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13. ABSTRACT (Maximum 200 words) The focus of this program is the development of extreme ultraviolet (XUV) lasers and their application to scientific problems. Laser development concentrates on the laboratory-scale lasers near 100 nm. The current application area is the development of very high resolution, microscopic imaging techniques. Such techniques should have a number of applications and be particularly useful in the life sciences. This report presents recent progress on XUV laser development.				
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Development and Application of XUV Lasers

Final Technical Report

for the period

1 June 1990 — 30 November 1993

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1. Introduction

This program supported two related types of activities because there was a change of Principal Investigator (PI) during the period. The work of the original PI, Dr. P. J. K. Wisoff, concentrated on the development of short-pulse, blue-green dye laser systems, and their amplification to terawatt levels in a XeF($C \rightarrow A$) electron-beam pumped, excimer amplifier system. This research, initiated under an earlier AFOSR grant, was very successful and of significant interest to the scientific community, as indicated by the list of publications and presentations in Section XX. Early in the first year of the grant period, Dr. Wisoff left Rice University to join the NASA Astronaut program. Dr. J. F. Young joined the Rice faculty at that time and took over direction of the program. In collaboration with Dr. Wisoff, he continued the existing projects to natural and productive conclusions, while initiating a new, but related, program focused on the development of extreme ultraviolet (XUV) lasers and their application to scientific problems. Laser development concentrated on the laboratory-scale Xe and H₂ lasers near 100 nm. The current application area is the development of very high resolution, microscopic imaging techniques. Such techniques should have a number of applications, and be particularly useful in the life sciences. This report reviews both areas of activities.

2. Participating Personnel

Senior Personnel

James F. Young, Principle Investigator and Associate Professor; Summer salary only.

Dr. Csaba Tóth, Visiting Associate from the Research Institute for Solid State Physics, Budapest, Hungary.

Graduate Students

Ms. Tracy Sharp, awarded Ph.D., May 1993.

Mr. Marc Byrd.

Mr. Jun Wu.

Mr. Lim Nguyen, Air Force civilian employee, participant in the Palace Knight program, at Phillips Laboratory.

Mr. E. F. Ejeckam.

Mr. Tasshi Dennis.

Undergraduate Students

S. Hareland.

V. Cordrey.

Honors

James F. Young

Named IEEE Lasers and Electro-Optic Society Distinguished Lecturer for 1991-1992.

Promoted to Associate Professor with Tenure, Rice University, May 1992.

Elected Senior Member of the IEEE, August 1992.

Tracy E. Sharp

Awarded the L. S. Vaughn Graduate Fellowship for students exhibiting outstanding academic and professional potential, Rice University, 1991-1992.

Named an IBM Graduate Fellow, 1992-1993.

Picked for Director Fellowship, Los Alamos National Laboratory, 1993.

3. Program Highlights

- The dispersion of optical fibers around 500 nm was measured, and the data used to construct a fiber-prism pulse compressor. This was the first use of this technique in the blue-green spectral region.
- A two-stage dye amplifier for blue-green, subpicosecond pulses was designed to supply injection pulses to the XeF($C \rightarrow A$) excimer amplifier.
- The gain, energy output, saturation, and beam profile of the e-beam excited XeF($C \rightarrow A$) excimer amplifier was measured for various injection pulse lengths.
- Self-focusing effects at high fluences in the XeF($C \rightarrow A$) amplifier were studied, and a multipass, unstable resonator was constructed to minimize them.
- Lens-zone plate combinations were designed for achromatic focussing of ultrashort pulses.
- The transmission properties of glasses for high intensity visible pulses were measured.
- A practical, high repetition rate, 109 nm laser source pumped by an all-commercial laser system was constructed; it achieves saturated output power at a lower input pump power than any previous system.
- A reconfigured short-pulse injection source has been used to investigate the short-pulse characteristics of ArF excimer amplifiers. Small signal gain and saturation energy densities were measured.
- The transverse beam shape of the 109 nm laser was measured.
- An apparatus to measure the optical properties of thin films in the XUV was designed and constructed. An initial study of the absorption depth of PMMA was made.
- First observation of gain in a new class of excimer molecules, the alkali halide ionic excimers, specifically $Cs^{2+}F^{-}$, at 185 nm.
- Initial investigations of the time dependence of the gain of the Xe laser were made.

4. Blue-Green, Short-Pulse, High-Power Laser Systems

The realization of practical VUV and XUV laser sources will depend on the development of high power excitation technologies that are suitable for small, as well as large, laboratories. Laser-produced plasmas provide a convenient method of excitation and have been used to generate several VUV lasers. The extension of this technique to extremely short wavelength lasers will require very high power, ultrafast laser pulses in order to produce sufficient upper state densities for gain within the short lifetime of the excited state. We have studied a new class of ultrashort laser pulse amplifiers capable of directly amplifying pulses of about 10 fsec duration, and having high energy storage capability.

This new approach to femtosecond laser pulse amplification is based on broad bandwidth excimer transitions, in particular the XeF($C \rightarrow A$) transition. The properties of this gain transition have been previously studied at Rice using nanosecond dye laser pulse injection. These studies indicated that the large bandwidth of the transition should be capable of directly amplifying laser pulses of less than 10 fsec duration, with high energy storage and low amplified spontaneous emission.

Under this program we have constructed tunable, blue-green, subpicosecond source of laser pulses for injection into the XeF($C \rightarrow A$) amplifier, and measured the amplifier characteristics for short pulses. A two-stage dye amplifier was developed to amplify subpicosecond dye laser pulses that are generated from a hybrid, synchronously mode locked, dye oscillator. The pulses from the oscillator are either used directly (800 fsec), or shortened to 250 fsec by a fiber-prism compressor stage. This system has achieved single pulse energies of 2 mJ with an amplified spontaneous emission content of less than 0.1%.

Using this injection source, the gain characteristics of an electron-beam pumped XeF($C \rightarrow A$) excimer amplifier operating in the blue-green spectral region were investigated for several wavelengths. Saturation densities of 50 mJ cm^{-2} and 80 mJ cm^{-2} were measured for injected laser pulse durations of 250 fsec and ~ 100 psec, respectively. A gain bandwidth of 60 nm was observed. Using an optimized unstable resonator design, the laser amplifier produced 275 mJ pulses with a duration of 250 fsec, and a 2.5-times diffraction limited beam quality, making the XeF($C \rightarrow A$) amplifier the first compact laser system in the visible spectral region to reach peak powers of the terawatt level.

The short-pulse injection source was also reconfigured to provide pulses at 193 nm in order to investigate the short-pulse characteristics of ArF excimer amplifiers. The ArF laser, one of the most efficient excimer lasers in the nanosecond regime, offers a variety of potential new applications because of its short wavelength. To date, however, ultrashort pulse ArF excimer systems have shown significantly lower performance. In particular, the design of high power ArF systems is impeded by the fact that the saturation energy density for femtosecond pulses has not been previously determined. We modified the short-pulse injection source and combined it with a newly developed, spectrally compensated sum frequency mixing scheme to make such measurements.

The gain characteristics of a discharge pumped ArF excimer amplifier were measured with 700 fsec long pulses from the new injector system. The small signal gain and saturation was found to be 0.17 cm^{-1} and $3.7 \pm 0.15 \text{ mJ cm}^{-2}$, respectively. Output energy

densities as large as 10 mJ cm^{-2} were extracted from the highly saturated amplifier. These measurements indicate that by using sufficiently high seed pulse energies, the ArF excimer laser is capable of performance similar to a femtosecond, high brightness KrF excimer system.

Additional experimental details and results from these projects may be found in the papers listed in Section 7.

5. XUV Lasers and Applications

There are many exciting potential applications of coherent XUV sources in fields such as photochemistry, material science, atomic physics, and the life sciences. Such applications require a convenient laser source that can be easily assembled and that can operate in the laboratory with a minimum of attention. Under this program we have made a significant advance toward this goal: the first XUV laser system pumped by a standard, commercially available, Nd:YAG laser system at high repetition rate. The XUV laser utilizes the laser-produced plasma pumping method that has been used for several XUV gain systems. A grazing incidence, traveling-wave pumping geometry is used to reduce the pump energy required to achieve saturated energy outputs. Our work to date involves the Xe Auger laser at 109 nm, and is described in the preprint "A Reasonably Practical XUV Laser for Applications," included in Section 8.

