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LA-UR--88-2745

TI88 016236

TITLE OVERVIEW OF RECENT ADVANCES IN EXCIMER LASER
TECHNOLOGY AT LOS ALAMOS

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SUBMITTED TO International Congress on Optical Science
and Engineering "Extreme Laser Radiation"
Hamburg, West Germany
Sept. 22-23, 1988

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MASTER

Overview of Recent Advances in Excimer Laser Technology at Los Alamos

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ABSTRACT

From among the areas of excimer laser development at Los Alamos two are selected for further discussion: ultra-high brightness excimer laser systems and discharge-pumped XeF(C $\rightarrow$ A) lasers operating in the blue-green portion of the spectrum. Two different high brightness systems are described. One is based on small-aperture KrF amplifiers, while the other is based on a large-aperture XeCl amplifier. The XeF(C $\rightarrow$ A) laser is tunable from 435 to 525 nm, and may one day become a viable alternative to pulsed dye lasers for many applications.

1. INTRODUCTION

Recent advances in excimer laser technology at Los Alamos National Laboratory have occurred along a number of different fronts. The incorporation of a phase conjugating mirror (based on stimulated Brillouin scattering in a gas cell) in a KrF oscillator/amplifier system (e.g., a Lambda Physik model EMG 150) has resulted in improved beam quality and pointing stability while providing simple control of pulse durations with good energy extraction and excellent pulse/ASE contrast ratios¹. This and other efforts in the development of the 10-kJ Aurora KrF Laser Fusion Facility continue, with significant multiplexed beam-on-target demonstrations expected during the coming year. Separately², studies of active mode locking of a discharge-pumped XeCl laser have resulted in pulse durations as short as 120 ps.

We will focus, however, on two other areas of activity at Los Alamos: the "Bright Source" laser systems and work that is directed at the goal of efficient discharge-pumped XeF lasers operating on the C-to-A transition, which provides emission over the entire blue-green spectrum.

2. THE "BRIGHT SOURCE" LASER SYSTEMS

Subpicosecond, high brightness excimer laser systems are being used to explore the interaction of intense coherent ultraviolet radiation with matter. Applications of current systems include generation of picosecond x-ray pulses, investigation of possible x-ray laser pumping schemes, studies of multiphoton phenomena in atomic species, and time-resolved photochemistry. The first of these systems, Los Alamos Bright Source I (LABS I), is based on the amplification of subpicosecond pulses in small aperture (~ 1 cm²) KrF amplifiers^{3,4}, delivering focal spot intensities of $>10^{17}$ W/cm². Scaling to higher intensities, however, will require an additional large aperture amplifier which preserves near diffraction-limited beam quality and subpicosecond pulse duration⁵. We describe here both the small aperture KrF system, LABS I, which routinely provides pulses to several experiments, and a large aperture XeCl system, LABS II, designed to deliver ~ 1 J in subpicosecond pulses and yield intensities on target in excess of 10^{20} W/cm². We also discuss the effects of two-photon absorption on the optical components of large aperture, high brightness excimer lasers.

LABS I consists of a "front-end", which generates 248 nm seed pulses, followed by two KrF amplifiers. The seed pulses are initially generated at 648 nm with a mode-locked dye laser that uses DCM as the gain dye and DTDCl as the saturable absorber dye. The dye laser is synchronously pumped with the frequency doubled output of a cw mode-locked Nd:YAG laser. The dye pulses are then amplified at a 3 Hz repetition rate and frequency doubled in a 2 mm thick BBO crystal. The resulting pulses at 324 nm are finally sum frequency mixed with amplified 1064 nm pulses in a second 2 mm BBO crystal to produce 5- to 10 pJ subpicosecond (~ 0.7 ps) seed pulses at 248 nm. These pulses are then amplified by two Lambda Physik EMG 200 Series KrF amplifiers, separated by a vacuum spatial filter to suppress ASE and improve beam quality. The output beam diameter is 17 mm and the final output energy at 248 nm is 25 ± 3 mJ with $\sim 5\%$ ASE. The pulsewidth, measured using two-photon ionization in NO, is 700 fs. The focused spot size achievable with this system has been determined indirectly by measuring the confocal parameter of a beam focused by f/3 optics. The inferred focal spot diameter is 3.6 μ m (approximately twice the diffraction limit), which implies an intensity at the focal plane of 3.5×10^{17} W/cm². For all experiments parabolic mirrors are used as the focusing optics to preserve pulsewidth, minimize aberrations, and avoid nonlinear absorption and refraction.

