $l = 0.0396$ cm and $\Delta\nu_{film} = 1.1$ GHz for $l = 0.155$ cm.

Fig. 30. Upper: Typical photographic recording of Fabry-Perot interferometer output. Lower: Densitometer reduction of photograph displayed as contours at relative intensities 5, 6, 8, 10, 12. Length scale relates to image size on film.

Fig. 31. $\Delta\nu_{\text{film}}$ as a function of r for $L = 0.0396$ cm.

Data were taken at two cavity lengths in order to avoid the need for determining $\Delta\nu_{\text{f-p}}$ directly. In accordance with simple theory, it was assumed that $\Delta\nu_{\text{f-p}}$ was inversely proportional to L . Using this relation and Eq (4), $\Delta\nu_{\text{osc}}$ was determined to be 1.8 GHz. For a bandwidth limited Gaussian pulse of temporal FWHM τ , the spectral FWHM bandwidth is $0.44/\tau$. The pulses from the SAIL-1 oscillator have $\tau \approx 250$ ps and consequently have a spectral bandwidth of 1.8 GHz if they have bandwidth limited Gaussian character. The agreement of this value with the experimentally determined one completes the confirmation of the assumptions regarding the oscillator output characteristics made in theoretical modeling efforts.

Metal-Doped Microsecond Xenon Flashlamps

It has been shown by Gusinov^{11,12} that the addition of metal dopants can enhance the uv emission of xenon flashlamps in the spectral region of the atomic iodine laser pump band. However, there is some controversy as to whether the enhancement is due to an increase

in the lamp intensity, or merely due to the addition of a long tail to the emission.¹³ It has been demonstrated that in using the appropriate dopant, one observes an increase in intensity without pulse lengthening.¹ Since the present trend is to develop pump sources of ~ 1 - μ s duration for the atomic iodine laser, the degree and nature of enhancement for 1- μ s metal-doped xenon flashlamps must be investigated.

Work in References 11 and 12 was done with ~ 10 - μ s flashlamps. The overall goal of the present work is to extend the measurements on metal-doped xenon flashlamps down to 1- μ s pulsewidths. An attempt will be made to optimize the xenon pressure and dopant parameters for the lamp geometry used in the experiments. Finally, the overall absolute fluorescence efficiency will be measured under optimum conditions.

Experimental Apparatus -- The apparatus on which the metal-doped lamps are tested is shown in Fig. 32. The 0.218- μ F capacitor is charged to 38.8 kV and is discharged through the test lamp by means of a spark gap. In order to achieve microsecond pulsewidths at a linear energy deposition of ~ 16 J/cm in the lamp, the circuit had to be underdamped. The discharge current consists of approximately four ringing cycles with a period of 0.67 μ s. This implies a circuit inductance of about 50 nH.

Fig. 32. Metal-doped microsecond xenon flashlamp test circuit.

The use of Swagelok® fittings allows rapid changing of the lamp. Teflon ferrules seal to the outer wall of the 10-mm-ID x 1 mm-wall quartz tubing. The oxygen-free copper electrodes are internally connected directly to the vertical oxygen-free copper posts entering the bottom parts of the Swagelok® crosses shown in Fig. 32. The dopant (a metallic powder) is placed into the quartz tube before the lamp is assembled. The tube is evacuated and then back-filled with the appropriate fill pressure of rare gas.

The radiation is recorded photographically on a line perpendicular to the lamp axis by means of a 3/4-in Czerny-Turner spectrograph. A pinhole between the lamp and the spectrograph entrance slot acts as an imaging system. It is planned in the near future to replace the spectrograph with a PARC OMA II which will permit accurate absolute calibrations of the detection system. The radiation is also monitored on a line almost perpendicular to the lamp axis with an S-5 fast photodiode. An interference filter with a central wavelength of 280 nm and a full-width of 20 nm is placed before the photodiode.

Preliminary Results -- It was decided that the first metal dopant to be tried would be Te. This decision was based on the facts that Te has a relatively low vaporization energy and that a tail had not been observed on the emission from Te as had been for several other metal dopants.¹

Preliminary experiments indicate that Te does enhance the uv radiation from an Xe flashlamp. Figure 33a shows the spectrum from a 10-torr-Xe undoped flashlamp. Wavelength is along the horizontal axis and lamp radius is along the vertical axis. It can be seen that most of the radiation at these energies is line radiation. The lines that extend to farther radii have been identified as Si emission lines. The other lines are Xe emission lines. Figure 33b shows the signal from the photodiode. It can be seen that the light pulse has a width close to 2 μ s.

Figure 33c shows the spectrum from a 10-torr-Xe flashlamp to which 100 mg of Te has been added.² The radiation is greater, especially in the region of 280 to 300 nm, than a similar Xe-filled lamp without Te. The silicon lines are in general weaker, because the Te forms a film on the inner wall of the tube that shields the tube from the discharge. One other interesting point is that there is not an underlying continuum as the lines themselves are more dense. Presumably this is caused by radiation from and pressure broadening due to the hot, dense plasma produced when the Te is vaporized and bombarded by the electrons and ions of the initial xenon discharge.¹⁴ If the xenon is replaced by argon the emission is much less. Not only is the uv emission from argon less than from Xe, but the argon filament is also very inefficient in vaporizing the dopant. This may be due to the heavier electron being more efficient at transferring their energy and momentum during bombardment of the Te. Figure 33d is a trace of the photodiode signal for the doped lamps. Apparently, an appreciable fraction of the enhancement (at least in this wavelength region and under these conditions) is due to pulse stretching. However, the pulswidth is still close to 2 μ s.

Experiments are continuing to characterize Te-doped Xe flashlamps. Eventually other metals will be tried. When the spectrograph is replaced by the OMA II, absolute intensity measurements in a predetermined time window will be performed on the optimum doped flashlamp determined from the relative measurements. Reciprocity makes absolute intensity measurements on 2- μ s pulses with the spectrograph and photographic film a difficult and uncertain procedure; in general, only approximate limits can be determined.¹⁵

(a)

(b)

UNDOPED

(c)

(d)

Te- DOPED (100 mg Te)

Fig. 33. a) uv spectrum of undoped 10T Xe flashlamp
 b) photodiode signal at 280 ± 10 nm from undoped 10T Xe flashlamp
 c) uv spectrum of 10T Xe flashlamp doped with 100 mg Te
 d) photodiode signal at 280 ± 10 nm from 10T Xe flashlamp doped with 100 mg Te.

Cl₂ Pump Source for the Iodine Atomic Laser

The search for an efficient pump for an iodine atomic laser has led us to consider the near uv spectral emission of molecular chlorine. Chlorine exhibits two emission band systems in the region of the the absorption bands of the iodine-bearing compounds that are used to produce an atomic iodine laser.

At Cl₂ pressures below 1 torr, a broad-structured continuum has been observed^{16, 17} from 2200 to 3000 Å. This emission can probably be assigned to the

$$D(^1\Sigma_u) \rightarrow A(^1\Sigma_g)$$

transition in Cl₂ analogous to the McClellan bands of I₂.¹⁸ The fluorescence efficiency of a mixture of 0.042 torr Cl₂ and 730 torr of Ar in the region from 2200 to 3000 Å has been estimated as 25 percent, where fluorescence efficiency is defined as energy deposited in the gas mixture divided by energy emitted in the fluorescence band.

At Cl₂ pressures above 1 torr, a discrete band system, peaked at 2580 Å, predominates. It is believed that this band system is analogous to the 3420-, 2920-, and 1570-Å band systems of I₂, Br₂, and F₂, respectively. At one time the I₂,¹⁹ Br₂,¹⁹ Br₂,²⁰ and Cl₂²¹ transitions were all assigned as

$$E(^3\Pi_o + g) \rightarrow B(^3\Pi_o + u).$$

The I₂ and Br₂ transitions have been re-assigned,²² based on spectra of isotopically pure compounds, to be

$$D'(1432, 2_g^3\Pi) \rightarrow A'(2431, 2_u^3\Pi).$$

Molecular chlorine is currently undergoing a similar re-evaluation.²³

The fluorescence efficiencies of the I₂²⁴ and I₂²⁵ band systems have been measured to be on the order of 20 percent when pumped by high-energy electron beams.

These efficiencies prompted the measurement of the fluorescence efficiency of Cl₂ at 2580 Å under similar conditions. In addition, we anticipated laser oscillations from Cl₂, similar to those observed for I₂,²⁶⁻²⁸ Br₂,^{29, 30} and F₂.³¹

Experimental -- A schematic diagram of the experimental apparatus is shown in Fig. 34. A Febetron-705 electron-beam accelerator was used to pump gas mixtures containing a few torr of Cl₂ and 2000 to 5050 torr of He. The electron beam had a peak energy of 1.9 MeV, a current density of approximately 1000 amps/cm² and a pulse width of around 50 ns (FWHM). After exiting the anode of the accelerator, the electron beam was guided by a 4.5 k gauss axial magnetic field through an evacuated drift region. It passed through a 50-μ-thick stainless steel foil and entered the He/Cl₂ gas mixture. Then the beam was turned by the field through 45 degrees into the test cell, which was 1.8 meters long with a 6-cm inside diameter. The actual length excited by the electron beam was 1.4 meters. The portion of the electron beam that was not deposited in the He/Cl₂ gas mixture was turned into the wall of the test cell by Helmholtz coils in order to prevent window damage. A side-viewing port was located 59 cm from the 45-degree turn into the test cell. The helium gas used was obtained from Cryogenic Rare Gases at a stated purity of 99.9995 percent. The chlorine gas was obtained from Airco with a stated purity of 99.96 percent. Gases were used without further purification.

Electron-beam deposition studies were carried out by using a carbon calorimeter having 31 individual segments in a circular array. The fluorescence at the side viewing port and from the end window were each detected with photodiodes. The laser energy output was measured with a surface absorbing calorimeter. The laser mirrors were

dielectrically coated and were located inside the test cell separated by 1.3 meters. In all cases, the back mirror was 98 percent reflective with a 10-m radius of curvature. The reflectance of the flat front output coupling mirror was varied (98%, 75%, 50%, 25% and a sapphire flat). The fluorescence and laser spectra were recorded on Kodak Tri-X film by using a 1-m Spex monochromator in second order with 100 μ slits.

Fig. 34. Schematic diagram of the experimental apparatus used in Cl_2 pump source study.

Results -- The primary objective of the Cl_2 study was to determine the fluorescence efficiency of Cl_2 in the region of the absorption band of iodine-bearing compounds used to produce an atomic iodine laser. In order to do this, the energy deposited in the He/ Cl_2 gas mixture was measured at various distances along the test cell and at a variety of He pressures. An important piece of data was the temporal output of the photodiodes placed behind double apertures. The double apertures defined a cone of view for the photodiodes and allowed us to calculate the number of photons emitted by a rectangular parallelepiped of dimensions 1 cm x 1 cm x l

where l is the length of the column of gas excited by the electron beam along the axis of the photodiode. (The implicit assumption is that one is dealing with an optically-thin medium.) This means that l was 6 cm for the side photodiode and 140 cm for the end viewing photodiode. Figure 35 shows a plot of the sidelight and endlight as a function of He pressure. Sidelight and endlight are reported in photons per cm^2 (integrated along the column length). It should be noted that both the sidelight and endlight continued to increase over the pressure range used. At 2050 torr of He, only 50 to 60 percent of the electron beam making the turn into the test cell was deposited in the 140-cm-long excitation region. Pressure safety requirements did not allow us to go to higher He pressures. Figure 36 shows a plot of the sidelight and endlight as a function of Cl_2 pressure. Both sidelight and endlight peaked at relatively low Cl_2 pressures (2 to 4 torr) for the highest He pressure possible.

Fig. 35. Plot of sidelight and endlight vs He pressure at a constant Cl_2 pressure of 2 torr.

Fig. 36. Plot of sidelight and endlight versus Cl₂ pressure at a constant He pressure of 9050 torr.

A fluorescence efficiency of 5 $\frac{1}{2}$ percent was calculated at 2 torr of Cl₂ and 9050 torr of He. The efficiency was constant with He pressure at a Cl₂ pressure of 2 torr. After measuring the fluorescence efficiency, dielectrically coated laser mirrors were installed. At 2 torr of Cl₂ and 9050 torr of He, using a 98-percent reflecting back mirror and a front mirror the relativity of which varied from 25 to 50 percent, we obtained a constant 96-mJ output in a 3.75-cm-diameter beam. This corresponded to a laser efficiency of ~0.4 percent. The dielectrically coated front mirror was then replaced with an aligned sapphire flat (~4 percent reflectance) and the laser energy dropped to 36 mJ. As a matter of interest, the laser energy output was measured as a function of Cl₂ pressure at 9050 torr of He (see Fig. 37). The laser energy output peaked at the same Cl₂ pressure (2 torr) as the fluorescence. The laser spectrum of a mixture of 2 torr of Cl₂ and 9050 torr of He and the fluorescence spectrum of 2 torr of Cl₂ and 4140 torr of He were recorded as shown in Fig. 38.

Fig. 37. Plot of laser energy output vs Cl₂ pressure at a constant He pressure of 9050 torr.

Fig. 38. The laser spectrum of a mixture of 2 torr of Cl₂ and 9050 torr of He. The fluorescence spectrum of a mixture of 2 torr of Cl₂ and 4140 torr of He.

