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Recent advances in excimer laser technology at Los Alamos

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ABSTRACT

Current research in excimer laser technology at Los Alamos progresses in two major areas: 1) In the Bright Source program, the development of ultra-high brightness (sub-picosecond) laser systems, based on discharge-pumped excimer laser amplifiers, continues. Recently we have completed rigorous measurements of the saturation parameter for ultra-short pulses. 2) In the laser fusion program, implementation of the large KrF laser fusion amplifiers have been accompanied by numerous studies of the laser physics and kinetics of large e-beam pumped devices.

1. ULTRASHORT-PULSE ENERGY EXTRACTION MEASUREMENTS IN XeCl AMPLIFIERS

The bandwidth of the XeCl gain medium is 120 cm^{-1} and can be fully exploited to produce femtosecond ultraviolet pulses by seeding of a XeCl amplifier with the frequency-doubled output of a standard ultrafast dye oscillator/amplifier. Such ultrafast XeCl laser sources[1-4] have in recent years delivered peak optical powers of up to one terawatt. However, as pulse durations drop below the characteristic recovery times of the gain medium, and otherwise begin to approach the bandwidth limit, output energies and pulse shapes can be expected to depart from those derived from well-established incoherent pulse-propagation models such as the Frantz-Nodvik theory.[5] The intelligent design and application of ultrafast XeCl amplifiers therefore strongly depends upon new measurements of the time- and frequency-dependent properties of XeCl amplifiers in the short-pulse regime.

Short-pulse amplification in XeCl was first studied by Corkum and Taylor.[6] With 2-ps pulses, they measured a saturation energy, E_{sat}, of 1.0 mJ/cm^2 and found that following transit of the ultrashort pulse, the time dependence of the gain recovery consisted of two components: a fast 40-ps recovery that restored 59% of the original amplitude, and a slow 2.5-ns recovery that restored the remaining 41% of the amplitude. The short recovery time was identified with a combination of population redistribution within the rotational manifolds of both the upper and lower laser levels, vibrational relaxation in the ground state, and possibly dissociation of the ground state, while the long recovery time was identified with the energy storage time of the gain medium. From these data Corkum and Taylor predicted that for pulse durations τ significantly longer than the short recovery time, but shorter than the energy storage time (say 200 ps $< \tau <$ 1 ns), E_{sat} would increase to 2.5 mJ/cm^2. More significantly, however, they predicted that for subpicosecond pulses with spectral bandwidths approaching the XeCl limit of 120 cm^{-1}, an effective E_{sat} of 2.5 mJ/cm^2 would also be observed, since such pulses would then have full access to the rotational manifolds of the upper and lower levels.

Unfortunately, previous experimental efforts to determine the effective E_{sat} in XeCl for near-bandwidth-limited pulses appear to be inconclusive. While some workers[1-3] report $E_{sat} \sim$ 2 mJ/cm^2, others[7] report E_{sat} = 0.85 mJ/cm^2, a value comparable to the 1.0 mJ/cm^2 result of Corkum and Taylor for 2-ps pulses. The importance of a reliable value for E_{sat} and the apparent disagreement among previously published measurements and theoretical predictions have motivated our own study of short-pulse amplification in XeCl. In order to provide as reliable and comprehensive results as possible, we have made the first contiguous measurements of short-pulse gain in all three of the pulsewidth regimes identified by Corkum and Taylor.

In our energy extraction measurements, the input pulse is split into two beams of equal energy. One beam is directed onto a pyroelectric calorimeter (Gentec ED-100) to determine the input pulse energy, E_{in}, while the second beam propagates through the center of the test amplifier's discharge volume and is then detected with an identical calorimeter to determine the output pulse energy E_{out}. The spatial extent of the beam is defined by 3-mm apertures placed before and after the amplifier. The input pulse energy is varied over four orders of magnitude using calibrated neutral-density filters.

These measurements are made with pulses of 160 fs, 3 ps and 600 ps duration. The 160-fs and 3-ps input pulses are generated using a frequency-doubled ultrafast dye laser and amplifier. (The spectral bandwidths of these pulses are 105 and 8 cm^{-1}, respectively.) The 600-ps input pulse is obtained using near-threshold stimulated Brillouin scattering in ethylene glycol to shape the output of a commercial XeCl oscillator.[9] Further, a variety of experimental conditions are employed in order to replicate those used by previous workers, including 1) the use of two different discharge amplifiers: a Lambda Physik MSC 101 with a gain length of 45 cm and a Lambda Physik MSC 201 with a gain length of 87 cm, 2) the use of gas mixtures with either helium or neon buffers as well as varying halogen content, 3) input pulses generated with and without pre-shaping by a XeCl preamplifier.