This system is routinely operated as a source for several physics experiments⁶. An x-ray spectroscopy experiment has shown that multiphoton processes in a solid aluminum target produce an aluminum ion plasma exhibiting line radiation at energies exceeding 2 keV. Another experiment has shown that highly charged ion

states can be produced by multiphoton ionization. In xenon, for example, absorption of 213 248-nm photons yields a Xe^{+1} ion.

We are currently building a second high-brightness system, based on amplification in XeCl , that uses a large aperture ($\sim 100 \text{ cm}^2$) final amplifier designed to deliver pulse energies approaching 1 J at a maximum repetition rate of 1 Hz. The seed pulse generator scheme is sketched in Figure 1. Pulses of 175 fs duration at 616 nm are initially generated in a linear-cavity, dispersion-compensated dye laser (Rhodamine 6G/DDOCl) that is synchronously pumped by a cw mode-locked Nd:YAG laser. The Nd:YAG laser also provides $1.06 \mu\text{m}$ seed pulses to a regenerative amplifier. This synchronous amplification scheme has the advantages of low amplified spontaneous emission, good beam quality, and the elimination of a separate pump laser for the dye amplifier. We observe no temporal broadening of our 175-fs pulses through this amplifier. The amplified 616-nm pulses are then frequency doubled to 308 nm in a BBO crystal. Preamplification of these 30- μJ uv pulses to the 3-mJ level is accomplished with a single small aperture commercial XeCl discharge amplifier. The beam is then expanded in a vacuum spatial filter before entering the final amplifier (Figure 2). This $10 \times 10 \text{ cm}^2$ aperture device consists of two independently pumped discharge gain regions that share a common x-ray preionizer. The small signal gain, g_0 , in each discharge region is ~ 5 . The discharges are pumped by low-jitter, thyatron-switched pulse modulators with three-stage magnetic pulse compression. The resultant jitter is less than 5 ns over a ~ 50 -ns gain time. In order to maintain near-diffraction-limited beam quality at a sustained 1-Hz repetition rate, a transverse gas flow system is used. The wave front distortion at 632.8 nm is less than $\lambda/20$ over 80% of the aperture and the hot gas clears within 30 ms after a shot. Barring additional distortion due to nonlinear refraction in the output window, focal-spot intensities $> 10^{20} \text{ W/cm}^2$ should be obtained with this system.

Figure 1. Subpicosecond, 308 nm seed pulse generator

Figure 2. Large-aperture XeCl amplifier

An understanding of nonlinear optical phenomena such as two-photon absorption and nonlinear refraction in ultraviolet window materials at excimer wavelengths is essential for the design of large aperture amplifiers. We have measured the two-photon absorption coefficient β at 248 nm for those materials (fused silica and CaF_2) that can be obtained in large apertures. For fused silica $\beta = 4.5 \pm 2.2 \times 10^{11} \text{ cm/W}$, while for CaF_2 , $\beta = 8.3 \pm 4.1 \times 10^{12} \text{ cm/W}$. To evaluate the implications of these values of β for KrF laser systems, we plot in Figure 3, $I_{\text{out}}/I_{\text{in}}$ versus I_{in} for 1.27 cm thick windows of fused silica and CaF_2 . Both two-photon absorption and its associate pulsewidth broadening are included in the calculation of I_{out} . At $I_{\text{in}} = 3 \times 10^{10} \text{ W/cm}^2$, a typical intensity for current systems, $I_{\text{out}}/I_{\text{in}} = 0.33$ for fused silica and $I_{\text{out}}/I_{\text{in}} = 0.66$ for CaF_2 . Therefore, for large-aperture, KrF-based, high brightness lasers, the material for the output window, as well as for any subsequent windows should be carefully chosen, and the total thickness after the gain medium should be minimized. In contrast, XeCl-based high brightness lasers, where the photon energy is 4 eV, become attractive, since two-photon absorption in CaF_2 is not possible.