The fluorescence efficiency of 5½ percent, using longitudinal electron-beam excitation to excite He/Cl₂ mixtures, is too low to make Cl₂ an attractive pump for an atomic iodine laser. It has been proposed²⁶ that ion-ion recombination, not energy transfer from a metastable inert gas, is responsible for the formation of these excited halogen molecules. If this is the case, then a chlorine-bearing compound that has a large electron attachment rate may be the key to obtaining an interesting fluorescence efficiency. Both CCl₄ and HCl are candidates for this new set of experiments.

CF₃I Absorption Spectrum Modification

The efficiency of the atomic iodine photo-dissociation laser when pumped by broad band emitters is limited by the degree of spectral overlap between the emitter and the absorption profile of the parent compound used as a source of excited iodine atoms. One possible way to increase the efficiency is to broaden the absorption spectrum of the parent compound by adding vibration energy to the molecule; this can be accomplished by thermal heating or by absorption of IR photons in a vibrational mode of the molecule.

Pummer et al.³² have investigated the broadening of the ultraviolet absorption band of CF₃I centered near 270 nm following absorption of CO₂ laser photons in the ν_1 vibrational mode of CF₃I at 9.6 μ m. In that experiment, the absorption of 350 nm Kr⁺ ion laser light by CF₃I in a 180-cm-long cell was monitored for a pressure range of 0.03 to 3.0-torr CF₃I. The transmittance at 350 nm is 100 percent for ground state CF₃I; however, when the cell was irradiated with the R(14) line of the 9.6 μ m CO₂ laser transition at a power level of 2.5×10^5 W/cm², a reduced transmittance of ~87 percent was observed for 1-torr CF₃I. This corresponded with an average absorption of 1 to 2 CO₂ laser photons per molecule in the ν_1 vibrational mode of CF₃I. The results of Ref. 32 suggest that broadening of the CF₃I absorption spectrum has occurred by vibrational excitation; however, no quantitative number is given to indicate the extent of the broadening. A

knowledge of the modified absorption profile would be required to obtain a broadening factor from their results.

Because of the potential significance of this effect on the atomic iodine laser efficiency, we have continued this investigation into the broadening of the CF₃I absorption spectrum by vibrational excitation. We have extended the work of Ref. 32 to include measurement of the complete absorption profile of CF₃I, with and without CO₂ laser irradiation. Figure 39 is a schematic representation of the experimental apparatus used in this investigation. The single line output of a Lumonics T103 CO₂ laser is directed along the axis of a 5-cm-long 2.5-cm-diameter Germisil quartz cell fitted with NaCl windows. Prior to passing through the cell, a portion of the CO₂ radiation is deflected to a calorimeter by a Germisil flat to monitor the shot-to-shot relative energy of the CO₂ laser pulse. The flashlamp produces a 10- μ s FWHM pulse of light from a capacitor discharge circuit of 4.4 μ Fds charged to 15 kV. Light from the flashlamp passes through the cell and is reflected off the Germanium flat through a collection lens to the entrance slit of a 1-m spectrograph. Flashlamp light is also directed through a 1/4-m Jarrel-Ash monochromator to an S-5 photocathode which records the relative flashlamp intensity. The central wavelength of the 1/4 m monochromator was set to match the central wavelength of the spectrograph. The ~50-nm output of the spectrograph is recorded on Kodak Tri-X Pan 4164 film. The film was calibrated at 270 nm in the system by using the flashlamp as a source to generate a response versus exposure curve.

For each set of experimental conditions, a spectrum was recorded for an empty cell, a cell with the appropriate CF₃I pressure, and a cell of the same CF₃I pressure with CO₂ laser irradiation. The film records were then scanned on a microdensitometer, yielding plots of the light transmitted through the cell for the various conditions. Figures 40 and 41 show typical microdensitometer traces for the flashlamp light transmitted through an empty cell and

through 10-torr CF_3I without and with CO_2 irradiation, respectively. Using the film calibration curve, the absorption profile of CF_3I can be constructed (shown in the lower part of the figures). The FWHM for the CF_3I without CO_2 irradiation is in good agreement

with that determined more conventionally.⁴⁰ It should be noted that the broadened absorption profile with CO_2 irradiation is asymmetric with more broadening to longer wavelength. This was observed in all broadened spectra.

Fig. 39. Schematic diagram of the apparatus used to monitor the broadening of the CF_3I absorption spectrum by CO_2 laser invitation.

Fig. 40. Microdensitometer records of the flashlamp light transmitted through (a) an empty cell (b) 10 torr CF_3I without CO_2 laser irradiation. Lower part of figure is absorption profile of CF_3I calculated from the two upper traces.

Fig. 41. Microdensitometer records of the flashlamp transmitted through (a) an empty cell (b) 10-torr CF_3I with CO_2 laser irradiation. Lower part of figure is absorption profile of CF_3I calculated from the two upper traces.

The CF_3I pressure, the CO_2 laser line, and the CO_2 laser power were varied to determine the optimum conditions for broadening the CF_3I absorption spectrum. The largest broadening that was observed was an increase in the FWHM by ~ 2.25 for 5-torr CF_3I irradiated, using the R(14) line of the $9.6\text{ }\mu\text{m}$ CO_2 laser transition at a power level of $\sim 1 \times 10^6\text{ W/cm}^2$. This broadening was accompanied by a reduction in the number density at line center of ~ 3 . It was anticipated that these two numbers should agree if those molecules shifted from line center moved to a useful absorption region. A check of the absorption of the CO_2 laser by the 5 torr CF_3I before and after irradiation at $1 \times 10^6\text{ W/cm}^2$ indicated a 30-percent decomposition of CF_3I . This agreed with the missing molecules removed from line center, but did not show up in the broadened profile. For the same conditions, a reduction in CO_2 laser power to $3 \times 10^5\text{ W/cm}^2$ decreased the decomposition to 11 percent but also decreased the broadening factor to ~ 1.4 . Continued reduction in CO_2 laser power resulted in further reduced broadening.

The above experiments involved absorption by the ν_1 vibrational mode of CF_3I . This band was selected by Pummer et al.³² because of the high density of transitions, particularly in the region of the Q-branch, which would facilitate multiquantum absorption processes leading to highly excited vibrational states. Because of the problems with decomposition using ν_1 , we also irradiated CF_3I in the $\nu_2 + \nu_3$ vibrational mode. Because the CO_2 laser is less strongly absorbed, a higher CF_3I pressure is required to deposit the same CO_2 laser energy, thus decreasing the number of photons absorbed per molecule on the average. At 40-torr CF_3I and a power level of $1 \times 10^6\text{ W/cm}^2$ on the P(34) line of the $9.6\text{-}\mu\text{m}$ CO_2 laser transition, a broadening of 25 percent was observed. The broadening was found to increase as the ratio of absorbed photons/molecules increased. However, decomposition was also observed, although not quite as severe as with ν_1 .

Using ν_1 and R(14), data were recorded at several delays of up to $30\text{ }\mu\text{s}$ after CO_2 irradiation and found to be the same. Based

on our estimates, this should be sufficient time for vibrational equilibration of the CF_3I . This suggested that deposition of the same CO_2 laser energy over a longer time scale would result in the same broadening and might reduce the amount of decomposition because of the lower flux. By varying the flow rate of gases in the CO_2 laser, the pulse width of the laser was changed from $\sim 300\text{ ns}$ FWHM to $\sim 2\text{ }\mu\text{s}$ FWHM (Figure 42). With R(14) and 5-torr CF_3I , the same energy was deposited in the cell for the two pulse widths. In each case, the percentage CF_3I decomposition was the same.

Fig. 42. Temporal profiles of the output pulses from a Lumonics T103 CO_2 laser for two different ratios of gases in the flow mixture.

While we have observed broadening of the CF_3I absorption spectrum for many conditions, the broadening was accompanied by the decomposition of CF_3I by the CO_2 laser. Unfortunately, it appears that the degree of broadening correlates directly with the degree of CF_3I decomposition. The decomposition of CF_3I by CO_2 laser photons limits the usefulness of this technique for two reasons. First, the decomposition products are CF_3 radicals and ground state iodine atoms. Thus, for 30-percent decomposition by the CO_2 laser and 25-percent photolysis by the flashlamp producing excited iodine atoms, a condition of zero inversion exists. Secondly, the

decomposition of CF_3I increases the amount of material for chemical regeneration. If one could somehow avoid the decomposition problem, this technique would be an attractive way to improve the efficiency of the atomic iodine photodissociation laser.

Chemical Regeneration

As is well known, the usual starting material employed in an iodine laser is a perfluoroalkyl iodide, RI. The operation of an iodine laser results in the conversion of some of the RI into R_2 and I_2 . Because RI is expensive in a power plant it would be necessary to regenerate RI from R_2 and I_2 rather than to replace it by purchase of new material. References 1 and 34 present calculations estimating the amount of R_2 formed as a function of relevant laser operating parameters and discuss the economic motivation for chemical regeneration. These reports describe in detail the theoretical basis for a regeneration scheme based on dissociation of R_2 via multiple photon absorption in the infrared. Reference 1 also suggests that chemical reactions in low-frequency discharges might lead to efficient regeneration. The following material presents current results of ongoing regeneration experiments at Sandia, and gives projected values of chemical regeneration figures of merit ("efficiencies") where appropriate.

Ideally, a simple, purely chemical procedure might be found to carry out the process

(Throughout this report chemical reactions are labeled with Roman numerals.) Unfortunately, the R_2 molecule, a perfluoroalkane, is very stable both thermodynamically and kinetically. The specific carbon-carbon bond holding the two R groups together must be attacked in order to make (I) proceed and there is no known chemical route for this attack. Before shelving the search for a purely chemical technique for regeneration, Sandia personnel discussed the problem with

chemists of several disciplines in both industry and universities. The unanimous response was that an efficient purely chemical scheme did not exist. It should be mentioned that destructive pyrolysis of R_2 has been discussed as the basis of a regeneration scheme³⁵ but that it does not lead to an efficient process because the pyrolysis step is not selective enough in the manner in which it attacks R_2 .

In the absence of any purely chemical procedure for carrying out (I), schemes involving physical steps have been pursued. The most promising physical step appears to be

because the subsequent reaction of R with I_2 or I to form RI is spontaneous. Two physical schemes for carrying out (II) are presently being investigated: the excitation of R_2 via multiple photon absorption in the CO_2 laser frequency region and dissociation of R_2 in continuous wave alternating current electrical discharges. Studies using each of these techniques are described below.

Dissociation Via Multiple Photon

Absorption -- The absorption of a single infrared photon leaves a molecule vibrationally excited but stable. The absorption of a large number of such photons leaves a molecule sufficiently excited that it can dissociate if a large fraction of the energy finds its way into one bond. Reference 34 presents calculations of dissociation rate as a function of absorbed energy based on a statistical theory for the rate at which energy migrates through the bonds of a molecule and builds up in a particular one. The calculations in Reference 34 indicate that it should be possible to use a CO_2 laser to excite R_2 in such a way that (II) proceeds at an appreciable rate and that all other dissociation processes proceed at negligible rates. Based on the work in Reference 34, the experiments described below were initiated.

Effort has been concentrated on the simplest R1 of interest, CF_3I , and the corresponding R2, C_2F_6 . The first step of the experimental work was determining the absorption strength of molecules of interest in the CO_2 laser frequency region. Figure 43 is a simplified

lines P(30)-P(46). Detailed work focused on these two regions. Absorption strengths were determined for C_2F_6 , CF_3I , $\text{C}_2\text{F}_5\text{I}$, CF_3H , and $\text{C}_2\text{F}_5\text{H}$. Spectroscopic data were obtained at fluences of about 0.3 J/cm^2 (intensities of about $2 \times 10^5 \text{ W/cm}^2$). These values are low enough that saturation effects

Fig. 43. Simplified schematic of apparatus for spectroscopy and laser-induced chemistry. M_1 , M_2 , M_3 are flat molybdenum mirrors. M_2 is removed during spectroscopy experiments. S_1 and S_2 are sodium chloride plates. Cell windows are of sodium chloride.

schematic of the apparatus used for spectroscopic studies and for high intensity irradiations. The same laser is used in both kinds of studies so that there will be no change in the temporal behavior or frequency distribution from one experiment to the other.

It was determined that C_2F_6 absorbs in two different parts of the CO_2 $9\text{-}\mu\text{m}$ ($00^0_1 - 02^0_0$) laser band. The ν_5 fundamental absorbs R(30) and lines of shorter wavelength. The $\nu_2 + \nu_{12}$ and $\nu_7 - \nu_{12}$ combinations absorb the

should not be important at the typical operating pressures of several torr.