We have made the first observation of gain in a new class of excimer molecules, the alkali halide ionic excimers. The alkali halide ionic excimers are isoelectronic to the well-known rare gas halides and have emission wavelengths in the range from 210 nm to 130 nm. Soft x-rays from a laser-produced plasma were used to photoionize CsF vapor and produce excited state $\text{Cs}^{2+}\text{F}^{-}$; an average gain coefficient of 0.3 cm^{-1} at 185 nm was measured over a 6 cm length. This is the first demonstration of a new type of molecular ultraviolet laser with broad bandwidth and large saturation intensity. The details of this work are given in the reprint "Optical gain in the ionic excimer $\text{Cs}^{2+}\text{F}^{-}$ excited by soft x-rays from a laser-produced plasma," included in Section 8.

The characterization and improvement of XUV recording films is a critical aspect of developing high resolution microscopic imaging systems. Very little data on the XUV optical properties of materials is available because of a lack of sources and the experimental difficulties of working in this region. We have developed an apparatus to measure the absorption depth of thin films in the XUV. Such basic information is necessary, for example, to choose polymer compounds for films that will allow the XUV radiation to penetrate and initiate photochemical reactions to record the intensity pattern. The wavelength region of interest is from 200 nm to 30 nm. The long wavelength end is chosen to connect to the available UV data, while beyond 30 nm, reliable calculations are available. The basic experiment is shown in Fig. 2. A laser-produced plasma is used as a bright, broad-band light source, and a 0.2 m vacuum spectrometer provides wavelength selectivity and tuning. A thin film of the sample is placed at the exit slit of the spectrometer and the transmission is compared to a reference spectrum without the sample. Laser-produced plasmas using high atomic weight targets have a continuous spectrum often modeled as a black body having a temperature of about 25 eV. Figure 3 shows our initial measurements of the absorption spectrum of PMMA, the film that we have used to date. Also shown are some

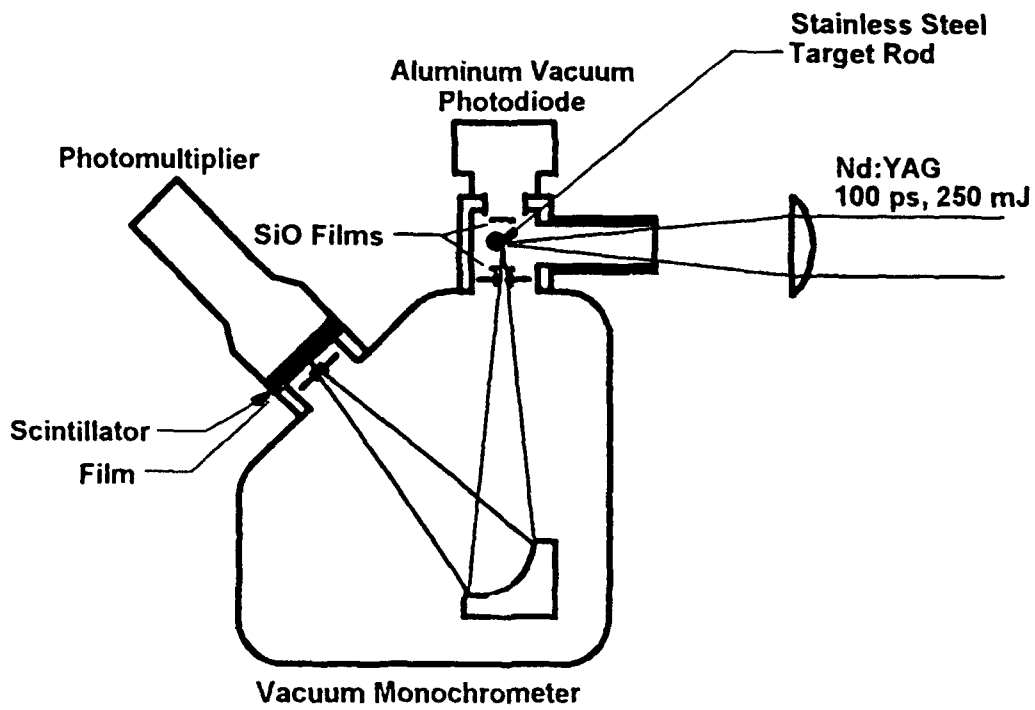


Fig. 1—Schematic of apparatus to measure absorption of films in the XUV.

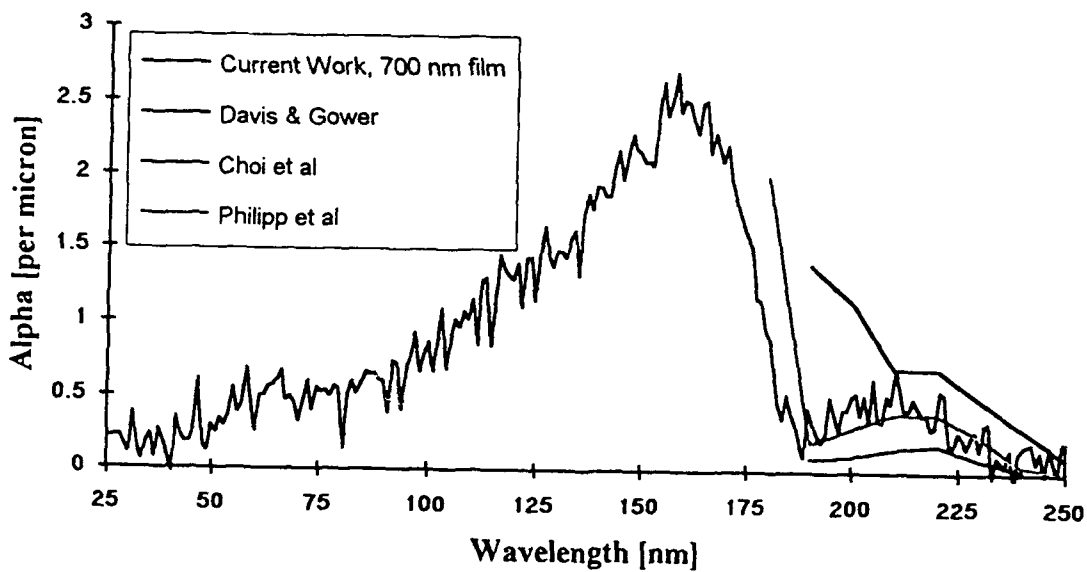


Fig. 2—Absorption coefficient of PMMA

previous measurements at long wavelengths. For optimum sensitivity and good dynamic range, the absorption depth should be on order of the wavelength, and the data shows that the characteristics of PMMA are a good match for the Xe and H₂ lasers. Unfortunately, PMMA is also quite an insensitive film. We will use our absorption apparatus to investigate the properties of other, higher sensitivity, films.

6. Relationship to Other Programs and Support

The excimer amplifier studies have been done in collaboration with Prof. Frank Tittel of Rice, and Prof. Gabor Szabó, a visitor from JATE University, Hungary. The electron-beam facility is supported by ONR.

The Cs²⁺F⁻ investigations were jointly supported by NSF. The Xe laser development has been jointly supported by Rice University, in the form of start-up funds to the PI.

7. Publications and Presentations of Supported Work

Journal Articles

- "High intensity transmission of optical glasses," T. E. Sharp, F. K. Tittel, P. J. Wisoff, and G. Szabó, *Appl. Optics* (submitted).
- "Characterization of an ultrahigh peak power XeF(C → A) excimer laser system," Th. Hofmann, T. E. Sharp, P. J. Wisoff, W. L. Wilson, F. K. Tittel, and G. Szabó, *IEEE J. Quantum Electron.* **28**, 1366–1375 (1992).
- "Analysis of lens and zone plate combinations for achromatic focusing of ultrashort laser pulses," T. E. Sharp and P. J. Wisoff, *Appl. Opt.* **31**, 2765–2769 (1992).
- "Blue-green dye laser seeded operation of a terawatt excimer amplifier," F. K. Tittel, Th. Hofmann, T. E. Sharp, P. J. Wisoff, W. L. Wilson, and G. Szabó, in *The Dye Laser: 25 Years*, Michael Stuke, ed., (Springer-Verlag: Berlin, 1992) pp. 141–152.
- "Ultrashort-laser-pulse amplification in a XeF(C → A) excimer amplifier," T. E. Sharp, T. Hofmann, C. B. Dane, W. L. Wilson, Jr., F. K. Tittel, P. J. Wisoff, and G. Szabó, in *Selected Papers on Ultrafast Laser Technology*, T. R. Gosnell and A. J. Taylor, eds., (SPIE: Bellingham, Washington, 1991).
- "Dispersion measurements of single-mode fibers in the blue-green spectral region by an interferometric method," G. Szabó, T. E. Sharp, F. K. Tittel, and P. J. Wisoff, *Appl. Optics* **30**, 5224–5225 (1991).
- "Tunable, high-power, subpicosecond blue-green dye laser system with a two-stage dye amplifier," T. E. Sharp, C. B. Dane, D. Barber, F. K. Tittel, P. J. Wisoff, and G. Szabó, *IEEE J. Quant. Elect.* **27**, 1221–1227 (1991).
- "Ultrashort-laser-pulse amplification in a XeF(C → A) excimer amplifier," T. E. Sharp, T. Hofmann, C. B. Dane, W. L. Wilson, Jr., F. K. Tittel, P. J. Wisoff, and G. Szabó, *Opt. Lett.* **15**, 1461–1463 (1990).
- "Optical Gain in the Ionic Excimer Cs²⁺F⁻ Excited by Soft X-Rays from a Laser-Produced Plasma," Cs. Tóth, J. F. Young, and R. Sauerbrey, *Optics Letters* **18**, 2120–2122 (1993).