Figure 3. $I_{\text{out}}/I_{\text{in}}$ versus I_{in} for 0.5 inch thick CaF_2 and fused silica samples

3. DISCHARGE-PUMPED XeF(C→A) LASERS

Our studies in discharge-pumping of the XeF(C→A) transition are motivated by the recent work of Tittel, Sauerbrey, Nighan and co-workers^{7,8} using intense short-pulse (10 ns) electron-beam excitation at pump energies of 135 Joules per liter, which has demonstrated efficiencies of ~3% and intrinsic energy extraction as high as 4 joules per liter. These results are obtained with complicated five-component gas mixtures that include argon as the buffer with xenon, krypton, NF₃ and F₂. The most remarkable change from more conventional mixes appears to be the addition of substantial amounts of krypton. It is believed⁸ that this lowers the absorption in the blue-green region by argon species [Ar₂(³Σ_u⁺) and Ar₃⁺], reduces the population of the B-state due to enhanced B-C state mixing by the krypton, and increases absorption at the B-X transition wavelength due to the formation of Kr₂F (thus impeding ASE buildup of the B-X emission). In addition, there is increased gain on the blue side of the C-A band due to emission from the Kr₂F trimer. Under e beam excitation the B-X fluorescence steadily decreases with increasing krypton pressure. The C-A fluorescence remains relatively constant since, as kinetic-code calculations show, at room temperature over ninety percent of the population starts off in the C-state. Figure 4 shows a typical fluorescence spectrum from such an optimized mixture. Time-resolved gain measurements show that during the e-beam excitation pulse there is net absorption, with net gain taking place only during the afterglow. Recently, however, results at Avco Everett Research Laboratory⁹ have shown that at lower energy deposition rates substantial gain is observed **during** a longer e-beam pump pulse (using such five component gas mixtures as described by the Rice University and United Technology work). We presume that this occurs, after an initial transient period, because of saturation of the absorbing species.

Figure 4 Time integrated fluorescence spectrum from an e-beam pumped 5 component gas mixture. The dashed lines represent the estimated contributions from the XeF(C→A) transition and from the Kr₂F trimer.

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Discharge pumped laser emission on the C-to-A transition has been previously demonstrated¹⁰⁻¹³, but always at efficiencies much lower than that of the B-X transition. The results of pumping with intense electron beams tend to indicate that to successfully implement similar high efficiency results in a discharge pumped device it is necessary to deposit power densities of the order of 10 MW/cm³ for ~10 nanoseconds, and this must be accomplished in gas mixes containing high concentrations of krypton at static filling pressures ~4 atmospheres. The question of the best buffer gas is a point of concern, because if argon is required kinetically for good discharge excited laser efficiency then the task becomes significantly more difficult. To address these issues we are conducting fluorescence studies on the XeF(C→A) transition in a number of different discharge devices. These include a long pulse inductively stabilized laser head at relatively low power deposition (560 kw/cm³), a commercial Lambda Physik EMG 50 laser at a power deposition rate estimated at 1 to 2 MW/cm³, and a high power deposition device with deposition densities of 20 MW/cm³.

Vacuum ultraviolet flash optical pumping of the C→A transition has also been demonstrated¹⁴, and we are also planning to study that avenue, mainly for the sake of a better understanding of the kinetics.

Measurements are ongoing at the time of this writing, but some conclusions can be drawn at this time regarding optimization of the kinetics for discharge pumping:

- The addition of large quantities of krypton is also important in discharge-pumped devices.
- NF_3 or a mixture of NF_3 and F_2 produces stronger fluorescence (and presumably higher gain) than just F_2 as the fluorine donor.
- A helium buffer yields results that are indicative of B/C state distributions that are similar to those observed with e-beam pumping, whereas with a neon buffer the population seems more evenly distributed between the two states.
- Initial indications are that the use of argon as the main buffer constituent may actually be important to the kinetics in discharge pumping, whereas it was thought that the role of argon in an electron-beam device was simply that of beam-stopping.

The electrical requirements of a discharge device would appear to be quite demanding based on the things we have learned thus far. No currently available commercial device will produce a stable glow discharge in 4 atmospheres of argon with 10 torr of NF_3/F_2 and 400-500 torr of krypton, while depositing $>10 \text{ MW/cm}^2$. Our efforts will next be guided towards the development of an x-ray preionized, high-power-deposition, short-pulse device capable of establishing an arc-free discharge in the hostile environment described above. One important feature of the discharge circuit, for the sake of efficiency, will probably be a current/voltage turn-off time that is as short as the rise time.

4. ACKNOWLEDGEMENTS

We wish to acknowledge the skilled assistance of Michael Maloney, Tatsuhiko Sakai, Jeffrey Roberts and Charles Lester. Also, we appreciate the discussions with and the advice received from Prof. Frank Tittel and Dr. Roland Sauerbrey. This research is supported by the Internal Research and Development program at the Los Alamos National Laboratory.

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