Table II reports absorption strengths in terms of the phenomenological, or effective, absorption cross section defined as

$$\sigma_{\text{eff}} (\text{cm}^2) = \left(\frac{1}{nL} \right) \ln \left(\frac{I_0}{I} \right) \quad (6)$$

where n is the total number density of the absorbing molecule, L is the absorption

TABLE II
Effective Absorption Cross Sections for Molecules of Interest in
the CO_2 00⁰1-02⁰0 Laser Band

R Branch					
Molecule	P (torr)	σ_{eff} (R34) (cm^2)	σ_{eff} (R36) (cm^2)	σ_{eff} (R40) (cm^2)	σ_{eff} (R42) (cm^2)
C_2F_6	15	6.3×10^{-21}	8.5×10^{-21}	1.1×10^{-20}	1.3×10^{-20}
CF_3I	1	3.5×10^{-19}	3.4×10^{-19}	3.1×10^{-19}	2.8×10^{-19}
$\text{C}_2\text{F}_5\text{I}$	10	3.6×10^{-20}	4.1×10^{-20}	5.0×10^{-20}	5.8×10^{-20}
CF_3H	30	9.6×10^{-22}	1.4×10^{-21}	3.1×10^{-21}	1.2×10^{-21}
$\text{C}_2\text{F}_5\text{H}$	2	2.9×10^{-20}	3.1×10^{-20}	3.3×10^{-20}	3.6×10^{-20}

P Branch				
Molecule	P (torr)	σ_{eff} (P30) (cm^2)	σ_{eff} (P34) (cm^2)	σ_{eff} (P38) (cm^2)
C_2F_6	15	3.2×10^{-21}	5.6×10^{-21}	5.6×10^{-21}
CF_3I	5	7.0×10^{-20}	1.1×10^{-19}	1.3×10^{-19}
$\text{C}_2\text{F}_5\text{I}$	20	3.4×10^{-21}	3.3×10^{-21}	3.9×10^{-21}
CF_3H	30	8.1×10^{-22}	1.1×10^{-21}	3.2×10^{-22}
$\text{C}_2\text{F}_5\text{H}$	50	7.2×10^{-22}	7.6×10^{-22}	8.1×10^{-22}

path length, and T_e and T are the fractional transmissions of the evacuated and filled cell, respectively. Data were taken at different pressures for different molecules in order to optimize signal/noise.

Even though the R branch lines are about 25 cm^{-1} to the red of the ν_5 band center of C_2F_6 , Table II indicates that the σ_{eff} values are larger in the R branch than in the P branch. For this reason, high intensity irradiations

were undertaken in the R branch. The line chosen was R(36) because the laser fluence on lines with larger values of σ_{eff} was appreciably lower than that for R(36). Table II shows that CF_3I absorbs much more strongly than C_2F_6 . Experiments in which CF_3I is regenerated from C_2F_6 and I_2 must therefore be carried out in such a way that the CF_3I product is never exposed to the laser radiation, lest it be decomposed. This will not be difficult in a well-designed commercial process, but it greatly complicates initial experiments. CF_3H absorbs

appreciably less strongly than C_2F_6 and can be exposed to fairly high-intensity laser radiation with little decomposition. This simplifies experimental design and is the reason that current work focuses on carrying out the process

via the intermediate step (III) followed by reaction of CF_3 with H_2 or H . Most of the information obtained in this study will be directly transferable to the C_2F_6/H_2 system.

In order to determine the conditions appropriate for carrying out (III), various mixtures of C_2F_6 and H_2 have been subjected to pulsed CO_2 laser irradiation by using the apparatus shown in Fig. 43. The R(36) line at $9.192 \mu m$ is used and the typical fluence is $6 J/cm^2$. The irradiation pulse width is approximately 10^{-7} seconds. Infrared spectra of samples are obtained before and after irradiation by using a commercial recording infrared spectrophotometer. In this way the nature and concentration of reaction products can be determined. It has been found that (III) proceeds, and that there is only one undesired by-product, C_2F_4 . It is not certain what the mechanism of C_2F_4 production is, but the most plausible one involves

followed by elimination of HF from $C_2F_5H^+$. The symbol $+$ denotes vibrational excitation. Unfortunately, there is no chemical kinetic information relevant to (IV) in the literature.

The overall reaction efficiency and the ratio of CF_3H production to C_2F_4 production have been determined for a number of C_2F_6/H_2 mixtures. The optimum pressure of C_2F_6 has been found to be 3 torr. Figure 44 shows the dependence of the fractional consumption of C_2F_6 in the irradiated volume per laser pulse on hydrogen pressure for a C_2F_6 pressure of 3 torr. In order to make inferences about

possible mechanisms for multiple photon absorption, data were obtained with C_6H_{14} in place of H_2 and these are also shown in Fig. 44. The pressure dependences of the H_2 and C_6H_{14} data are radically different. The most likely explanation is that adding H_2 enhances C_2F_6 IR absorption by increasing the rate of rotational relaxation and that adding C_6H_{14} decreases the rate of decomposition by increasing the rate of vibrational relaxation. Figure 45 presents the dependence of the CF_3H/C_2F_4 production ratio on H_2 and C_6H_{14} pressures for the same experimental conditions appropriate to Fig. 44. For both H_2 and C_6H_{14} , there is a maximum in the curve of production ratio versus pressure, and the curves have qualitatively the same shape. This supports the inference drawn above that H_2 and C_6H_{14} differ in the ways they affect the absorption and dissociation processes rather than in the ways they participate in chemical reactions.

Fig. 44. Fractional consumption of C_2F_6 in irradiated volume per laser pulse for 20 pulses of CO_2 R(36) radiation at fluence of $6 J/cm^2$ per pulse. Line segments connecting data points have no theoretical significance $P_{C_2F_6} = 3$ torr.

Fig. 45. $\text{CF}_3\text{H}/\text{C}_2\text{F}_4$ production ratio for 20 pulses of CO_2 R(36) radiation at fluence of $6.3/\text{cm}^2$ per pulse. Line segments connecting data points have no theoretical significance.

In order to evaluate the efficiency of the chemical regeneration process, a measurement was made of the energy absorbed by a mixture of 3-torr C_2F_6 and 3-torr H_2 . Preliminary results indicate that the energy absorbed per molecule of CF_3H formed was 3.9 eV, corresponding to 7.8 eV absorbed per carbon-carbon bond broken. The carbon-carbon bond energy, 4.2 eV, represents an absolute lower limit to the required absorbed energy, but any realistic process requires a larger absorbed energy as discussed in Reference 34.

The figure of merit (or "efficiency") of a chemical regeneration process is defined as

$$\eta_R = \frac{\text{extracted laser energy}}{\text{energy input to regeneration process}} \quad (7)$$

and theoretical estimates of η_R were made in Reference 34. The theoretical estimates of η_R for CF_3I , $i\text{-C}_3\text{F}_7\text{I}$, and $t\text{-C}_4\text{F}_9\text{I}$ were 0.035, 0.12, and 0.29, respectively, and included estimates of parameters such as CO_2 laser efficiency. The absorbed energy per CF_3H molecule formed in a $\text{C}_2\text{F}_6/\text{H}_2$ mixture can be used to determine an experimental regeneration efficiency. Using the same assumptions regarding certain process parameters as were used in Reference 34, the experimental value of η_R for CF_3H is 0.025. This value should be compared to the theoretical value for CF_3I because the absorbed energy required to break the carbon-carbon bond in $\text{C}_2\text{F}_6/\text{I}_2$ and in $\text{C}_2\text{F}_6/\text{H}_2$ mixtures should be the same. Within the precision of the preliminary measurements, theory and experiment agree, especially when the existence of the C_2F_4 product is considered. This is encouraging, as it suggests that the theoretical prediction of larger η_R for $i\text{-C}_3\text{F}_7\text{I}$ than for CF_3I may be reasonable.

The present status of this work is that values of η_R compatible with an overall system efficiency of 1 percent have been attained, assuming that the present work can be transferred directly to the $\text{C}_2\text{F}_6/\text{I}_2$ system. It is not known whether C_2F_4 will be a product in the $\text{C}_2\text{F}_6/\text{I}_2$ system. If it is, C_2F_4 can be fluorinated to C_2F_6 and recycled with some reduction in overall regeneration figure of merit. It must be emphasized that the present experimental conditions are probably not optimum and that appreciably larger values of η_R may be attained in the near future.

The development of a successful regeneration process is still in the early stages. In the near future, the dependence of the rate of consumption of C_2F_6 and of the $\text{CF}_3\text{H}/\text{C}_2\text{F}_4$ production ratio on fluence and on frequency of the exciting CO_2 laser line will be determined. Next, R_2 molecules other than C_2F_6 will be studied. Once the best combination of exciting frequency, fluence, and gas mixture has been found in R_2/H_2 systems, the most promising R_2/I_2 system will be studied with techniques that avoid the decomposition of RI .

Dissociation and Reaction in Electric Discharges

Another promising technique to regenerate RI compounds from RR compounds involves the use of electric discharges. An example of fluorine compound synthesis using an electric discharge technique can be found in Ref. 36 which was concerned with the formation of a number of trifluoromethyl organo-metallic compounds. With certain variations, the technique has also been used for synthesis of CF_3Cl from C_2F_6 and Cl_2 ,³⁷ and used in the synthesis of fluorinated polymeric compounds.³⁸ While the technique appears promising for the regeneration of RI compounds from RR compounds, little work exists in the literature which would suggest the most efficient way to use this technique for regeneration of RI compounds from RR compounds. A further difficulty is that there is not enough input from the literature to estimate in a realistic way what the energy cost per RI molecule would be for a large-scale regeneration facility.

The goals of this initial study are (1) to determine if it is possible to regenerate RI compounds of specific interest from RR compounds by using electrical discharge techniques, (2) to perform parametric studies concerning the optimization of formation efficiencies of the desired compounds, and (3) to use such information in the projection of a large facility for regeneration as required in a full-scale iodine laser system.

A block diagram of the initial setup used to investigate the interaction of electric discharges with RR compounds is shown in Fig. 46. The system is designed so that gas can flow through the discharge tube at a given mass flow rate at an independently controlled pressure. Two quadrupole mass spectrometers are used in this study. One spectrometer is located immediately downstream from the discharge so that species present in the discharge can be identified. The second spectrometer is located about 70 cm downstream from the discharge so that end-product analysis can be achieved. Although the primary gas flow is established at the flow rate controller, a second gas

injection orifice has been incorporated just downstream from the discharge unit so that gas reactants can be injected into the flow just after radical formation occurs in the flow system. This secondary gas injection system is used in connection with gas titration experiments as an adjunct to the mass spectrometers in determining the kinds and amounts of radical species formed in the discharge. It is also used to allow radicals to react with a second gas downstream from the discharge rather than directly in the discharge and thus prevent product breakup in the discharge.

Initial studies were carried out by using a microwave discharge unit operating at 2.4 GHz with total power dissipation of up to 100 W. Results obtained have shown that when C_2F_6 is passed through such a discharge a permanent modification of this gas can take place. The mass spectrometer cracking pattern contains a peak at atomic mass position 119 and is identified as C_2F_5^+ . The amplitude of this peak is proportional to the amount of C_2F_6 left in the flowing gas sample as it passes through the discharge. Studies of this mass peak as observed at the two mass spectrometer positions (Fig. 46) show that though most of the C_2F_6 can be destroyed in the discharge (depending upon discharge power, gas flow rate, and pressure) many of the CF_3 radicals formed in the discharge can associate to reform C_2F_6 downstream from the discharge. However, the compound C_2F_4 is also formed in the discharge. Initial results regarding C_2F_4 production indicate that the relative amount of C_2F_4 produced is minimized for low-discharge power, high-flow rate and low C_2F_6 pressure.

Some difficulty has been experienced in determining absolute yields of CF_3 radicals in C_2F_6 . Since the main cracking pattern peak observed for C_2F_6 occurs at atomic mass position 69 (i.e., the CF_3^+ ion), detection of the parent CF_3 radical peak in the presence of C_2F_6 is difficult. An approach to identify the presence of CF_3 in C_2F_6 has involved the reaction of Cl_2 with the radicals produced in C_2F_6 . The

CF_3 radicals can react with Cl_2 as follows:

The cracking pattern of CF_3Cl contains peaks at atomic mass positions 85 and 87 corresponding to the CF_2Cl^+ ion as well as having a strong peak at atomic mass position 69 corresponding to the CF_2^+ ion. When Cl_2 is injected into the flow through the orifice immediately below the electric discharge (Fig. 46), peaks at atomic mass positions 85 and 87 do occur during the time C_2F_6 is passed through the electric discharge (but not when the discharge is turned off). The buildup of CF_3Cl as a function of C_2F_6 pressure (with excess Cl_2) is shown in Fig. 47 for the case where a 5 W microwave discharge is used with a C_2F_6 flow rate of 100 cm/s. It is seen from this figure that the absolute yield of CF_3Cl increases with C_2F_6 pressure until a C_2F_6 pressure of 0.04 torr is reached. Higher C_2F_6 pressures do not result in higher absolute CF_3Cl yields.

Although absolute mass spectrometer calibrations concerning the amount of CF_3Cl actually produced in the system are not yet complete, an upper-limit estimation of the CF_3Cl yields can be obtained by observing the absolute amount of Cl_2 used in forming CF_3Cl . In Fig. 48, two curves are shown representing the mass 70 Cl_2 signal as a function of Cl_2 injection pressure in the flowing system for the case where C_2F_6 is flowing through the discharge at 100 cm/s. The top curve was obtained for the case where the discharge is off. With no discharge, the Cl_2 mass peak increases linearly with increasing Cl_2 pressure in the flow. When the discharge is turned on (5 W), the Cl_2 peak does not initially increase linearly with Cl_2 injection pressure because at the low Cl_2 pressures most of this gas is used in forming CF_3Cl . However, as shown from the lower curve, as the Cl_2 injection pressure is increased so that excess Cl_2 occurs in the flow system, the Cl_2 signal becomes linear with increasing Cl_2 injection pressure and has the same slope as the upper curve obtained in the absence of the discharge. The shift in Cl_2

Fig. 46. Block diagram of the flow-gas discharge system incorporating mass spectrometer diagnostics.

Fig. 47. The yield of CF_3Cl produced by injection of Cl_2 just downstream from the discharge in a flowing C_2F_6 gas stream at various pressures.