"A Reasonably Practical XUV Laser for Applications," T. E. Sharp, Cs. Tóth, J. Wu, and J. F. Young, *IEEE J. Quantum Electron.*, accepted for publication.

"Laser-Produced Plasmas as Short Wavelength, Incoherent Optical Sources," J. F. Young, in *Methods of Experimental Physics: Atomic, Molecular, and Optical Physics*, F. B. Dunning and R. G. Hulet, eds., (Academic Press, due 1994) Vol. III.

Selected Presentations

"First Observation of Gain in CsF Ionic Excimer Molecules at 185 nm," Cs. Tóth, J. F. Young, and R. Sauerbrey; Optical Society of America 1993 Annual Meeting, Toronto, Canada, October 1993.

"Characteristic Studies of an XUV Xe Laser at 109 nm," J. Wu, T. E. Sharp, Cs. Tóth, and J. F. Young; Seventh Annual Rice Quantum Institute Research Colloquium, Rice University, Houston, TX, August 1993.

"New Methods of Pumping Alkali-Halide Ionic Excimers: Direct Vaporization and Quasi-Traveling Wave Geometry," V. Cordrey, Cs. Tóth, J. F. Young, and R. Sauerbrey; Seventh Annual Rice Quantum Institute Research Colloquium, Rice University, Houston, TX, August 1993.

"A Practical XUV Laser for Holography," J. F. Young, M. J. Byrd, and T. E. Sharp; IEEE Lasers and Electro-Optics Society Annual Meeting, Boston, MA, November 1992 (Invited).

"Measurement of XUV Absorption Depth in Thin Films," M. J. Byrd, T. E. Sharp, and J. F. Young; Sixth Annual Rice Quantum Institute Research Colloquium, Rice University, Houston, TX, August 1992. "A Practical XUV Laser for Applications Research," T. E. Sharp, M. J. Byrd, J. Wu, and J. F. Young; Sixth Annual Rice Quantum Institute Research Colloquium, Rice University, Houston, TX, August 1992.

"Wide-Band Source for Optical Communication," L. Nguyen and J. F. Young; Sixth Annual Rice Quantum Institute Research Colloquium, Rice University, Houston, TX, August 1992.

"An Extreme Ultraviolet Holographic Microscope," J. F. Young; Ontario Laser and Lightwave Research Centre Seminar, Univ. of Toronto, Canada, October 1991 (Invited Speaker).

"An Extreme Ultraviolet Holographic Microscope," J. F. Young; IEEE Lasers and Electro-Optics Society Distinguished Lecturer Series: Princeton NJ Section, May 1992; Santa Clara Valley CA Section, May 1992; Central Texas Section, April 1992; Columbus OH Section, April 1992; Los Alamos NM Section, April 1992; Atlanta GA Section, March 1992; Dallas TX Section, February 1992; Boston, MA Section, October 1991.

"A Practical XUV Laser for Applications," T. E. Sharp, M. J. Byrd, and J. F. Young; Conference on Lasers and Electro-Optics, Anaheim, CA, May 1992.

"A terawatt XeF(C → A) excimer laser system," T. Hofmann, T. E. Sharp, G. Szabó, F. K. Tittel, and P. J. Wisoff; Int. Laser Science Conf., Monterey, CA, September 1991 (Invited).

- "Ultra-high intensity visible excimer laser system," F. K. Tittel, T. Hofmann, T. E. Sharp, G. Szabó, and P. J. Wisoff; Laser '91, Munich, Germany, June 1991.
- "Femtosecond pulse amplification by a XeF($C \rightarrow A$) excimer system," T. Hofmann, T. E. Sharp, P. J. Wisoff, W. L. Wilson, Jr., F. K. Tittel, and G. Szabó; Conf. on Lasers and Electro-Optics, Baltimore, MD, May 1991.
- "Self-focussing effects in a high power, ultrashort pulse, XeF($C \rightarrow A$) excimer amplifier," T. E. Sharp, T. Hofmann, W. L. Wilson, Jr., F. K. Tittel, P. J. Wisoff, and G. Szabó; Conf. on Lasers and Electro-Optics, Baltimore, MD, May 1991.
- "An Extreme Ultraviolet Holographic Microscope," J. F. Young and T. E. Sharp; Ninth Annual Conf. on Biomedical Engineering Research, Houston, TX, February 1991.
- "Characterization of a subpicosecond XeF($C \rightarrow A$) excimer laser," T. Hofmann, T. E. Sharp, C. B. Dane, P. J. Wisoff, W. L. Wilson, Jr., F. K. Tittel, and G. Szabó; SPIE Conf. on Gas and Metal Vapor Lasers and Applications, Los Angeles, CA, January 1991 (Proceedings: SPIE Vol. 1412, pp. 84-90).
- "Advanced concepts of electron beam pumped excimer lasers," F. K. Tittel, P. Canarelli, C. B. Dane, T. Hon, R. Sauerbrey, T. E. Sharp, G. Szabó, W. L. Wilson, Jr., P. J. Wisoff, and S. Yamaguchi; Eighth Int. Symp. on Gas Flow and Chemical Lasers, Madrid, Spain, September 1990 (Proceedings: SPIE Vol. 1397, pp. 21-29).
- "Amplification of high intensity ultrashort blue-green laser pulses using a XeF($C \rightarrow A$) excimer amplifier," C. B. Dane, T. E. Sharp, T. Hofmann, W. L. Wilson, Jr., F. K. Tittel, P. J. Wisoff, and G. Szabó; IEEE Lasers and Electro-Optics Society Annual Meeting, Boston, MA, November 1990.
- "A tunable, high power, subpicosecond blue-green dye laser system with two stage amplifier design," T. E. Sharp, C. B. Dane, F. K. Tittel, P. J. Wisoff, and G. Szabó; Rice Quantum Institute Colloquium, Houston, TX, August 1990.

8. Copies of Recent Publications

1. "Optical Gain in the Ionic Excimer Cs^{2+}F^- Excited by Soft X-Rays from a Laser-Produced Plasma."
2. "A Reasonably Practical XUV Laser for Applications."

Optical gain in the ionic excimer Cs^{2+}F^- excited by soft x rays from a laser-produced plasma

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Received August 5, 1993

Optical gain was observed on the $B \rightarrow X$ transition of the Cs^{2+}F^- ionic excimer molecule at 185 nm. The excimer was produced by photoionization of CaF vapor with a laser-produced plasma source. An average net gain coefficient of 0.3 cm^{-1} was measured for 7 Torr of CaF and a 225-mJ pump laser.

We have observed gain in a new class of excimer molecule, the alkali halide ionic excimers. Soft x rays from a laser-produced plasma were used to photoionize CsF vapor and produce excited-state Cs^{2+}F^- , with an average gain coefficient of 0.3 cm^{-1} at 185 nm measured over a 6-cm length. To our knowledge this is the first demonstration of a new type of molecular ultraviolet laser with broad bandwidth and large saturation intensity.

The alkali halide ionic excimers are isoelectronic to the well-known rare-gas halides¹ and have emission wavelengths in the range from 210 to 130 nm.²⁻⁴ The spectroscopy and kinetics of alkali halide ionic excimers were studied in detail by use of electron-beam,⁵ ion-beam,^{2,6} and soft-x-ray^{3,4,7,8} excitation. A schematic potential energy curve for Cs^{2+}F^- is shown in Fig. 1. Direct photoionization of the CsF ground-state molecule by soft x rays from a laser-produced plasma leads to the efficient formation of the Cs^{2+}F^- (B) state,^{4,7} which decays under emission of 185-nm radiation to the repulsive Cs^{2+}F^- (X) state. The stimulated-emission cross section for this transition is estimated to be $\sigma = 2 \times 10^{-16} \text{ cm}^{-2}$.^{1,9} Unlike the rare-gas halides, however, the ionic excimer precursor molecule CsF is slightly absorbing at the 185-nm excimer emission wavelength, with an absorption cross section of $3 \times 10^{-18} \text{ cm}^{-2}$.¹⁰ The primary products of CsF photoionization, however, have no measurable absorption at 185 nm, so the loss can be effectively eliminated with sufficient pumping flux. Detailed kinetic studies⁹ predict a net gain of 0.2 cm^{-1} near a pumping source having a blackbody temperature of 20 eV for an initial CsF pressure of 3 Torr.