Fig. 48. The effect of a 5-watt discharge on the downstream Cl_2 concentration due to interaction of Cl_2 with radicals produced in a C_2F_6 gas stream passing through the discharge.

pressure due to the presence of the 5-W microwave discharge reflects the absolute amount of Cl_2 which combined with the reactive species formed in C_2F_6 as it passes through the discharge. From the data obtained in Fig. 48, it can be determined that the partial pressure of Cl_2 used in combining with radicals produced in the C_2F_6 as it passed through the discharge was $\sim 0.6 \times 10^{-2}$ torr. Since the only significant product involving chlorine detected downstream from the discharge was CF_3Cl , a conclusion is reached that essentially all of the Cl_2 which reacted in the flow system resulted in the formation of CF_3Cl and that ~ 0.012 torr of CF_3Cl molecules were formed in the flowing gas stream.

The relative efficiency as a function of microwave power for the formation of CF_3Cl is shown in Fig. 49 for the case where 0.02 torr of C_2F_6 is flowing through the discharge at 100 cm/s and where excess Cl_2 is injected at the orifice just below the discharge. It is seen from the figure that the number of CF_3Cl molecules obtained per unit of microwave energy increases sharply as the discharge power is reduced.

Fig. 49. The relative efficiency of CF_3Cl formation as a function of microwave cavity input power for a stream of C_2F_6 with downstream injection of Cl_2 .

Initial experiments have been performed where the Cl_2 is premixed with C_2F_6 before passing through the discharge. The relative CF_3Cl formation efficiency appears to be more than three times greater in this case than was observed for the case where Cl_2 is injected immediately below the discharge. The main reason for this increase in efficiency most likely arises because where Cl_2 passes through the discharge along with C_2F_6 , there is more favorable competition for CF_3Cl formation in the discharge itself as compared to CF_3 association to reform C_2F_6 .

The results of this study have shown that CF_3 radicals can be produced in flowing C_2F_6 and that it is possible to react these radicals with Cl_2 to form CF_3Cl . The relative formation efficiency of CF_3Cl with respect to the formation of the undesirable compound C_2F_4 can be increased by working at low-discharge power levels, low C_2F_6 pressures and relatively high-flow rates. The CF_3Cl formation efficiency in the present laboratory setup is greatly improved by premixing C_2F_6 with Cl_2 upstream from the discharge rather than injecting Cl_2 into the flow immediately downstream from the discharge. Since the relative CF_3Cl formation efficiencies increase sharply with decreasing discharge power, it is expected that reducing the discharge power still further (after system modification) will result in significant improvement in the formation efficiency.

Since system modification is required before lower discharge powers can be used to maintain a discharge in a flowing gas system, such modifications are to be carried out as soon as possible. An RF generator operating in the range of 10 MHz is to be used in setting up a low-energy gas discharge system. Also, concerning the use of the microwave discharge system, work will be carried out to improve the Q of the resonant cavity-flow tube system so that less energy is absorbed in the tube and more in the gas flowing through the system.

Although Cl_2 has been used in this study as a label (and reactant) for purposes of facilitating the interpretation of the mass spectrometric data, work using I_2 with formation of CF_3I will be undertaken shortly.

The Iodine Amplification and Adaptive Propagation Code, ISTAR

The previous semiannual report¹ described the proposed theory for generalizing the $r - z - t$ Maxwell-Bloch code (TZYR) for iodine amplifiers into a code containing free-space propagation subroutines as well as provisions for the amplification of divergent coherent beams. The proposed coordinate transformations¹ were implemented and found to work very well. Considerable effort was devoted to making the propagation scheme adaptive so that the numerical gridwork could be revised periodically to avoid pulse walkoff or collapse. Subroutines were added to describe lensing, aperturing, beam expansion, and several necessary analysis operations related to the adaptive coordinate transformations. The total code is now referred to as ISTAR,³⁰ and it has been used for the study of anomalous dispersion effects on focal spot size and beam quality described elsewhere in this report. Some details of ISTAR and its performance will be elaborated here.

In cylindrical coordinates, the paraxial equation for the evolution of the electric field envelope in the Maxwell-Bloch equations is

$$\left[\frac{\partial}{\partial z} - \frac{1}{c} \frac{\partial}{\partial t} - \frac{1}{2ik} \left(\frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} \right) \right] E = 0 \quad (8)$$

$\mathcal{P}$ is the medium polarization, which is zero for free-space propagation. By transforming to retarded time coordinates, the time derivative disappears from the equation. Now change coordinates from the cylindrical r and z via the substitution,

$$s = z$$

$$\theta = r/(z + R) \quad (9)$$

Subsequently make a dependent variable transformation on the electric envelope,

$$\psi = \frac{R}{z + R} \exp \left(-i \frac{k}{2} (s + R)^2 \right) \bar{\psi} \quad (10)$$

$$\frac{R}{z + R} \exp \left(-i \frac{k r^2}{2(z + R)} \right) \bar{\psi} ,$$

and an analogous one on $\bar{\psi}$ to become $\bar{\bar{\psi}}$. The new differential equation is

$$\left[\frac{\partial}{\partial s} - \frac{1}{2ik} \frac{1}{(s + R)^2} \left(\frac{\partial^2}{\partial \theta^2} + \frac{1}{\theta} \frac{\partial}{\partial \theta} \right) \right] \bar{\bar{\psi}} = 0, \quad (11)$$

which is of the same form as the original equation in $r = z$ coordinates. The advantage is that we now have a finite difference grid network in $\theta = s$ that expands or contracts in just the proper manner in $r = z$ space to follow the electric field that would arise from a point source at the singular point, $z = s = -R$. The transformed dependent variables are smooth in the new coordinates because of the factorization of the radial phase that is associated with the paraxial approximation to the point source field. The amplitude factor $R/(z + R)$ removes a proper " $1/R^2$ " law behavior from the beam. The unspecified constant R has dimensions of length and is the distance to the hypothetical point source; it thus is identified as the phase front radius of curvature R_0 of the beam at the point where $z = 0$. This tells how to specify R and to determine a unique set of coordinates to use in a given region of space: the Maxwell-Bloch equations should be integrated in the new coordinate system with the transformation parameter R chosen to be as close as possible to the local radius of curvature of the beam. As soon as the radius of curvature deviates from the value of $R_0(z) = R + z$ at z , the coordinates are changed to a new set and the appropriate phase factorizations removed from the pulse. We note that coordinate

transforms are necessary whenever the beam is being affected by diffraction, by lensing, or by self-focusing, all of which slowly or abruptly change the radius of curvature away from the simple $R + z$ behavior. In the case of a lens, one knows exactly how to modify the coordinates to follow the beam after the lens; the coordinate transformation does not depend on the pulse. In the case of self-focusing or of diffractive propagation of a nonsimple beam, the code must analyze the pulse itself to determine the best coordinates to use to continue propagation of the pulse. This adaptive problem will be described shortly.

The finite difference integration method for the six level Maxwell-Bloch theory in cylindrical coordinates was reported previously.¹ We developed a predictor-corrector integrator in t and z combined with implicit Crank-Nicolson-Thomas advance in z . All this numerical description remained intact except the implicit solution. Let D denote the tridiagonal finite difference operator for the radial Laplacian. In cylindrical coordinates it is constant; however it now varies with s (or z) as one integrates the retarded paraxial equation (in vector-matrix notation):

$$\left(\frac{\partial}{\partial s} - \frac{1}{2ik} D(s) \right) \xi(s, t) = \eta(s, t) \quad (12)$$

One advances the solution in the s coordinate by solving the tridiagonal matrix equation,

$$\left(\frac{1}{2} - \frac{1}{2} D(s) \right) \xi(s, t) = \left(\frac{1}{2} + \frac{1}{2} D(s - \Delta s) \right) \xi(s - \Delta s, t) + \Delta s \eta, \quad (13)$$

for $\xi(s, t)$. Previously D was the same on the left and right of the above equation. There was concern that the difference scheme would now be unstable with this modification since one is not doing forward-

backward averaging exactly, but some mixture of that with an explicit forward step which can be unstable if Δs is unrestricted in relation to $\Delta \theta$. It was found that the new method was stable for all tested cases, apparently because we always are close to the Δs bound for explicit stability and because the forward-backward averaging of the $\partial/\partial s$ derivative suppresses some unstable growth. Since everything worked well, analysis was not attempted.

In order to change coordinate systems to accommodate the pulse, one needs an analysis procedure to determine an effective pulse radius and radius of curvature. This might appear trivial, but the pulse consists of as many as 50,000 complex numbers at each particular point in z . These are in the form of radial vectors stored end-to-end in order of increasing time on disc file. Other vectors relating to the medium are stored along with the field. An iterative method based on converging to a least-squares fit of the pulse to a Gaussian beam form would properly determine the radii, but it would require as much computer processing as the propagation part of the calculation. We decided to determine the radii by an operator method since this is fast (noniterative) and fits in well with the determination of power as a function of time and flux as a function of radius. Consider the radial functional dependence of a Gaussian beam form:

$$E_{GB} = c \exp(Zr^2) \quad (14)$$

$$Z = -1/w^2 - ik/2R_c$$

Both the transverse radius or waist and the radius of curvature are contained in the complex beam parameter Z . The simplest operator that picks out Z is

$$\frac{\partial}{\partial(r^2)} E_{GB} = Z E_{GB} \quad (15)$$

or

$$Z = \frac{1}{2(r^2)} \ln E_{GB} \quad (16)$$

For a Gaussian beam, Z is recovered exactly; in the case of a non-Gaussian beam, we perform averages of the operator over the pulse. Two such averages or expectation values that are useful are:

$$\bar{Z}_t(t) = \frac{Q_t(r)}{P(t)} \frac{2\pi \int_0^\infty r dr E^2 \frac{\partial}{\partial r^2}}{2\pi \int_0^\infty r dr E^2} \quad (17)$$

and

$$\bar{Z}_r(r) = \frac{Q_r(r)}{I(r)} \frac{\int_{-\infty}^\infty dt E^2 \frac{\partial}{\partial r^2}}{\int_{-\infty}^\infty dt E^2} \quad (18)$$

An average over the whole pulse may be formed from either one, e.g.:

$$\bar{Z} = \frac{\int_{-\infty}^\infty dt Q_t(t)}{\int_{-\infty}^\infty dt P(t)} \quad (19)$$

All of these average Z s reduce to the proper constant for a Gaussian. Note that $P(t)$ is the power as a function of time and $F(r)$ is the beam flux as a function of radius. These various quadratures are done numerically by one reading of the pulse from disc. The Z s may be decomposed into waist and curvature, e.g.:

$$w_0 = (-\text{Re } \bar{Z})^{-1/2} \quad (20a)$$

$$R_{c0} = -k(2 \text{Im } \bar{Z})^{-1} \quad (20b)$$

and $\bar{Z}_r(r)$ is used to determine a time-averaged radius-dependent phase front radius of curvature such as is recorded by a shearing interferometer. The above w_0 and R_{c0} are used in the choice of new coordinates.

We summarize the problem of adaptive integration for free-space propagation; the Maxwell-Bloch amplifier integrations are a special case. The pulse's temporal and radial behavior are given or have been computed at a particular location, $z = 0$. The beam analysis described in the previous paragraph gives a fit to the average waist and radius of curvature there. It is desired to propagate the beam over some total distance l which could include a focus, for example. We must determine (1) the appropriate subinterval l of l to propagate before re-analyzing the pulse and resetting the coordinates, (2) the proper coordinates and step size to use for the subinterval, and (3) how to modify the coordinates to compensate for beam walkoff or collapse. We do not wish to analyze and change coordinates at every Δz increment since this is inefficient, although it would circumvent the choice of subinterval. We will not elaborate here on the methods for selecting the numerical grid steps Δr , Δz , or Δt . We require the subinterval l to be less than the Rayleigh range,

$$l \leq \frac{1}{2} k w_0^2 \quad (21)$$

where w_0 is the average waist at the start. This restriction helps insure that diffraction does not cause beam walkoff within the subinterval over which we use a fixed set of $\theta - s$ (or $\theta - z$) coordinates. We require that l be less than the distance over which we would revise the Δs (Δz) increment by a factor of two:

$$l \leq 0.3 \left| R_{c0} \right| \quad (22)$$

This is derived from Gaussian beam behavior and the criterion for choosing Δz . It prevents overshooting a focal point as well as allowing Δz to readjust periodically. Now, having fixed l , the remaining choice is the precise coordinates to use to cover the subinterval. We cannot simply transform to

$$s = z \quad (23a)$$

$$\theta = r / (z + R_{c0}) \quad (23b)$$

where R_{c0} is the radius of curvature at the last analysis position, $z = 0$. This is because the pulse is diffracting and so is spreading transversely faster than given by the radius of curvature at the start of the interval. This creates a large amount of pulse walkoff in the vicinity of a focus (beam waist). An unsuccessful solution to this problem was to increase the rate of spread of the coordinate system by such an amount that over the interval l the increased grid expansion would compensate for walkoff in the previous interval as could be seen in the waist at $z = 0$. This turned out to be an unstable procedure which broke into oscillations. The successful solution to the choice of coordinates was to use simple Gaussian beam theory to extrapolate the waist w_L at $z = l$ given the fit quantities w_0 and R_{c0} at $z = 0$. This is an algebraic relation. The $\theta - s$ coordinate system is chosen so that it expands by the factor w_L/w_0 in the distance l . In between there is a slight contraction of the beam which is limited by the choice of l to prevent any significant beam collapse.