As discussed and demonstrated in Refs. 11-16, the oblique-incidence laser-produced plasma geometry has several advantages for efficient pumping of photoionization lasers. The spectral output corresponds approximately to a blackbody radiator with a temperature of 10-20 eV.^{7,11,12,17} Long high-aspect-ratio excitation regions can be produced by use of moderate-energy pump lasers, and the traveling-wave plasma results in nearly synchronous excitation, which is important for short-lifetime upper levels. Our experimental setup is shown in Fig. 2. The plasma pumping source is a Nd:YAG-based mode-locked oscillator/regenerative amplifier

system¹⁶ that produces 225-mJ 100-ps 1064-nm pulses at 6 Hz. The beam diameter was enlarged to 3 cm and focused by a cylindrical mirror onto a stainless-steel plasma target rod inside a CsF heat-pipe cell. The mirror had a 60-cm concave radius of curvature, was coated with Al, and was used at a 60-deg angle of incidence relative to the normal, resulting in a 60-cm sagittal focal length and a 6-cm-long line focus. The laser intensity at the target surface was $\sim 3 \times 10^{10} \text{ W cm}^{-2}$. The target surface was grooved, as previously described, to produce a distributed series of pointlike plasma sources,¹³⁻¹⁵ and it rotated continuously during the experiments.

The CsF vapor was produced in a heat pipe operating at temperatures as high as 850 °C. We estimate the CsF number density by measuring the vacuum ultraviolet absorption through the pumping region, using a laser-produced plasma point source at the end of the target rod. We compared the measured absorption in the region from 220 to 120 nm with previous data to determine the molecular density, assuming a uniform density along the length. The results were consistent ($\pm 20\%$) with the equilibrium vapor pressure corresponding to the cell temperature measured with external thermocouples.¹⁰ CsF vapor pressures in the range of 1-10 Torr were used. He was used as a buffer gas to maintain the heat-pipe

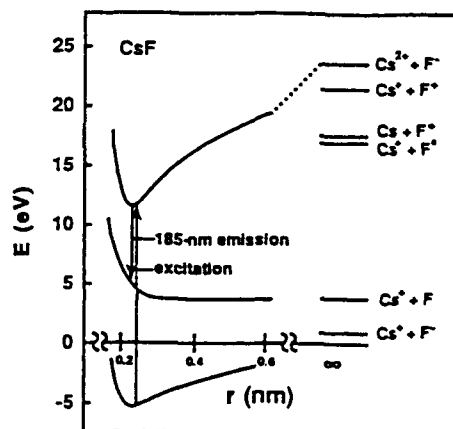


Fig. 1. Potential energy E versus internuclear separation r for the CsF ionic excimer.

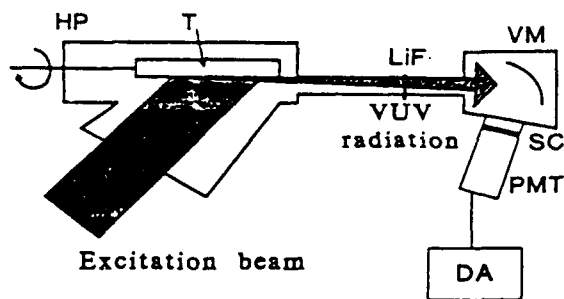


Fig. 2. Experimental setup for measuring VUV gain of CsF ionic excimer molecules. HP, heat-pipe vacuum cell; T, rotating target rod with grooved surface; LiF, lithium fluoride window; VM, VUV monochromator; SC, sodium salicylate scintillator; PMT, photomultiplier tube; DA, digital data-acquisition system.

operation and to reduce CsF deposition on the cell windows.

Radiation from the cell was analyzed by a 20-cm focal-length vacuum spectrometer and detected with a sodium salicylate scintillator and a fast photomultiplier. The spectrometer was located ~ 40 cm from the end of the plasma and isolated by a LiF window; the spectral resolution was ~ 0.5 nm. The photomultiplier signal was captured by a transient digitizer connected to a personal computer for digital data acquisition and data processing. The temporal resolution of the system was limited to ~ 10 ns by the scintillator fluorescence decay time. The signal was integrated during a 10-ns gate at the rising edge of the photomultiplier signal. The early positioning of the gate was critical for achievement of a high contrast ratio between the short-lifetime molecular radiation and the longer laser-produced plasma radiation. Figure 3 shows a portion of a typical spectrum displaying the background plasma emission, the $\text{Cs}^{2+}\text{F}^{-}$ ($B \rightarrow X$) emission band at 185 nm, and two peaks near 160 nm that are believed to be F_2 emission. The spectrum was taken with a continuous wavelength scan of 0.12 nm per shot, with an instrumental resolution of ~ 0.5 nm.

The determination of net gain in this system is complicated by the low predicted gain coefficient, the ground-state CsF loss, the background plasma radiation, and the nonuniform pumping distribution along the excitation length. The basic data consist of measurements of the emission strength at particular wavelengths as a function of excitation length. We varied the plasma length by moving a shutter across the pumping beam; in all cases the data were identical for shutter movement either toward or away from the spectrometer, indicating that background losses in shielded regions were not significant. Figure 4(a) is a plot of emission versus excitation length for two wavelengths: the peak of the $\text{Cs}^{2+}\text{F}^{-}$ band at 185 nm and a reference wavelength of 240 nm, chosen to be near the signals of interest, yet with no observed stimulated emission or absorption features. Each experimental point represents the average of 100 laser shots. The 185-nm excimer emission increases more rapidly than the plasma reference, indicating gain. Figure 4(b) shows the results when the spectrometer was moved off the axis of the linear plasma;

the excimer emission and plasma background show similar length dependence. We can make a crude estimate of the gain coefficient from Fig. 4(a) by fitting an exponential to the ratio of the 185- and 240-nm reference emission intensities. The result is $g = 0.36 \text{ cm}^{-1}$ for a CsF pressure of 7 Torr, but the uncertainty is large.

We improved our estimate of the gain coefficient by including the effects of inhomogeneous pumping. We measured the spatial distribution of excited $\text{Cs}^{2+}\text{F}^{-}$ molecules by scanning a 0.5-cm-wide aperture across the plasma pumping beam and measuring the 185-nm emission. The signal is proportional to the number density $n(z)$ of the excited excimer molecules along the laser focal line. The data, which are shown in Fig. 5, can be approximated by the parabolic dependence $n(z) = n_{\text{max}} f(z)$, where $f(z) = 4z(L - z)/L^2$, $0 \leq z \leq L$, L is the maximum excited length, and n_{max}

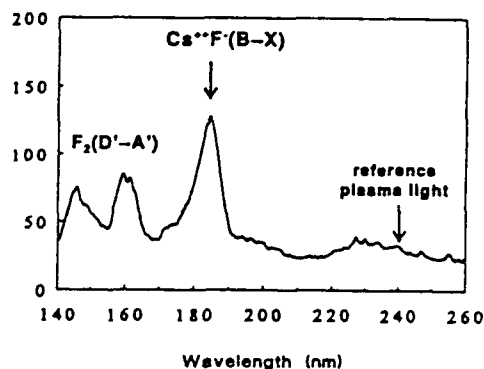


Fig. 3. Emission spectrum of soft-x-ray-excited CsF vapor at ~ 7 Torr. The arrows show the spectral regions used in the evaluation of the data.

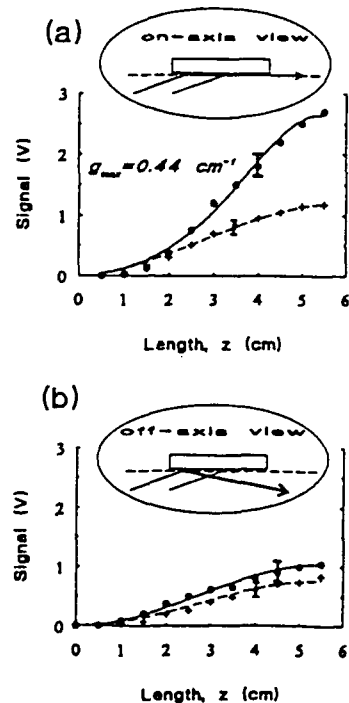


Fig. 4. Emission intensity as a function of plasma length at 185 nm (filled circles) and at 240 nm (pluses), for a CsF pressure of 7 Torr, viewed (a) on the plasma axis and (b) off axis.

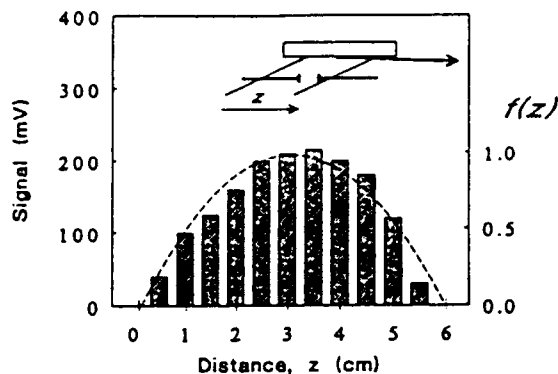


Fig. 5. Spatial distribution of 185-nm radiation along the excited region measured by scanning of a 0.5-cm aperture across the pumping beam. The dashed curve is a parabolic fit used to estimate the small-signal gain coefficients.

is the maximum excimer number density. Emission at the 240-nm reference wavelength showed the same dependence. Since the small-signal gain $g(z)$ is proportional to $n(z)$, the emission intensity, if we ignore constant atomic and geometrical factors, from an excited length z should be given by

$$I(z) \propto \exp\left[g_{\max} \int_0^z f(z') dz'\right] - 1. \quad (1)$$

An average gain coefficient for the total excited length L can be defined as

$$g_{\text{avg}} = \frac{g_{\max}}{L} \int_0^L f(z) dz = \frac{2}{3} g_{\max}. \quad (2)$$

We determined values of g_{\max} and g_{avg} by fitting relation (1) to the measured 185-nm emission signal. For a CsF pressure of 7 Torr, $g_{\max} = 0.44 \text{ cm}^{-1}$ and $g_{\text{avg}} = 0.30 \text{ cm}^{-1}$; at 4 Torr, $g_{\max} = 0.36 \text{ cm}^{-1}$ and $g_{\text{avg}} = 0.24 \text{ cm}^{-1}$. The solid curve through the 185-nm data of Fig. 4(a) is the best fit of relation (1). However, the emission intensity in the absence of gain, either at the reference wavelength or off axis at 185 nm, should be proportional to the simple integral $\int_0^z f(z') dz'$. This function is plotted through the 240-nm data in Fig. 4(a) and for both curves in Fig. 4(b). In all cases there is good agreement between the measured points and the calculated curves.

As Fig. 3 shows, we also observed emission peaks at 158 and 161 nm. These signals were not present at the start of the experiment but grew in intensity in proportion to the number of plasma excitation pulses. The emission intensity remained approximately constant as the cell was cooled, reducing the CsF density to near zero. We attribute these peaks to the accumulation of F_2 in the interaction chamber. F_2 can be formed both directly from CsF photodissociation products and by means of Cs^{2+}F^- (X) state dissociation following emission at 185 nm. The emission peak at 161 nm and the shift of the 158-nm peak from the well-known 157-nm F_2 laser

line¹⁸ can be attributed to vibrationally excited levels of the molecule in the high-temperature environment.

Molecular transitions typically have broader linewidths and lower stimulated-emission cross sections than their atomic counterparts. Although they require higher population inversions in order to reach the same gain, they also exhibit higher saturation intensities and output. The ionic excimers provide a welcome expansion of the available UV molecular laser lines. We have measured gain in Cs^{2+}F^- that is consistent with theoretical predictions^{1,9} and have demonstrated that substantial gain can be achieved with a moderately sized practical pumping system.

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A Reasonably Practical XUV Laser for Applications*

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Abstract

We describe a laboratory-scale, 109 nm, Xe Auger laser pumped by an all-commercial, high repetition rate, Nd:YAG laser system. The Xe laser provides pulse energies up to $1 \mu\text{J}$, and an average flux of 3×10^{12} photons per second at 11.5 eV. Measurements of the gain, output energy, and output beam shape are reported, along with an investigation of the time dependence of the gain.

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1 Introduction

There are many exciting applications of coherent extreme ultraviolet (XUV) sources in photochemistry, material science, atomic physics, and the life sciences, but to become practical, these applications require a convenient laser source that can be easily assembled and that can operate in the laboratory with a minimum of attention. We report a significant advance toward this goal: the first XUV laser system pumped by a standard, commercially available, Nd:YAG laser system at high repetition rate. Gain in the XUV has been observed in several atomic species, including C[1], Cs[2, 3], Zn[4], Kr, and Xe[5, 6]. We chose to develop the Xe^{+2} Auger laser at 109 nm because of its unique combination of high gain, efficiency, benign physical properties, and short wavelength. In this paper we describe the construction of this source and present data on gain, output energy, and beamshape; initial investigations of the time dependence of gain are also reported.

The 109 nm Xe laser is pumped by soft x-rays from a laser-produced plasma through a process of inner shell photoionization and subsequent rapid, selective Auger decay to the upper laser level. Gains of about $\exp(7)$ were first observed in the Xe^{+2} Auger system using a 55 J 1064 nm pump laser[5]. Subsequent optimization of operating parameters[7], and the development of new pumping geometries[8] resulted in an equivalent small signal gain of $\exp(33)$, and a saturated output energy of one microjoule using a 0.5 J pump laser[9]. Our present system achieves gains of about $\exp(25)$ and output energies of $1 \mu\text{J}$ using a 250 mJ, ~ 100 psec pump pulse, the lowest pump energy reported for saturated output to date. This is also the highest repetition rate Xe Auger laser to date; it provides an average flux of 3×10^{12} photons per second at 11.5 eV.

2 System Description

A schematic of the system is shown in Fig. 1. A Coherent Antares 76-s Nd:YAG cw mode-locked oscillator provides a 76 MHz train of ~ 100 psec long pulses at 1064 nm. A Continuum RGA-69 pulsed amplifier system is used to amplify selected pulses to an energy of about 250 mJ. The amplifier system consists of a regenerative amplifier stage followed by a single pass, 9 mm diameter Nd:YAG amplifier, all in a common housing. The amplifier system normally operates at a repetition rate of 10 Hz, but we have reduced the rate to 6 Hz because of the synchronization requirements of another experiment that also uses this amplifier. The entire pumping system occupies less than 2.5 m^2 of optical table space.

The 1064 nm pump beam is expanded to a diameter of about 2.5 cm and brought to a line focus on the plasma target using a cylindrical mirror at an ~ 83 deg angle of incidence. The several advantages of this grazing incidence geometry have been presented previously[8, 9]. The mirror, fabricated from BK7 glass, is 27.9 cm long by 7.6 cm high with a radius of curvature (concave) of 15.5 cm and is coated with aluminum. Used at this steep angle, the mirror produces a line focus about 20 cm long, parallel to the mirror, about 127 cm away, the sagittal focal length. A cylindrical mirror, rather than a lens, was first used to pump the H_2

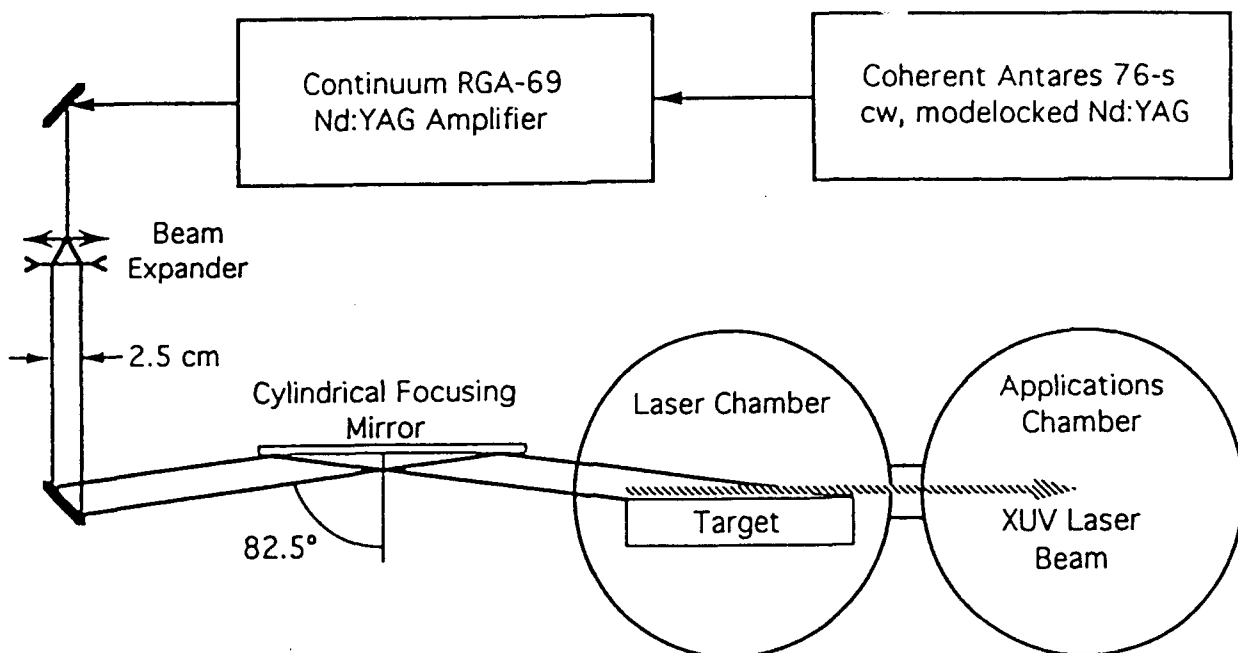


Figure 1: Schematic of the Xe Auger 109 nm laser system.