The new $\theta - s$ coordinates were programmed into the old TZYR code for amplifiers. Free-space propagation, lensing, etc. options were added and the new code (ISTAR) was checked and evaluated in the process of assembly. It was found necessary to extend the radial grids farther out into the wings of the pulse in order to improve the imaging properties of the propagation. Table III

illustrates the performance of ISTAR for the case of amplification (with significant pulse distortion due to saturation) and subsequent focusing of the pulse. The pulse undergoes considerable "squaring-out" due to the saturated amplification; this causes w to grow relative to the growth of RMAX in the amplifier interval. However, from this point onward the grid maximum is seen to remain a near-constant multiple of w which proves that the grid is following (perhaps predicting) the pulse expansion. This is also shown by

the energy column which deviates less than one-third percent from the amplified value throughout the lensing, focusing, and expansion region. The fact that the waist returns to 7.65 mm at 4 m beyond the 2-m focal length lens demonstrates the true imaging potential of the code. R_c^{GIB} is the radius of curvature predicted by the Gaussian beam formula; it is given to prove that the beam is not a simple Gaussian, in which case the ability of RMAX to follow w would not be surprising.

TABLE III

Example of Adaptive Integration Ability of ISTAR Code

Position	L (m)	Steps to reach	w (mm)	RMAX (mm)	R_c (m)	R_c^{GIB} (m)	Energy (J)
Start of amplifier		0	2.20	13.13	3.62	3.62	0.0060
Finish of amplifier	5.300	66	6.09	21.21	13.25	13.92	0.4983
Propagation	3.700	5	7.65	27.15	17.03	16.92	0.4969
Propagation	4.000	4	9.30	33.53	21.09	21.00	0.4969
Lensing, f = 2m	0	0	9.30	33.53	-2.21	-2.21	0.4969
Propagation over many (about 2 m subintervals total)		50	0.175	0.709	-0.06	-0.07	0.4974
Propagation	0.0241	20	0.140	0.549	-0.12	-0.08	0.4974
Propagation	0.0346	46	0.131	0.560	0.13	1.80	0.4973
Propagation	0.0393	50	0.210	0.907	0.07	0.10	0.4973
Propagation	0.0197	11	0.299	1.191	0.08	0.08	0.4975
Propagation	0.0232	8	0.425	1.554	0.10	0.10	0.4976
Propagation over many (4 m from subintervals lens) to image plane of 3rd line above.		38	7.65	27.02	1.79	1.79	0.4980

* L, w, and R_c are explained in text. RMAX is outermost radial grid point and R_c^{GIB} is the predicted radius of curvature.

Laser Beam Aberrations and Focusability

A general discussion concerning the operation of a lateral-shearing interferometer has recently been given in Ref. 2. Of primary concern in that work was using this instrument for measuring laser beam divergence. In practice, one is typically dealing with an overall optical system (laser plus optics) that exhibits some aberration, however, one expects deviations from linear interferometric fringes. In the presence of these aberrations, enough of the fringe may be linear so that a simple divergence measurement is still possible. The question arises as to what one can infer about the focusability of such an aberrated beam from the interferogram.

In this report results are presented for the appearance of the interferometric fringes and beam focusability in the presence of third order spherical aberration. From Ref. 2 the equation describing the fringe pattern is

$$\left\{ x^2 + y^2 - \left[(x - s)^2 + (y - \theta l)^2 \right] \right\} = 2R \frac{\lambda}{R_0} \left\{ (x^2 + y^2)^{K/2} - \left[(x - s)^2 + (y - \theta l)^2 \right]^{K/2} \right\} \quad (24)$$

$$+ 2R\theta y - 2R \left(\frac{m}{2} + 1 \right) = 0$$

where x and y are the coordinates of the fringes, λ is the laser wavelength, s is the shear, R is the radius of curvature of the beam phase front at the observation plane, θ is one of the angles describing the beam¹ as it exits the shear plate, D is the optical path length difference due to the finite thickness of the shear plate, m is an odd integer corresponding to the fringe being described, and λ/R_0 are best described by writing the aberration term used:

Aberration term in wavelengths =

$$\epsilon \left(\frac{x^2 + y^2}{R_0^2} \right)^{K/2} \quad (25)$$

where for third order spherical aberration $K = 4$. The scale length R_0 is chosen to define that radius for which the maximum optical path length difference from a spherical phase front is a fraction, ϵ , of one wavelength.

Figure 50 illustrates a representative example of a calculated shear plate interferogram (l is the distance from the shear plate to the observation plane). The dashed fringes refer to an unaberrated beam, and the numbers refer to the relative fringe orders. There are several points worthy of mention. First of all, it is seen that the familiar S shape is not symmetrical relative to the straight fringes except for that order going through the center of the interferogram. Secondly, it must be emphasized that this device measures the phase difference between points of the phase front separated by the shear distance in the direction of the shear. Consequently, the interpretation of a phase distortion due to aberrations depends upon where it is measured on the interferogram. If the center of the laser beam is relatively unaberrated, then clearly one would prefer to measure phases relative to this region. Hence, a large shear may be desirable and the fringe differences due to the presence of aberration is best measured on the circle diameter along the direction of shear. The amount of shear is chosen so that the point on the phase front for which information is required lies near the center of the un-sheared beam. Experimentally, if one wanted to infer the amount of aberration from an interferogram, it seems that the most direct approach would be to calculate the interferogram patterns for different amounts of aberration until a fit is obtained.

Fig. 50. Calculated shear plate interferogram demonstrating the effect of spherical aberration.

Figure 51 shows the smallest focal spot size as computed from FOCUS⁴⁰ (solid curves) for 90 and 60 percent of the encircled incident power. The wriggles on the 90 percent power curve arise due to diffraction effects. The result of geometric optics is shown as the dashed line, extrapolated to fit the diffraction limit at zero OPD (optical path difference). N is the refractive index of the focusing lens.

KINETIC PROCESSES IN PHOTOLYTICALLY PRODUCED GROUP VI ¹S ATOMS

Overview

The class of gas-laser systems based on the excitation of the $p^4 1S$ state of Group VIA atoms (e.g., O, S, Se, Te) with energy extraction on the $p^4 1S \rightarrow p^4 1D$ or $p^4 1S \rightarrow p^4 3P$ transition is an extremely promising one for laser-fusion applications. Long energy-storage times are possible since the $1S$ state exhibits a long radiative lifetime and is resistant to deactivation by a wide variety of

Fig. 51. Calculated focal spot sizes (at the plane of minimum spot radius) as a function of the OPD (optical path difference) at the edge of the laser beam.

other species. Also, these transitions occur at useful wavelengths (450 to 700 nm) and exhibit stimulated emission cross sections that are low enough to inhibit parasitic amplified spontaneous emission but not so low that the medium cannot be saturated at reasonable energy fluxes.

The most efficient method yet discovered for producing ^1S atoms is photodissociation of a Group VI-atom-bearing compound. As noted in the last report,¹ our recently discovered electron-beam-driven F_2 laser at 157 nm produces radiation within an OCS absorption band that is expected to give a near-unit quantum yield of $\text{S}(^1\text{S})$. In the remainder of this section, we describe our attempt to further optimize the performance of the F_2 laser, our use of the F_2 laser to photolytically produce $\text{S}(^1\text{S})$ atoms, and some theoretical investigations of the role photoionization might play in the kinetics of Group VI ^1S atomic systems.

Scaling Experiments with the 3-meter F_2 Laser

Our initial attempts in scaling the F_2 laser to higher excitation fluxes (energy deposited/ cm^2/sec) in which we went from excitation fluxes of $1.5 \times 10^7 \text{ W/cm}^2$ (on the L.I.I.I. device) to $2 \times 10^8 \text{ W/cm}^2$ (on the Febetron 705) resulted in almost a factor of five increase in laser efficiency. As a relatively quick second-step experiment, we increased the active length of the F_2 laser cell from 1.4 m to 3.0 m. It was anticipated that this would allow us to test the efficiency of the F_2 laser at excitation fluxes on the order of $4 \times 10^8 \text{ W/cm}^2$. A schematic of the 3-m-long F_2 laser system is shown in Fig. 52. Like both of our previous F_2 laser systems, this laser utilized a "u" geometry in which the electron beam, after drifting through a short low-pressure region, was magnetically guided around a corner and down the gas-filled laser cell.

The magnetic field used to guide the electron beam down the 3-m-long F_2 laser cell had the same intensity (4.5 k gauss) as the guiding field used on the 1.4-m cell. However, radial spreading of the electron beam, which was barely noticeable in the 1.4-m laser, now

became a very important effect as is shown in Fig. 53. The set of data points denoted by $\bullet$ in this figure shows total energy reaching the electron-beam calorimeter as a function of the pressure in the cell, p , and location of the calorimeter, x . The reasonable agreement between these data and the deposition rate predicted by Sandia's TIGER code indicate that loss of electron-beam energy to the walls of the cell is still small. The set of data points denoted by $\circ$, which show energy reaching the central 7-cm^2 area of the calorimeter, show a much more rapid decline in energy due to radial spreading of the electron beam. This radial spreading of the electron beam reduced the excitation flux density on the laser axis to $3.2 \times 10^{11} \text{ W/cm}^2$ or an absorbed energy density of 15 J/cm^2 .

APPARATUS FOR F_2^* LASER STUDIES

Fig. 52. Schematic diagram of the 3-meter F_2^* laser apparatus.

Fig. 53. Electron-beam energy in relative units reaching the segmented calorimeter as a function of helium pressure p times distance down the cell x for the 3-metre F_2 laser apparatus. The points labeled + represent the total energy reaching the calorimeter whereas the points labeled o represent electron-beam energy reaching the central 7 cm² of the calorimeter.

Experiments with the 3-m laser at He pressures up to 9000 torr demonstrated that the laser output from this cell was dominated by a single-pass amplified spontaneous emission. Completely misaligning the optical cavity only decreased the laser output by ~ 40 percent. This was probably due partly to the fact that the round trip transit time of the optical cavity was only about a factor of two shorter than the pulse width of the gain. Thus, the laser pulse controlled by the cavity only had a few passes in which to build-up, while the gain was greater than unity. Under these conditions, the highest value of laser efficiency observed was 0.7 percent. Comparing the peak F_2 sidelight emissions with and without an aligned optical cavity on the test cell,

sidelight saturation effects as large as 32 percent were observed. It should be noted that this is only a lower limit to the actual saturation of the medium by laser photons as there clearly was some saturation of the medium even with the cavity misaligned.

One improvement was made in the F_2 handling system during this set of experiments which greatly enhanced the overall reliability of the F_2 laser. Previously, a single gas-handling system, which was mechanically pumped to $\sim 10^{-2}$ torr was used both to inject F_2 into the laser cell and to pump out spent He/ F_2 laser mixes. A soda lime trap was located just before the mechanical pump to passivate the F_2 in the mixtures to prevent it from damaging the mechanical pump. Several times during the early F_2 laser experiments, the entire F_2 laser cell became seriously contaminated either with mechanical pump oil or with soda lime. A separate F_2 handling system was built to inject F_2 into the laser cell (this system could be pumped to $\sim 1 \times 10^{-5}$ torr) and the original mechanically pumped system was used only to exhaust spent laser mixes. This improvement appears to have completely eliminated the contamination problems.

It is clear that simply increasing the excited length of the F_2 laser was not successful in improving the laser's efficiency. This seems to be due, at least in part, to the fact that the 3-m system operated in a regime in which the optical cavity had little effect on the laser's performance. Future attempts to scale-up the F_2 laser should include a test cell which has a short round-trip time, compared to the length of the excitation pulse and possibly a relatively small "prepulse" to allow the buildup of a steady-state flux of laser photons in the cavity which would control amplified spontaneous emission during the main excitation pulse.

Laser Pumping of $S(^1S)$

Introduction -- In the following sections, we describe our use of the F_2 laser to

photodissociate OCS to $(\text{CO} + \text{S}^1\text{S})$. The goals of these experiments were to determine at relatively high atom densities, the S^1S production efficiency and effective lifetime and to assess the importance of photoelectron collision processes. Further work will be required to develop a detailed kinetic model with which to interpret our experimental observations.

Experimental Configuration -- A schematic of the experimental setup is shown in Fig. 54. The molecular fluorine laser is essentially identical to the 1.4 m F_2 laser described in our previous annual report¹ and in Ref. 31. The vacuum ultraviolet output beam from this laser was coupled through an evacuated pipe to the S^1S kinetics cell. Apertures in the path of the laser beam reduced its diameter to 1.4 cm before it entered the S^1S kinetics cell. The intensity of the laser beam was monitored during the experiments by inserting a 45-degree MgF_2 window into the coupling pipe which deflected ~10 percent of the laser beam to a VUV photodiode mounted behind apertures and a diffuser. VUV laser light entered the S^1S kinetics cell through a MgF_2 window located 7 mm from the optic axis of the S^1S kinetics cell. The absolute intensity of the laser beam transmitted through the kinetics cell was monitored by a VUV-coupled laser calorimeter. Emissions from the $\text{S}^1\text{S} - \text{S}^1\text{D}$ transition at 7727 Å were monitored by either of two photomultiplier tubes mounted vertically above the cell. The PM tubes were carefully apertured to insure that they viewed a well-defined area passing through the center of the F_2 laser beam and which was centered 7 mm from the entrance window of the S^1S kinetics cell. Extensive amounts of lead and mu-metal shielding reduced the x-ray and RF-induced PM-tube noise level to less than ten millivolts within 100 ns after the laser pulse entered the S^1S kinetics cell. Two additional MgF_2 windows looked transversely through the kinetics cell with an optic axis which also was located 7 mm from the edge of the F_2 laser input window. The mounts for these windows extended well into the kinetics cell. As a result, the distance between these two windows was ~12.5 mm and the entire volume of gas between these windows was excited by

the F_2 laser. During most of the experiments, the transverse windows were used to monitor transmission of 35-GHz microwaves through the laser-excited OCS mixtures. The physical placement of the windows guaranteed that detection of waves that are diffracted around the excited medium would be small. These transverse windows were also used to monitor the intensity, time history and spectral distribution of S_2 emissions observed near 4000 Å. The F_2 laser system, the S^1S cell and its gas-handling system, and the vacuum coupling regions for the VUV laser were pumped by liquid-nitrogen-trapped oil-diffusion-pump systems and were evacuated to pressures of less than 1×10^{-5} torr.