laser[10], and was later used for a Xe laser[11]. The mirror has lower aberrations than a lens at this angle, along with greater throughput, since the lens requires an antireflection coating that is difficult to design for this high angle of incidence. The lens coatings used in the past were absorbing, prone to damage, and very sensitive to polarization, wavelength, and angle of incidence. Current aluminium coatings can withstand fluences of about 50 mJ cm^{-2} , and because of the large projected beam area on the mirror at this angle, we are about 10 times below that value. Nevertheless, contamination or dust can lead to damage, and it is important to protect the surface; we enclose the mirror in a plastic box with openings for the beams. A multilayer dielectric coating on the mirror would be more robust than aluminum, but it would also suffer from polarization, wavelength, and angle sensitivity.

On the negative side, a mirror is somewhat more difficult to align initially than a lens, and it is important to use a mount that provides adjustments on all axes. In particular, the mirror sagittal focal length is a very sensitive function of the angle of incidence at this oblique angle, so it is necessary to have fine control of this angle, both by adjusting the mirror and by steering the input pump beam. Since it is difficult to make fine adjustments of the target position, the cylindrical mirror is first adjusted so that the focal line is parallel to the target surface, and then the input pump beam angle is adjusted, changing the sagittal focal length, to place the focal line on the target surface. Once the system is aligned, very little adjustment is needed.

The plasma target is a stainless steel rod 3.8 cm in diameter, 20 cm long, threaded at ~ 30

grooves per cm, rotated by a stepper motor. Burn patterns taken with minimum energy on thin metal shim stock formed to the the target surface indicate that the mirror produces a line focus about $75\ \mu\text{m}$ wide. Because of the grooved target surface, the pumping beam forms a series of point plasmas uniformly spaced along a 20 cm length[9]. The effective total plasma length is just 2.5 cm, the diameter of the pump beam, yielding a power density on target of about $10^{11}\ \text{Wcm}^{-2}$. The plasma target is located in a commercial circular vacuum feedthrough collar, 15 cm high and 28 cm inside diameter, with four ports. The pump beam enters through one port, and the XUV beam exits the opposite port into a similiar chamber used for applications and diagnostics. We have found it very useful to include a He-Ne laser alignment beam propagating parallel to, and just above the surface of the target, toward the application chamber. When aligned colinear to the plasma, and thus also to the XUV beam, it provides a convenient visible reference for locating detectors, etc.

While it is convenient to fill both the laser and application chambers with Xe, we found that Xe has a small absorption coefficient at 109 nm, corresponding to a cross section of $2 \times 10^{-20}\ \text{cm}^{-2}$. The cause of this absorption is unknown, but there are several neutral Xe levels near 109 nm, the closest ($300\ \text{cm}^{-1}$) being $5p^5({}^2P_{3/2})7d(J = 1)$ [12]. The absorption results in significant losses for our typical pressures of ~ 15 Torr and > 50 cm path lengths, and it is usually advantageous to isolate the laser chamber from the rest of the system with a LiF window. Although the absorption loss of even a 1 mm thick LiF window is also significant (85%), at least it is constant and does not distort measurements, e.g., determination of gain versus pressure. It should be possible to use specially fabricated thinner windows having lower loss because of the small area and low pressure differential. The window is mounted at a small angle, and the reflection from the surface is used to monitor the laser energy on each pulse. The pump focusing optics, XUV laser chamber, and the applications chamber also occupy less than $2.5\ \text{m}^2$ of optical table space.

3 Source Characterization

We have characterized the operational parameters of this source. Figure 2 shows the 109 nm output energy from Xe as a function of the pumped gain length. The measurement was made at a Xe pressure of 14 torr, the pressure that maximized the output energy for our pumping conditions. The output curve was measured using a scintillator-photomultiplier combination and adjusting the gain to avoid saturation. Each point represents the average of 40 laser shots; the maximum variation in these points was about $\pm 10\%$ at low signal levels. The peak output energy was measured using a windowless vacuum photodiode, and calibrated using the known quantum yield of the Al photocathode. The uncertainty of the peak energy is estimated to be $\pm 25\%$. The linear region of the semilog plot indicates an exponential gain coefficient of about $1.2\ \text{cm}^{-1}$, for a total equivalent small signal gain of $\exp(25)$; the output clearly reaches saturated energy densities at the end of the target. The output energy in the forward direction is about $1\ \mu\text{J}$, most of which is extracted from the relatively short saturated region. Because of the traveling wave excitation produced by this pumping geometry, we expect that very little energy is emitted in the backward direction, based on previous observations[8].

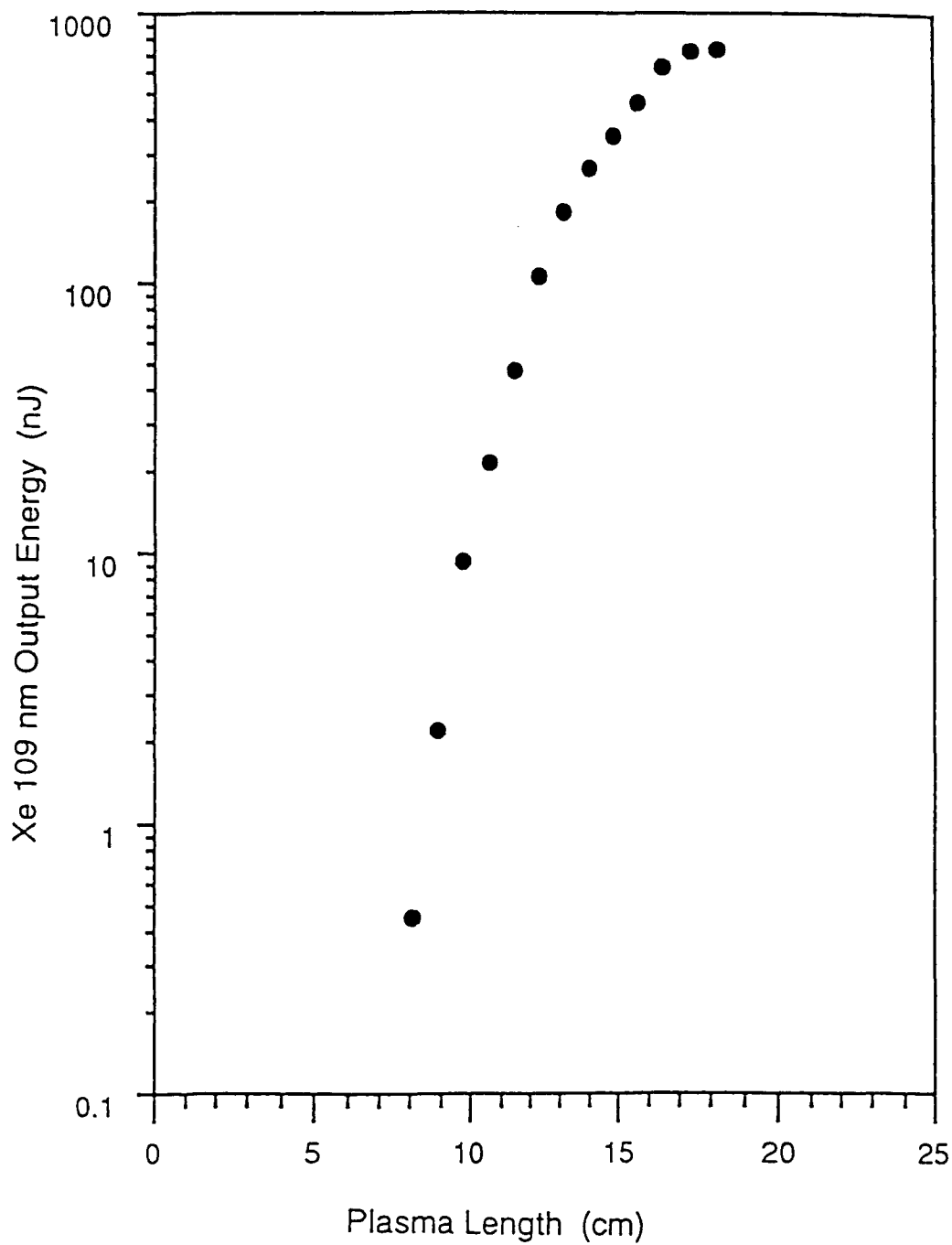


Figure 2: Output energy versus pumped length for the 109 nm Xe Auger laser, for a Xe pressure of 14 Torr.