Results and Discussion --

Laser Characteristics. As discussed thoroughly in the last report¹, the F_2 laser operates efficiently in mixtures of high-pressure helium (typically 7500 torr) and a few torr of molecular fluorine excited by a high-energy electron beam. The spectral distribution of the laser emission is shown in Fig. 55. The splitting in the doublet at 157.5 nm probably corresponds to rotational spacing in the upper laser state, whereas the third feature at 156.7 nm represents transitions to a different vibrational level. All three of these lines fall within the OCS absorption band that yields S^1S .

Fig. 54. Schematic diagram of the F_2 laser and S^1S kinetics cell.

Fig. 55. Spectrum of the F_2^* laser used to photolyze OCS. The peaks occur at 157.60 ± 0.01 , 157.49 , and 156.71 nm. The instrument resolution is about 0.03 nm. The half-height positions are marked by the number 0.5 .

The usable laser energy is limited by laser-induced damage to the output window on the F_2^* laser cell. This window separates the high-pressure helium/fluorine mixture from an evacuated coupling tube. At flux levels near 100 mJ/cm^2 in a 70-ns pulse (1.4 MW/cm^2), the MgF_2 output window typically exhibits damage on its outer surface, the side toward the vacuum, after a few dozen shots. This damage causes the window's surface to appear frosted, which enhances diffuse scattering of the laser beam with considerable loss in laser intensity at the OCS cell. We have not observed any laser damage to either of the two optical elements which comprise the laser cavity, a highly reflecting (>98 percent) dielectric-coated mirror and a polished MgF_2 flat, even after several hundred shots.

Depending on the desired experimental conditions, the laser beam actually entering the OCS photolysis cell was varied in energy from less than 1 mJ/cm^2 to near 80 mJ/cm^2 in either a rectangular beam 1.1 by 1.5 cm or a cylindrical beam 1.6 cm in diameter.

Bleaching-Wave Considerations. At the high laser fluxes employed in this study, we expect the photodissociation transition

to be partially saturated by the incident radiation field. For this case, the familiar Beer's law description of photon absorption is not appropriate. Instead, the radiation transport equations that describe propagation of the laser beam in one space dimension take the form

$$\frac{\partial I(x, t)}{\partial x} + \frac{1}{c} \frac{\partial I(x, t)}{\partial t} = -\sigma I(x, t) \rho(x, t) \quad (26)$$

$$\frac{\partial \rho(x, t)}{\partial t} = -\sigma I(x, t) \rho(x, t) \quad (27)$$

where I is the laser intensity, ρ is the density of OCS, σ is the dissociation cross section, c is the velocity of light in the medium, and the laser is assumed to be monochromatic. The solution of these equations is

$$I(x, t') = \frac{I_0(t') e^{\sigma \int_{-\infty}^{t'} du I_0(u)}}{\Gamma} \quad (28)$$

$$\rho(x, t) = \frac{\rho_0(x) e^{\int_{-x}^x du \rho_0(u)}}{\Gamma} \quad (29)$$

where

$$\Gamma = e^{\int_{-x}^{t'} du \rho_0(u)} - 1 + e^{\int_{-x}^x du \rho_0(u)},$$

$$t' = t - x/c,$$

and $I_0(t)$ is the time-dependent laser flux entering the initial OCS density profile $\rho_0(x)$. For simplicity, we assume a laser pulse of width, τ , and energy/cm² E_0 entering a uniform density of OCS. The resultant solutions for two special cases, which will be useful later on, are presented here. The energy that passes a distance, x , through the OCS-filled cell is

$$E_x = E_0 \ln \left\{ 1 + e^{-\sigma \rho_0 x} \left(e^{E_0/E_s} - 1 \right) \right\} \quad (30)$$

where $E_s = h\nu/\sigma$. The fraction of the initial OCS that is actually photolyzed at each position is

$$\frac{\Delta \rho}{\rho_0} = \frac{u}{1+u} \quad (31)$$

where

$$u = e^{-\sigma \rho_0 x} \left(e^{E_0/E_s} - 1 \right).$$

The evaluation of this fraction will be necessary for determining the absolute efficiencies presented later in the section entitled Absolute Calibrations and Quantum Efficiency of S(¹S) Production.

In order to apply these formulas, we need to know the absorption cross sections for the precise wavelength(s) of our laser. We determined this quantity at several different E_0 values by filling the OCS cell with various pressures of OCS and measuring both the incident laser energy flux E_0 and the transmitted energy flux E_x . A fit of these data to Eq. (30) yields the absorption cross section. For E_0 values of ≥ 5 mJ/cm², all the data can be well represented by Eq. (30) with $\sigma = 7 \times 10^{-17}$ cm². However, as the input flux is increased, a curious phenomenon occurs; the effective measured absorption cross section of pure OCS decreases. By the time $E_0 \approx 70$ mJ/cm², σ has dropped to $\sim 3 \times 10^{-17}$ cm². If high pressure N₂ is added to the OCS, prior to irradiation at high laser fluxes, the inferred σ value increases markedly even though N₂ is essentially transparent at 157 nm. These results suggest that ground state OCS is being depleted by some process in addition to absorption of a laser photon, and that the addition of N₂ inhibits this depletion mechanism. We believe from the microwave studies described later in the section entitled Microwave Transmission Experiments that the addition of N₂ to OCS can control an electron avalanche process that might otherwise occur. Thus, a likely explanation for the anomalous behavior of the effective absorption cross section is that for high incident laser fluxes, without added N₂, electrons are collisionally heating (or dissociating) the OCS, resulting in a lowered absorption of the laser radiation.

Time Dependence of S(¹S) Emission

The time dependence of the S(¹S → ¹D) emissions from laser dissociated OCS-buffer gas mixtures is complex. With no buffer gas, no emission is observed. With added rare gases or molecular gases, the emission is readily detected and is characterized by a rise time that is equal to or faster than that of the dissociating laser pulse and a decay time that is typically faster at early times than at late times. However, for sufficiently low fluxes, the

decay can be well characterized by a single time constant.

Figure 56 illustrates examples of the early-time behavior of $S(^1S)$ emissions for 1.0-torr OCS and added buffer gas for both large and small incident laser fluxes. From the analysis presented earlier, the rise time of the bleaching wave, and hence the $S(^1S)$ emission is approximately $\tau_r \approx (\sigma I)^{-1}$ (see Eq. (26)). For an incident laser flux of 50 mJ/cm^2 in 70 ns, τ_r is predicted to be 25 ns, in good agreement with the data in Fig. 56. Similarly, τ_r is calculated to be ~ 400 ns for the case of 3.2 mJ/cm^2 incident energy. However, the true rise time cannot exceed the length of the laser pulse, whose base width is about 90 ns. Again, the rise time of the data agrees well with this interpretation. It is also clear from the data in Fig. 56 that the emission associated with the higher flux exhibits a fast early-time decay and that the emission corresponding to the lower-flux data does not. Identification of the cause of this early-time fast decay will require further study.

Fig. 56. Photomultiplier signal of the $S(^1S - ^1D)$ emission vs time at two incident flux levels in 1.0-torr OCS with added buffer gas. For 45 mJ/cm^2 , 1500-torr N_2 was added to the OCS. For 3.2 mJ/cm^2 , 1500-torr of Kr was added.

The late-time decay of the $S(^1S)$ emission is typified by the curves presented in Fig. 57. These data were taken with a laser flux of about 30 mJ/cm^2 . The intriguing feature of these curves is that while each of the three OCS pressures gives nearly the same efficiency for $S(^1S)$ production at $t = 0$, each curve decays with a time constant that is inversely proportional to its $t = 0$ intercept.

Fig. 57. Photomultiplier signal of the $S(^1S - ^1D)$ emission vs times for an incident laser flux of 30 mJ/cm^2 . Each different OCS concentration included 1500 torr N_2 .

These time constants are not dependent on OCS pressure itself, but rather on the amount of OCS photolyzed (Fig. 58). Figure 58 shows the variation in decay frequency (τ^{-1}) with photolyzed OCS. The point denoted by * is for 1.0-torr OCS and a low laser flux. The decay frequency for that condition is considerably smaller than it is for lower initial OCS pressures with higher laser fluxes. Therefore, the species causing the late-time decay is not OCS or an

impurity contained in the OCS, but it is produced by the photolysis process. In addition, these data suggest that if the quenching agent is an excited species, it is long lived, lasting for at least 20 μ s, and that it is not consumed by the ^{1}S -quenching process.

Fig. 58. Decay frequency of the $\text{S}(^1\text{S} - ^1\text{D})$ emission vs the photolyzed amount of OCS. The open circles are for mixtures that include 1500-torr N_2 , the filled circle represents a mixture containing 500-torr N_2 , and the point labeled * is for 1.0-torr OCS and 1500-torr N_2 with a low laser flux (3 mJ/cm^2).

Microwave transmission Experiments -- First order calculations of the electron temperature distribution in a flash photolyzed OCS medium⁴¹ have suggested the possibility of a process in which electrons rapidly gain energy by superelastic collisions with $\text{S}(^1\text{S})$ atoms. After the electrons have gained enough energy to ionize $\text{S}(^1\text{S})$ they can generate more electrons which would repeat the process in a runaway fashion, rapidly leading to the quenching of most of the $\text{S}(^1\text{S})$ and production

of electron densities exceeding $10^{14}/\text{cm}^3$. However, the calculations also indicate that this process cannot be effective unless the excited OCS medium has an initial electron density of $10^{11} - 10^{10}/\text{cm}^3$. Photoionization of $\text{S}(^1\text{S})$ by F_2 laser photons represents an electron production mechanism which could quite possibly generate electron densities in excess of $10^{10}/\text{cm}^3$ by the end of the F_2 laser pulse. Therefore, we decided to investigate the time-dependence of the electron density in the laser excited OCS medium.

The most convenient method for us to obtain an approximate measure of the electron density in the OCS medium was to monitor transmission through the medium of 35-GHz microwaves. For this purpose microwave waveguides were set up to look transversely through the $\text{S}(^1\text{S})$ kinetics cell as shown in Fig. 54. Normally, both the transmitted microwave signal and a reference were observed to assure that the microwave power supply itself was not affected by the x-rays, RF interference, and magnetic fields associated with firing the F_2 laser.

For a microwave frequency of 35 GHz, one would normally expect the microwave transmission through the $\text{S}(^1\text{S})$ kinetics cell to be completely cut off by an electron density $\geq 1.5 \times 10^{13}/\text{cm}^3$. However, when a buffer gas at near atmospheric pressure is added to the medium, dephasing collisions between electrons and the buffer gas can shift the electron density at which complete cutoff occurs upwards by almost an order of magnitude. Thus, transmission cutoff in our experiments indicates electron densities that are in all cases greater than $2 \times 10^{13}/\text{cm}^3$ and in some cases (i.e., 760-torr Kr) greater than $\sim 10^{14}/\text{cm}^3$.

A summary of the microwave transmission data is shown in Table IV. For 0.8-torr OCS with no added buffer gas, the electron density immediately rose above the transmission cutoff level and remained above cutoff for $\sim 30 \mu$ s before decaying back to zero with a time constant of $\sim 50 \mu$ s. We consider these data to be strong evidence that the runaway electron process postulated

in the previous section is actually occurring. Adding any of the rare gases or H_2 or N_2 at the 1000-torr level caused a much more rapid quenching of the electron density, and in most cases prevented the electron density from rising to the cutoff level for experiments with 0.05- and 0.1-torr OCS in the kinetics cell. To test the hypothesis that an important property of the buffer gas in limiting the electron density is its electron attachment cross section, CF_4 was added to the kinetics cell. This molecule is presumed to have a large electron attachment cross section for a broad range of electron energies but has negligible optical absorption at 1575 Å. Addition of 400-torr CF_4 to 0.1-torr OCS in the kinetics cell reduced the electron concentration dramatically; in essence, the time history of the electron density followed the time history of the F_2 laser pulse.

TABLE IV

Summary of 35-GHz Microwave Transmission Data

Added Gas	Added Gas Pressure (torr)	F_2 Laser Energy (J/cm^2)	Decay Time of Electron Density (μs)
0.05 Torr OCS			
None	0	0.04	50
He	1000	0.05	15
Ne	1000	0.03	12
Ar	1000	0.05	10
Kr	1000	0.06	5
Ne	1000	0.05	1.5
H_2	1000	0.03	0.7
N_2	1000	0.05	1.5
0.1 Torr OCS			
He	1500	0.05	7
Kr	1500	0.05	3
H_2	1500	0.05	0.3
N_2	1500	0.04	0.6
CF_4	400	0.03	0.1

It is also clear that reducing the electron density dramatically lengthened the lifetime of the $S(1S)$ states in the afterglow of each F_2 laser pulse. For shots with comparable F_2 laser intensities at 0.2-torr OCS density, the decay time of the $S(1S)$ light was only ~ 500 ns with 55 torr of added Kr but was ~ 10 μs with 400 torr of added CF_4 and 1300 torr of added Kr.