We measured the spatial distribution of 109 nm energy in the laser output beam using a scanning knife edge technique, and used the data to calculate beam divergence and effective emitting area. The data (filled circles) in Fig. 3 represent the energy transmitted past a scanning knife edge, and detected by a vacuum photodiode. The knife edge was much larger than the beam size, and was located 80 cm from the front, pump-input end, of the target rod. Figure 3(a) shows the data for a horizontal scan, which is the direction normal to the target surface, and Fig. 3(b) shows data for a vertical scan, tangent to the target rod. In both cases, the abscissa gives the relative distance that the knife edge was moved, and the dashed lines represent a simple polynomial fit to the data.

Although the detailed shape of the transmitted energy curves for the normal and tangential scans are quite different, in both cases the data indicate that 90% of the energy was transmitted through a 1.8 mm wide region. Thus, the output beam divergence in both planes was about 2.3 mrad, for a total solid angle of $16 \mu\text{sr}$. At the output end of the pumped region, this solid angle corresponds to an effective emitting area of $8.1 \times 10^{-3} \text{ cm}^2$, or a circle of 0.9 mm diameter. This result is consistent with the $\sim 1 \text{ mm}$ absorption depth of the pumping soft x-rays from the laser-produced plasma into the surrounding Xe gas.

The lifetime of the Xe Auger 109 nm upper laser level has been measured to be 4.75 nsec, with a collisional quenching rate of $2.8 \times 10^7 (\text{Torr} \cdot \text{sec})^{-1}$ [13, 14, 6]. This means that the lifetime should be longer than a nanosecond up to pressures of 28 Torr, and that relatively long pumping pulse lengths should be effective in creating an inversion. (The lower laser level lifetime is calculated to be 8 to 10 nsec, meaning the laser is self-terminating on our timescale.) Subsequent measurements of the time-integrated gain coefficient for various pumping pulse lengths, however, showed that the effective gain coefficient decreased for pumping pulse lengths longer than a few hundred picoseconds, even for pressures of about 5 Torr[7]. This suggests that other processes, probably electron collisions, reduce the effective inversion time under the experimental conditions normally used for lasers. To date, however, neither the exact nature of the competing process nor the time dependence of the gain have been determined. We investigated the time dependence of gain by dividing the pumping plasma into two regions: a 12 cm length to provide a probe beam, and a 3 cm long gain section to be measured. Figure 4 shows the prism beam delay arrangement that provided a variable time delay between the pumping of the gain region and the generation of the probe beam.

Figure 5 shows the amplified probe output as a function of probe delay for two pressures. Each point on the graph represents the average of 20 measurements, yet the variability is still significant at low pressures where the signal levels are low, about $\pm 20\%$. Nevertheless, it is clear that the gain lifetime decreases with pressure, and is much shorter than predicted by collisional quenching alone. Quantitative conclusions, however, are difficult because the measured curves are convolutions of the gain decay function with the probe pulse shape. Sher measured the output pulse shape of a Xe system pumped with a 300 psec long pulse, both for unsaturated conditions—a total gain of $\exp(13)$ —and for saturated conditions, a gain of $\exp(40)$ [15]. The two cases were quite different; the unsaturated output pulse was symmetric in time with a width of 120 psec, while the saturated pulse was highly asymmetric and shortened to 50 psec. For all of our measurements, the probe region operated in the unsaturated regime, since the maximum gain-length product under optimum conditions (15 Torr) was

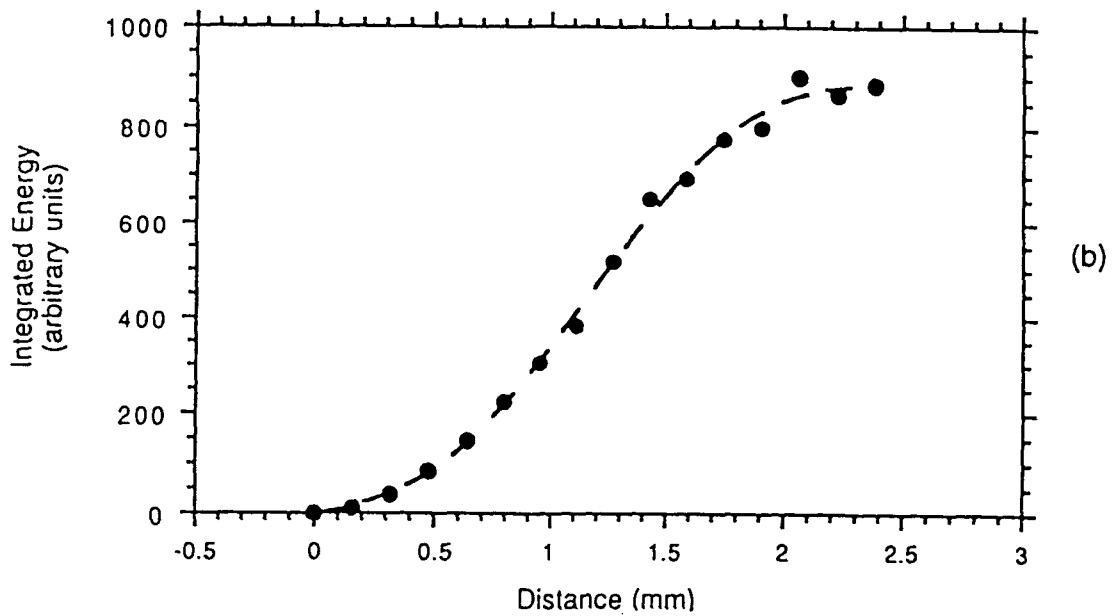
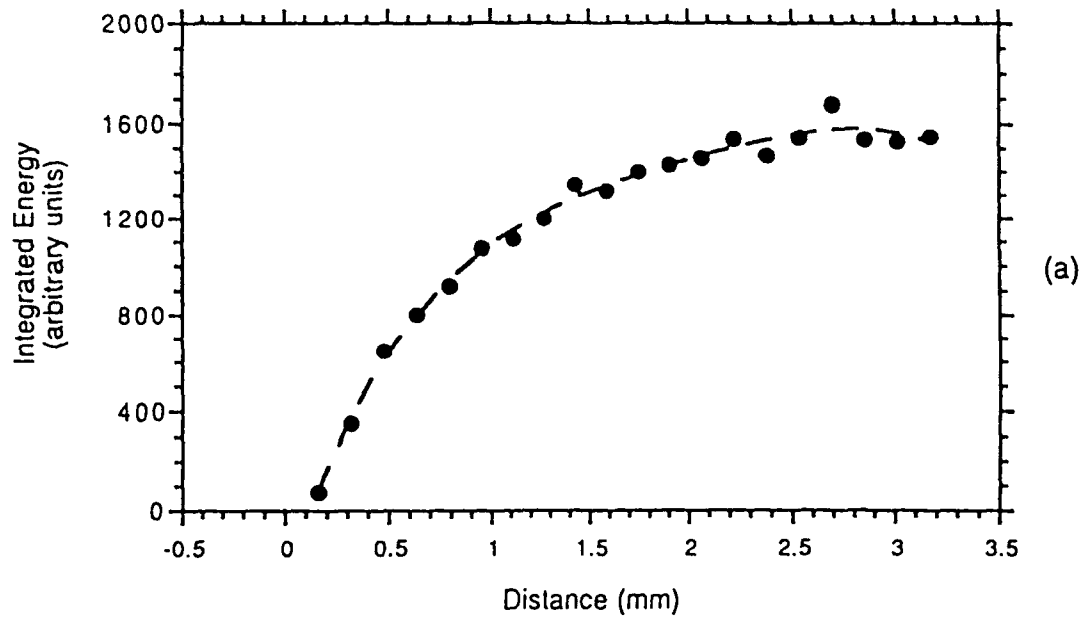


Figure 3: Laser beam energy transmitted past a scanning knife edge, for a scan (a) normal to the target surface (horizontal), and (b) tangential to the target surface (vertical). The knife edge was 80 cm from the front of the pumped region; the dashed curves are polynomial fits to the data.