Absolute Calibrations and Quantum Efficiency of $S(1S)$ Production

In order to determine the quantum efficiency for $S(1S)$ production by F_2 laser photons on OCS, four types of absolute calibrations were needed: (1) the absorption cross section of OCS for F_2 laser light and the behavior of the bleaching of OCS at high-laser-flux densities had to be determined, (2) the total flux of F_2 laser photons entering the $S(1S)$ kinetics cell had to be accurately measured, (3) the absolute efficiency at 773 Å of the photomultipliers used to detect the $S(1S)$ auroral emissions had to be determined, and (4) the transmission of the filters used in front of the PM tubes and the degree to which their transmission curves overlapped the $S(1S) - S(1D)$ emissions had to be evaluated.

The VUV absorption cross section and bleaching wave behavior of OCS are discussed in a previous section. The total energy of the F_2 laser beam was determined with a Scientec calorimeter. In-house calibration of the response of this calorimeter to a quartz iodide lamp with a known bolometric output led to a value of the calorimeter's sensitivity, which was within 8 percent of the value claimed by the manufacturer. During the experiments, the calorimeter was separated from the $S(1S)$ kinetics cell by a MgF_2 window. The transmission of this window was remeasured several times during the course of the experiments to assure that its transmission had not changed. After each major realignment of the F_2 laser - $S(1S)$ cell system,

several F_2^+ laser shots were taken with a blackened copper foil at the back of the evacuated $S(^1S)$ cell. Not all of the laser energy reaching the calorimeter was due to VUV lasing; therefore, shots were taken periodically with the $S(^1S)$ cell filled with room air to determine how much of the total laser energy was due to atomic fluorine laser lines between 6500 and 7500 Å.

The sulphur auroral emissions at 7727 Å were viewed with an RCA photomultiplier tube having a multi-alkali cathode or with a Varian photomultiplier tube having a Ga-As photocathode. Both of these detectors were calibrated against two standard quartz iodide lamps which had had their spectral irradiance calibrated by the Epply Research Laboratories. The values of quantum efficiency obtained with the two different lamps consistently differed from each other by 9 percent. This was probably due to the fact that while one of the calibration lamps was new, the other had been in use for several years. Quantum efficiency values obtained with the new lamp were used in the calculation of $S(^1S)$ production efficiencies. Values obtained for $S(^1S)$ production efficiencies by using the two different photomultiplier tubes were in agreement to better than 15 percent with much of the remaining uncertainty attributable to the difficulty in comparing results taken under slightly different experimental conditions. The experimental conditions under which the PM tubes were used were rather hostile, with intense red-laser light, electron-beam-produced x-rays, and a 45-Gauss magnetic field in close proximity to the PM tube. Tests of the PM tubes responses to a dc light source indicated that they were not significantly affected by the magnetic field, and tests with a Xe flashlamp (~2-μs pulse-width) indicated that while the x-rays and red laser contributed to the noise level observed during the F_2^+ laser pulse, the sensitivities of the tubes were not affected by the presence of this noise.

Two Corning glass cutoff filters and either of two interference filters were used in front of the PM tube. The transmission curves of all of the filters versus wavelength were measured on a Beckman I. R. Spectrophotometer. The

two Corning filters together transmitted 76 percent at 7727 Å and less than 1 percent below 7300 Å and were principally used to block scattered light from the red atomic fluorine laser. In addition, either a 7700-Å filter 100-Å-wide, or a 7727-Å filter 16-Å wide, were used to further isolate the $S(^1S \rightarrow ^1D)$ emission. The transmission curves of both of these filters are shown in Fig. 59. Due to the narrow half-width of the 7727-Å filter, this filter's transmission was checked at 7723.8 Å by using an argon lamp plus monochromator as a light source. This test indicated a transmission at 7723.8 Å slightly higher than the value obtained with the Beckman spectrophotometer indicating that the wavelength scale on the Beckman was probably shifted about 3 Å. A correction for this shift was made in data taken by using the 16-Å-wide filter, but this correction was negligible in data involving the 100-Å-wide filter.

Fig. 59. The curves labeled A, B, and C refer to the wavelength dependent transmission filters used in front of the photomultiplier as discussed in the text. The other curve is the $S(^1S \rightarrow ^1D)$ emission with 760 torr added N_2 reported by Black et al.⁴

As of February 1974, the only data available to us on the spectral width of $S(^1S) \rightarrow S(^1D)$ emissions was data taken at Sandia in 1975 on direct-electron-beam excitation of mixtures of 1-torr OCS and 2×10^4 torr Kr.⁴² These results showed a $S(^1S \rightarrow ^1D)$ emission feature with about a 20-Å FWHM. We assumed, therefore, that buffer gases that are lighter than Kr would provide narrower spectra which would be entirely included within the 16-Å pass band of our 7727 Å interference filter.

On the basis of these assumptions, we inferred $S(^1S)$ production efficiencies of 20 to 25 percent. Very recent data taken by Black, Sharpless, and Slanger⁴³ indicate that the spectrum of the $S(^1S \rightarrow ^1D)$ transition in most buffer gases is well over 100-Å wide and that the $S(^1S)$ spectrum with krypton as a buffer gas has the same width (~ 20 Å) at 55 torr that it has at 20,000 torr. As an example, their spectrum of $S(^1S \rightarrow ^1D)$ emission with 760 torr with added N_2 is shown in Fig. 5b, together with the transmission curves of the filters used in these experiments. Very recently, we have also taken $S(^1S)$ emission data with the 100-Å-wide interference filter using N_2 , H_2 and Kr as buffer gases at pressures for which Black et al.⁴³ have determined the $S(^1S)$ induced spectrum. Folding our filter transmissions into the observed spectra, we obtain the production efficiencies listed in Table V.

For the data taken with N_2 as a buffer gas, the $S(^1S)$ signal levels were large enough that it was possible to accurately determine the $S(^1S)$ density during the F_2 laser pulse. This is important in determining production efficiency as the $S(^1S)$ density typically exhibited a fast early-time decay. For H_2 and Kr, the signal levels were relatively low so that the $S(^1S)$ density could only be determined after the laser pulse and its accompanying PM tube noise were over. Thus the production efficiencies for mixtures with H_2 and Kr as buffer gases should be considered only as lower limits to the true production efficiency.

The only emission from the excited medium in the $S(^1S)$ cell visible to the eye was a deep blue. Investigating these emissions with a spectrograph and film, we determined that these bands were $^3\Sigma^- \rightarrow ^3\Sigma^-$ emissions from S_2 ending on the ground electronic state. These bands are known to have a radiative decay time on the order of 100 ns. Using a photodiode behind a blue-color filter and using manufacturer's calibration curves for the diode efficiency and filter transmission, we obtained a preliminary value of $\sim 10^{13}/\text{cm}^3$ for the total number of S_2 molecules created during

TABLE V
Production Efficiency of $S(^1S)$ from OCS by vuv F_2^+
Laser Photons

OCS Pressure (torr)	Added Gas	Added Gas Pressure (torr)	FWHM of Filter Used (Å)	$S(^1S)$ Production Efficiency (%)	Decay Time of $S(^1S)$ Emissions (Ye) (μs)
0.2	N_2	810	100	84	12
0.2	N_2	810	16	79	12
1.0	N_2	1500	16	91	1.5
0.2	H_2	760	100	65	2
0.2	Kr	55	100	56	0.5

an excitation pulse of the OCS medium. Since the $S(^1S)$ densities produced in the kinetics cell were on the order of $10^{15}/\text{cm}^3$ we conclude that the formation of S_2 cannot represent a significant loss mechanism of $S(^1S)$ atoms.

Photoionization of Sulphur $(3p)^4\ ^1S$ Via Autoionizing Levels

In systematic close-coupling calculations of photoionization of atoms with configurations $(3s)^2(3p)^4$, Connely, Smith and Lipsky,⁴ hereinafter (CSL), determined the position and widths of the $(3s)^2(3p)^3\ ^2D(nl)$ and $(3s)^2(3p)^3\ ^2P(nl)$ autoionizing levels of sulphur. With these they calculated the photoionization cross section from $(3s)^2(3p)^4\ ^3P$, 1D , and 1S ground state terms. Tondello⁴⁵ later measured many of the autoionization structures via UV photoabsorption from the ground state. One might then think that the sulphur $(3p)^4\ ^1S$ photoionization cross section in the UV is established. Unfortunately this is not the case. The agreement between the calculation and measurement for $(3p)^4\ ^3P$ photoabsorption is poor. Further, an examination of the CSL calculations indicates that they considered $(3p)^4\ ^1S - (3p)^3\ ^2P(nl)\ ^1P$ transitions only. CSL did not include the $(3s)^1(3p)^5\ ^1P$ and 3P odd parity levels.

The $(3s)^2(3p)^4\ ^1S - (3s)^1(3p)^5\ ^1P$ transition has a large oscillator strength but the transition is at a wavelength $< 1000\ \text{\AA}$. However in a 1-electron model, the calculated Auger decay rate for $(3s)^1(3p)^5\ ^1P - (3s)^2(3p)^3\ ^2D(\epsilon l)$ 1P and $(3s)^2(3p)^3\ ^2P(\epsilon l)\ ^1P$ leads to a $(3s)^1(3p)^5\ ^1P$ level width of about 13 eV. The calculated line width is comparable to the energy above the ground state, and the 1-electron calculation is inconsistent with conservation of energy, since the final state in the Auger decay process $(3s)^2(3p)^3\ ^2D(\epsilon l)\ ^1P$ is well above the ground state.

To go beyond the 1-electron picture, one needs either a many-body or a configuration interaction (CI) calculation. To motivate the CI calculation we consider a simple model. Suppose we have a ground state O, and two

excited states in the continuum, a and b. Suppose the oscillator strength between O and b is f_{ob} . Suppose state b can decay to state a, and the linewidth is Γ_{ba} which is much larger than any radiative width. Then the oscillator strength f_{ob} is distributed in energy as a Lorentzian with width Γ_{ba} . This is a standard description of x-ray absorption. But if the energy difference $E_b - E_a \approx \Gamma_{ba}$ one must truncate the oscillator strength distribution at E_a , since otherwise the low-energy wing of the oscillator strength distribution would require a negative energy continuum electron. But if we truncate the oscillator strength distribution at E_a , what happens to the missing oscillator strength? The answer is that when the electrostatic interaction between b and a is large, then it is large between state b and other levels with the same quantum numbers as level a. In the case of sulphur there is a large electrostatic matrix element between $(3s)^1(3p)^5\ ^1P$ and $(3s)^2(3p)^3\ ^2D(\epsilon l)\ ^1P$, but then there are large matrix elements between $(3s)^1(3p)^5\ ^1P$ and $(3s)^2(3p)^3\ ^2D(\epsilon l)\ ^1P$. This latter is a strong configuration interaction and it was neglected in CSL. As a consequence of the strong CI two effects are possible. One, the level b can be shifted to an energy below the continuum limit; it retains most of the o-b oscillator strength, but now the b-a Auger transition is energetically forbidden and so the level b has only a radiative width. As a consequence of the CI some other levels must be shifted to a higher energy. Those that are shifted into the continuum should have either a small oscillator strength or a small linewidth to recover an oscillator state distribution consistent with energy conservation in the autoionization process. This scenario has been verified for the $(ns)^1(np)^6\ ^2S$ level in the halogens by Cowan et al⁴⁶ and Hansen⁴⁷, by using different approximate Hartree-Fock techniques. Comparison with levels in Moore's⁴⁸ tables shows that either ab initio technique is accurate to only $5000\text{--}10000\ \text{cm}^{-1}$.

The other possible consequence is that level b interacts so strongly with levels both lower and higher in energy than itself, so that, while level b remains in the continuum, the

oscillator strength is strongly redistributed to either discrete-levels or to continuum levels which autoionize weakly. This scenario has been verified to occur for 4p photoionization in Xe. The analysis was done by Wendin and Ohno,⁴⁹ by using many-body theory techniques. This approach does not appear sufficiently accurate to use in calculating the group VI (np)⁴ 1S photo-absorption.

In our sulphur calculations, we include 67 levels with $j = 1$; the (3s)¹(3p)⁵ 1P and 3P levels, 4 levels from each (3s)²(3p)³(ns) configuration, with $4 \leq n \leq 8$, and 9 levels from each (3s)²(3p)³(nd) configuration with $3 \leq n \leq 7$. We include all CI terms among these discrete levels via CI interaction matrices corrected for Brillouin's theorem.⁵⁰ The details can be found in a forthcoming publication. The autoionization rates can be found by inserting continuum matrix elements in the CI interaction matrices. Spin-orbit interaction was included for the 67 levels, as well as for the ground state terms of S and S'. The small admixture of (3p)⁴ 3P₀ in (3p)⁴ 1S₀ has a prominent role in the photoionization. We used two electron matrix elements calculated with Herman-Skillman-like orbitals.⁵¹ However, the initial energy positions were obtained by using experimentally measured⁴⁸ effective principle quantum numbers. In addition, we determined the initial position

of the (3s)¹(3p)⁵ 3P and 1P levels by diagonalizing the CI matrix with the (3s)¹(3p)⁵ configuration energy as a parameter. The final choice of (3s)¹(3p)⁵ configuration energy was that initial value which, after diagonalization of the CI matrix, brought the (3s)¹(3p)⁵ 3P₁ level into reasonable agreement with the experimental value.⁴⁸

In Table VI, we list a level number, the level designation, the initial estimate of the term energy (measured relative to the S' (3s)²(3p)³ 4S term), the result of the CI matrix diagonalization (K_f), and experimental values from Moore's tables⁴⁸ and Tondello's⁴⁵ measurements. In the next column are energy positions calculated by CSL,⁴⁴ but we have changed their designations, which appear incorrect. In the final three columns, we list our calculated line-widths, those measured by Tondello,⁴⁵ and those calculated by CSL.⁴⁴

Our calculated photoionization cross section for sulphur (3p)¹ 1S between 1629 Å and 1171 Å is shown in Fig. 60. Between 1500 and 1600 Å, there is only one absorption band; unfortunately it is located near the F₂ laser output. Below 1500 Å, the cross section is dominated by autoionization structure.