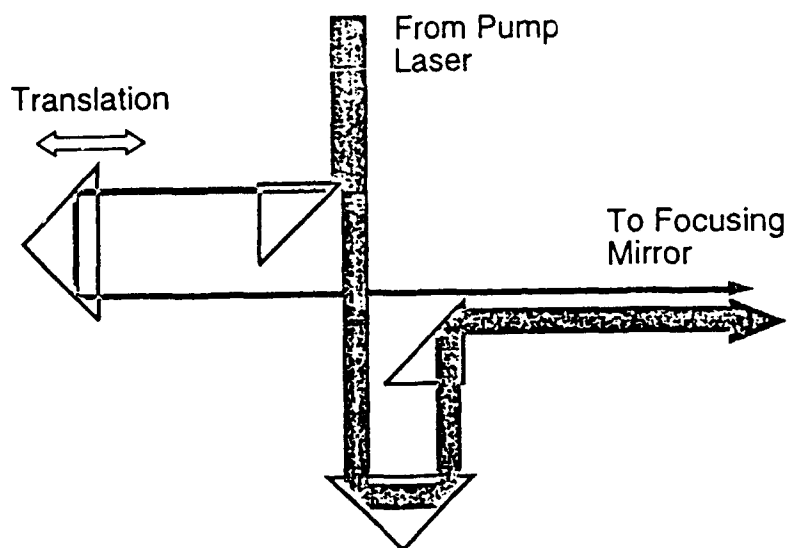


Figure 4: Schematic of the prism assembly used to spatially divide the pump beam, delay the probe portion, and recombine the two parts for measurements of the time dependence of gain.

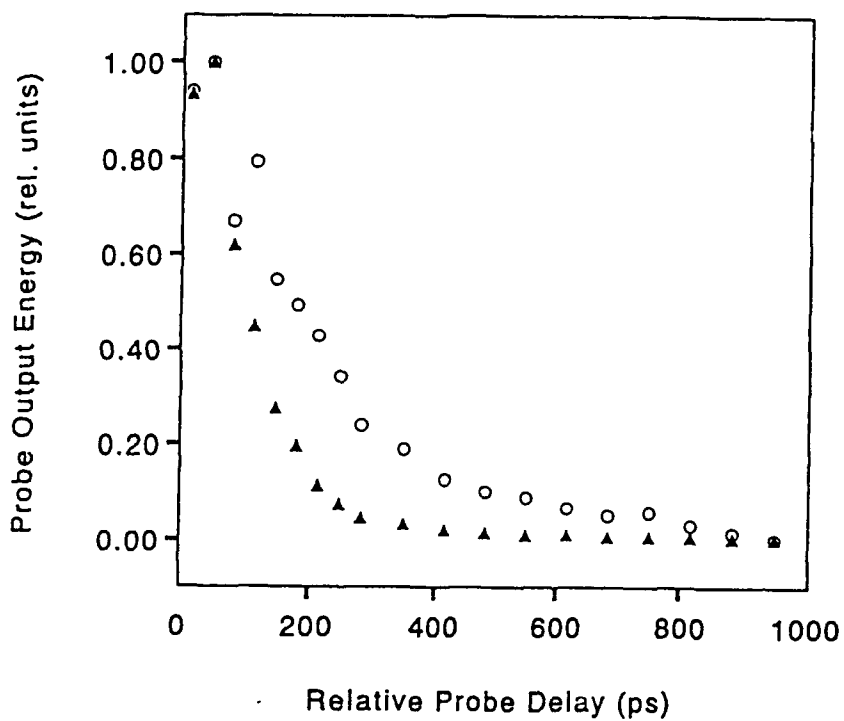


Figure 5: Amplified 109nm probe output as a function of probe delay for Xe pressures of 5 (open circles) and 34 Torr (filled triangles).

exp(14). Based on Sher's results, we expect the probe output to be symmetrical in time and somewhat shorter than the pump pulse length.

To analyze the data of Fig. 5, we assumed the gain decayed exponentially in time, $\alpha(t) = \alpha_0 \exp(-t/\tau)$, and fit the measured curves using two assumed probe pulse widths. The open circles in Fig. 6 show the gain time decay constant τ as a function of pressure assuming a very short probe pulse length, which effectively ignores the effect of probe pulse shape. The closed circles show the best fit time constants assuming a gaussian shaped probe pulse with a full width at half maximum of 80 psec, somewhat less than our pump pulse length. Both assumptions yield results consistent with previous observations: that sub-nanosecond pump pulses are required for significant gain, and that higher pressures require shorter pulses. But the most conspicuous feature of the data is the rather small variation of decay constant with pressure in this range, especially considering the uncertainty in the data and the fitting technique. This indicates that the most interesting pressure region for study is probably below 5 Torr. Unfortunately, we were not able to make measurements at lower pressures because of low probe signal levels.

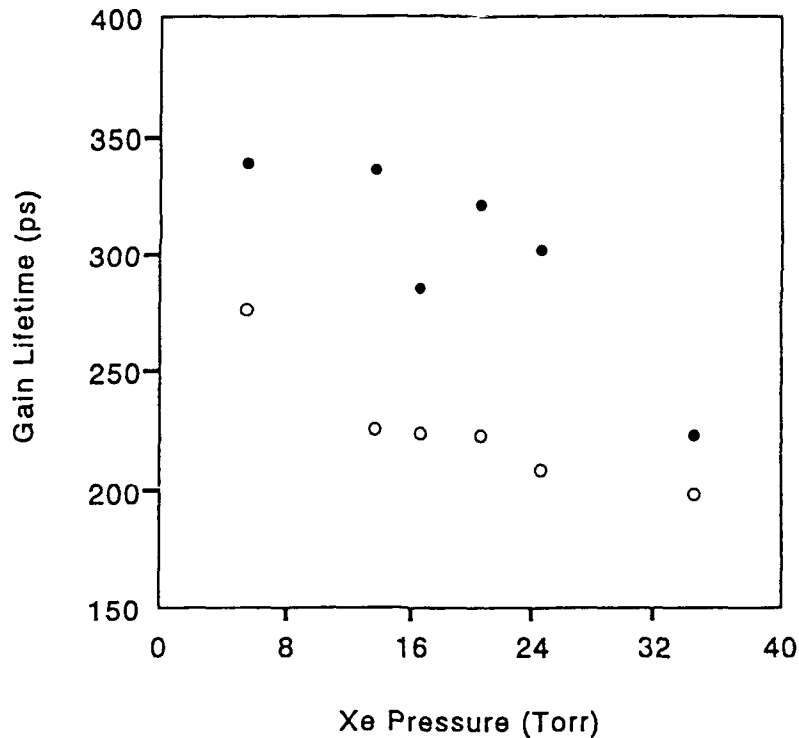


Figure 6: Time decay constant of gain at 109 nm as a function of pressure assuming: a very short probe pulse (open circles), and a gaussian shaped probe pulse with a full width at half maximum of 80 psec (filled circles).

4 Conclusions

The 109 nm Xe Auger laser is a promising practical source of XUV radiation, but improvements in output energy and beam quality are desirable. Because of the exponential nature of the system, small changes can yield large improvements. As can be seen from Fig. 2, most of the output energy is extracted from the last 5 cm of excitation length, where the 109 nm traveling wave reaches saturation energy density. Relatively modest increases in pumping energy or efficiency would reduce the target length needed to reach saturation and thus lead to significant increases in output energy. We used a plain stainless steel target for convenience, but there is evidence that a gold plasma would result in higher gains[8, 16]; for example, Ref. [8] reports gain coefficients about 4 times greater than we measured using only 2.3 times the energy density, at the same power density on target. In addition, using a fraction of the pump beam energy as a pre-pulse to initiate a plasma has produced output enhancements of ~ 10 under conditions similar to those used here[17]. The high optimum pressures we measured also indicate a low pumping flux relative to earlier work. The output of the pump laser system is constrained by a power density damage limit, so greater energy could be obtained from the same laser system by increasing the pulse length. Pulse lengths three or four times longer should not reduce pumping efficiency.

A better understanding of the gain dynamics might lead to several important improvements. A double-pass geometry would allow efficient energy extraction from the entire pumped volume, but our measurements indicate that the gain does not last long enough under our current conditions for this to be effective. Similarly, if excitation conditions could be found that would permit pumping with a Q-switched laser, considerable simplification of the pumping laser system would be possible. The method we used to measure the gain time dependence had the advantage of simplicity, but it provided only limited information. We need to isolate the probe and gain regions so that we can control their parameters independently and explore lower pressure and excitation conditions.

5 Acknowledgement

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