TABLE VI
Spectroscopic Parameters for Odd-Parity, $J = 1$ Levels Sulphur

Level No.	Designation	K _i (cm ⁻¹)	K _f (cm ⁻¹)	K _{exp} (Moore)	K _{exp} (Tondello)	K (CSL)	Γ (cm ⁻¹) (calc)	Γ (cm ⁻¹) exp	Γ (CSL)
1	(3p) ³ 4S(4s) 3S	-27071	-27259	-28228					
2	2D 3D	-15101	-15740	-15729	-15744				
3	4S(3d) 5D	-15681	-15394	-15674					
4	3D	-14146	-14255	-13369	-13393				
5	(5s) 3S	-11889	-11885	-12206					
6	(3s) ¹ (3p) ⁵ 3P	-4225	-11398	-11259					
7	(3p) ³ 4S(4d) 5D	-8585	-8607	-8583	-8585				

(cont)

TABLE VI
(cont)

Level No.	Designation	K_i (cm ⁻¹)	K_f (cm ⁻¹)	K_{exp} (Moore)	K_{exp} (Tonjello)	K (CSL)	Γ (cm ⁻¹) (calc)	Γ (cm ⁻¹) exp	Γ (CSL)
8	³ D	-7669	-7752	-7606					
9	(6s) ³ S	-6706	-6697	-6838	-6839				
10	2p(4s) ³ P	-5412	-5006	-6399	-6377				
11	4s(5d) ⁵ D	-5287	-5293	-5239					
12	2p(4s) ¹ P	-3478	-4399	-5269					
13	4s(5d) ³ D	-4756	-4772	-4867	-4867				
14	(7s) ³ S	-4305	-4299	-4373	-4374				
15	(6d) ⁵ D	-3567	-3564	-3567					
16	³ D	-3228	-3210	-3376	-3375				
17	(8s) ³ S	-2993	-2985	-3037	-3037				
18	(7d) ⁵ D	-2563	-2537	-2564					
19	³ D	-2295	-1175	-2177	-2469				
20	2p(3d) ³ D	-299	-323		123	1514			724
21	¹ P	-71	-500						
22	³ P	256	2074		2927		0.89		
23	(5s) ³ D	2258	2232		2121	2764	80.6	81	102
24	(3d) ³ S	631	567		1872	2534	179.0	95	59
25	(4d) ³ D	6590	6586		6939	7239	639.0		582
26	¹ P	6726	6041				0.49		
27	³ P	6916	7723		7935		0.002		
28	(6s) ³ D	7872	7837		7822	8182	50.3	100	19
29	(4d) ³ S	7130	7105		7628	7974	93.0	62	16
30	(5d) ¹ P	9297	8896				0.19		
31	³ D	9852	9857		10338	9838	28.3		123
32	¹ P	9949	10651				0.95		
33	(7s) ³ D	10415	10391		10338	10507	3.23		33
34	(5d) ³ S	10073	10077			10485	57.9		13
35	(6d) ³ D	11415	11405				233.0		
36	(3p) ³ ² D(6d) ¹ P	11466	11209				0.12		
37	³ P	11537	13972				0.11		
38	³ S	11616	11639				42.8		

(cont)

TABLE VI
(cont)

Level No.	Designation	K_i (cm^{-1})	K_f (cm^{-1})	K_{exp} (Moore)	K_{exp} (Tondello)	K (CSL)	$\Gamma(\text{cm}^{-1})$ (calc)	$\Gamma(\text{cm}^{-1})$ exp	Γ (CSL)
39	(8s) 3D	11789	11757				10.4		33
40	(3p) $^3P(3d)$	3P 9234	9547				1.49		
41	3D	9718	9818		10338		1053.0		
42	(5s) 3P	11947	11915		11631		0.31		
43	$^2D(7d)$	1P 12436	12236				0.18		
44	3D	12444	12447				214.0		
45	$^2P(5s)$	1P 12430	12672		12082		3.47		
46	$^2D(7d)$	3P 12493	12044				0.065		
47	3S	12554	12648				64.8		
48	$^2P(3d)$	1P 10987	10680		12316		0.40		
49	(4d) 3P	16186	16811				3197.0		
50	3D	16415	16478		16401		482.0		
51	(6s) 3P	17561	17591		17461	17747	184.0		800
52	1P	17756	17843			17889	88.5		30
53	(4d) 1P	17295	17081				1.76		
54	(5d) 3P	19392	19694			19775	1078.0		55
55	3D	19532	19580		19587		308.0		
56	(7s) 3P	20104	20124		20068	20203	52.7		47
57	1P	20204	20274			20269	72.2		16
58	(5d) 1P	20040	19926				6.81		
59	(6d) 3P	21070	21269			21289	578.0		44
60	3D	21155	21195		21186		214.0	79	
61	(8s) 3P	21472	21492		21461	21530	24.4		17
62	1P	21533	21604			21464	73.7		9
63	(6d) 1P	21482	21432				18.3		
64	(7d) 3P	22058	22208				490.0		
65	3D	22125	22154		22116		193.0	61	
66	1P	22383	22556				725.0		
67	(3s) $^1(3p)^5$	1P 42656	45853				108550.0

Fig. 60. The contribution of autoionizing levels to the photoionization cross section of sulphur $(3p)^4 1S$ for $1628 \text{ \AA} \geq \lambda \geq 1171 \text{ \AA}$. The final states are designated in accordance with Table III.

A blowup of the photoionization between 1558 and 1592 Å is shown in Fig. 61. The dashed curve comes directly from the input in Table VI. However, Tondello⁴⁵ has measured the position of the two levels contributing to the photoionization in this region, levels 22 and 23. By varying the input parameters for levels 22 and 23, until the output of the CI diagonalization leads to energy levels within 5 cm^{-1} of Tondello's values, we find the solid curve in Fig. 61.

Our conclusion is that while sulphur $(3p)^4 1S$ photoionization is substantial at 1575 Å ($\sigma = 4 \times 10^{-18} \text{ cm}^2$), it is negligible at 1565 Å ($\sigma < 10^{-19} \text{ cm}^2$). Note also that for level 22 leading to the peaked photoionization at 1575 Å, our calculated linewidth (80.6 cm^{-1}) is in excellent agreement with Tondello's⁴⁵ measured value (81 cm^{-1}).

The difference between the dashed and solid curves indicates the importance of ground state photoionization measurements of Group VI elements in locating autoionization structure, which can then be used to fine-tune calculations.

Fig. 61. Photoionization cross section from sulphur $(3p)^4 1S$ due to autoionization for $1592 \text{ \AA} \geq \lambda \geq 1557 \text{ \AA}$. The dashed curve results from using K_f in Table III, while the solid curve results from varying K_f to bring K_f into 5 cm^{-1} agreement with the measurements of Tondello.

A blowup of the sulphur $(3p)^4 1S$ photoionization cross section in the 1449-1480 Å range is shown in Fig. 62. Again, the dashed curve is directly from Table VI, while the solid curve arises from adjusting the calculation to Tondello's measured values for levels 25 and 29. The cross section is broad; the calculated linewidth for level 25 is 639 cm^{-1} . Note in Table VI that this is close to the value calculated by CSI. (582 cm^{-1}). Since level 25 is $(3p)^3 2D(4d)^3 1D$, it is likely that the neglect of CI via $(3s)^4(3p)^5 1P$ and $3P$ by CSI, does not significantly modify their line-width calculation. Thus, we have good reason to believe that near 1465 Å, the sulphur $(3p)^4 1S$ photoionization cross section is dominated by a broad peak with $\sigma \approx 3 \times 10^{-19} \text{ cm}^2$. These calculations are being extended to selenium and tellurium.

Fig. 62. Photoionization cross section from sulphur $(3p)^4 1S$ due to autoionization for $1479 \text{ Å} \geq \lambda \geq 1449 \text{ Å}$. The dashed curve results from using K_I in Table III, while the solid curve results from varying K_I to bring K_f into 5 cm^{-1} agreement with the measurements of Tondello.

Summary and Conclusion

The F_2 molecular laser is operating reliably as an excitation source at 157 nm at laser energy levels variable from $< 1 \text{ mJ/cm}^2$ to 100 mJ/cm^2 . Our initial attempts to further increase the efficiency and output energy of the F_2 laser resulted in a system which was dominated by single-pass amplified spontaneous emission.

Initial experiments in which the F_2 laser beam pumped pure OCS mixtures at 0.1 to 1.0 torr pressure revealed the existence of an avalanching process which quickly drove the electron density above $\sim 2 \times 10^{13}/\text{cm}^3$. The electron density remained above this level for tens of microseconds. A very fast decay of the $S(1S)$ density was observed under these conditions indicating that quenching of $S(1S)$ was an integral part of the electron avalanche process.

Addition of buffer gases to the OCS mixtures, particularly buffer gases with high electron attachment or energy-loss cross sections prevented the electron density from reaching $2 \times 10^{13}/\text{cm}^3$ and caused the remaining electrons to disappear with decay times from 100 ns to a few microseconds. Under these conditions and at relatively high F_2 laser fluxes ($40\text{--}60 \text{ mJ/cm}^2$), the $S(1S)$ emissions exhibited two distinct time decays. During the laser pulse, a fast-time decay was observed. The fraction of the $S(1S)$ quenched during this time decay appears, qualitatively, to increase as the initial $S(1S)$ density increases. After the end of the F_2 laser pulse, the $S(1S)$ density decays at a much slower rate which also was proportional to the initial $S(1S)$ density. The fact that the slow-decay rate depended only on the initial $S(1S)$ density rather than on the initial OCS or buffer gas pressures indicates that the quenching agent is a daughter product of the photoionization of OCS.

For cases in which the electron density was well-controlled, we observed $S(1S \rightarrow 1D)$ signals indicating that $S(1S)$ was being produced by photolysis of OCS with at least 80 percent efficiency. These results, which were obtained with initial $S(1S)$

densities of $\sim 2 \times 10^{15}/\text{cm}^3$, are in good agreement with earlier published results,⁵² which measured high production efficiencies at S^{18} densities which were many orders of magnitude lower.

Calculated S^{18} photoionization cross sections in the VUV imply limits on the VUV excitation flux for an S^{18} laser of $\sim 10^{17}$ photons/ cm^2 and 10^{18} photons/ cm^2 for F_2 and Kr_2 pumps, respectively. This in turn implies that the most successful pumping scheme for an S^{18} laser will be one which minimizes the flux of pump photons throughout all parts of the laser medium.

OSCILLATOR TECHNOLOGY

Overview

The objective of this program is to design, construct, and test highly reliable laser oscillator systems. Currently, it is felt that most wavelength regions of interest to OLF can be generated by using a laser oscillator system based on a microprocessor-controlled Nd:YAG-pumped dye laser system. These oscillator systems cannot only be used as the front end of a laser fusion device, but they can be used in testing and making germane laser amplifier measurements of candidate laser systems in the wavelength region of 4000 to 8000 Å and extended range with harmonic generation and frequency mixing.

The design characteristics of such an oscillator system include long-term stability, high reliability, hands-off operation, and variable pulse-width generation in the nanosecond regime. These requirements necessitate the application of microprocessor technology to laser technology, thereby resulting in an advanced state-of-the-art laser system capable of satisfying all of the above requirements.

Nd:YAG Pump Source

The Nd:YAG oscillator/amplifier dye laser pump source, operating at 10 pps, has been assembled and tested. The operating parameters are as follows: pulse width - 10 ns, energy at 355 nm - 30 mJ, energy at 532 nm - 200 mJ, and energy at 1.064 microns - 700 mJ. The 532-nm pump wavelength will be utilized for dyes such as Rhodamine 6G in the 5600 to 6000 Å regime while the 355-nm pump wavelength will be used for the various blue wavelength dyes.

Absolute Wavelength Meter

A microprocessor controlled wavelength meter, based on a wedged interference plate, has been designed. It is anticipated that wavelength accuracies of about 10^{-7} will be realized. The software for this system has been tested and it has been found to take 2 seconds of running time to yield a wavelength calibration from a single laser pulse. Software techniques to reduce this time are currently being investigated.

Absolute Wavelength Controller

A microprocessor based Fabry-Perot dye laser wavelength controller has been designed. Software design is currently under way.

Computer System Design

An overall microprocessor based computer system design concept has been formulated and steps are being made to test these concepts. Basically, a master microprocessor system will be used to communicate with and give instructions to several microprocessor "slave" controllers, such as the absolute wavelength meter, the wavelength controller, and the dye-laser microprocessor controller.

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