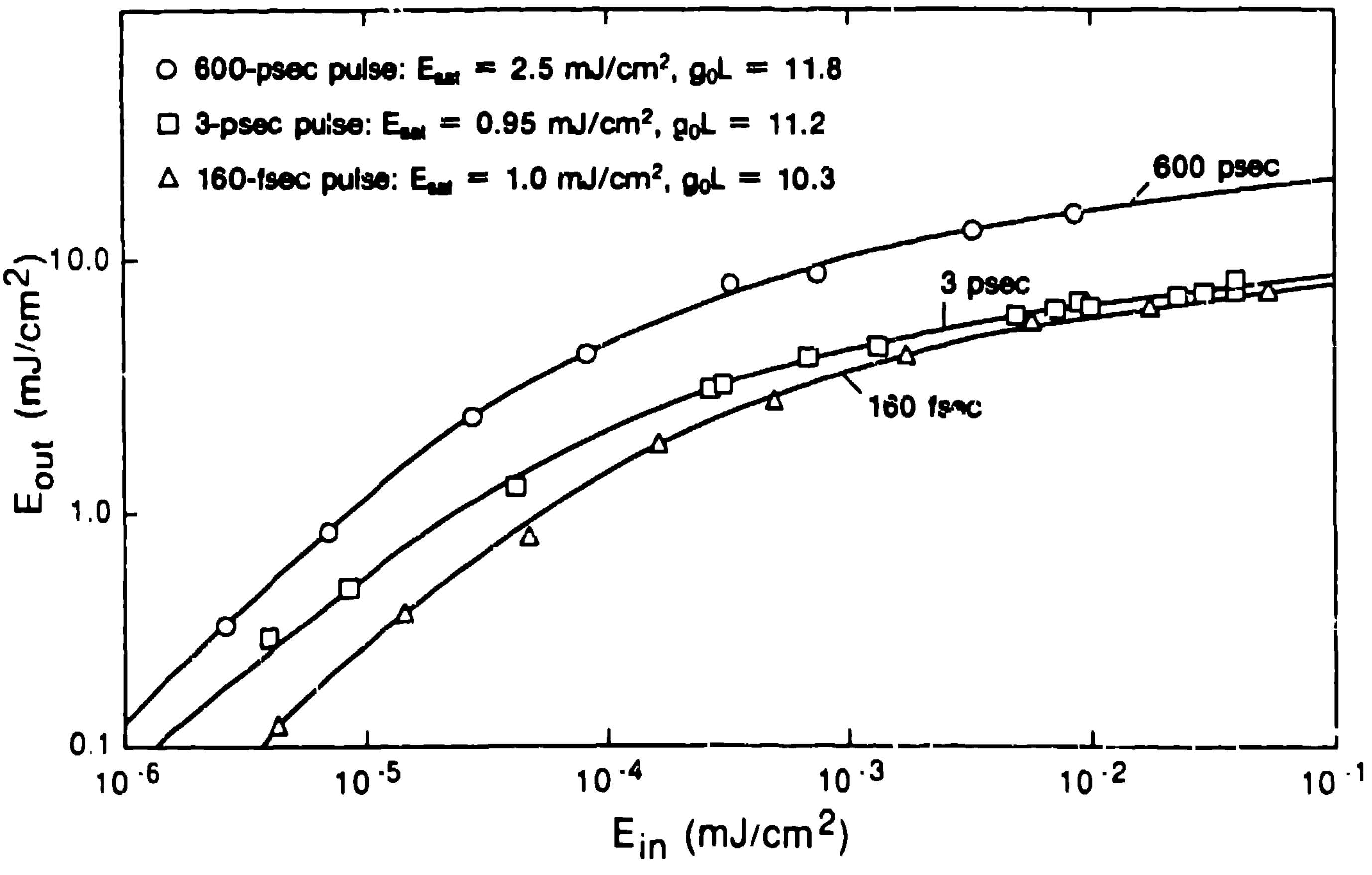

Fig. 1. Energy-extraction curves for various pulsewidths measured using a Lambda Physik 201 amplifier with a gas mixture of 60 T Xe, 4 T HCl and 2740 T Ne. The solid lines are fits of the data to the Frantz-Nodvik equation with the derived values of E_{sat} and g_0L listed in the inset.

In Figure 1 we present for the three different pulse widths typical data sets taken with a Lambda Physik 201 amplifier using a neon-based gas mixture and unshaped input pulses. The solid lines in Figure 1 represent fits to the data of the Frantz-Nodvik equation[5]

$$E_{out} = E_{sat} \ln\left[1 + e^{g_0 L}\left(e^{E_{in}/E_{sat}} - 1\right)\right] \quad (1)$$

where E_{sat} is the saturation energy and g_0L is the small signal-gain length of the amplifier. The values of E_{sat} and g_0L derived from the fits are shown in the Figure. Note that the small-signal gain, $G = \exp(g_0 L)$, increases significantly with pulsewidth. Further note that E_{sat} is about 1 mJ/cm^2 for both bandwidth-limited 160-fs pulses and 3-ps pulses, but increases to 2.5 mJ/cm^2 for 600-ps pulses. These results are found to be independent of the gain length of the test amplifier, the composition of the gas mixture, and of any pre-shaping of the input pulse introduced by a preamplifier. We conclude that over a very wide range of amplifier conditions the saturation energies given by fits to the Frantz-Nodvik curve (Eq. (1)) are $E_{sat} = 1.0 \pm 0.15$ mJ/cm^2 for 160-fs and 3-ps pulse durations and $E_{sat} = 2.5 \pm 0.30$ mJ/cm^2 for the 600-ps pulse duration.

Since in going from the intermediate-pulse to the long-pulse regime we indeed observe the expected increase in E_{sat}, we believe that our technique would have been sensitive to a relative increase in E_{sat} in the bandwidth-limited regime. As we have attempted to duplicate the experimental conditions used by other groups, we can offer no simple explanation for the quantitative disagreement between our results and those of Refs. 1-3. We note that this disagreement is especially significant in the case of Ref. 3, since these workers alone also observe a relative increase in E_{sat} between the intermediate and bandwidth-limited regimes. Otherwise, it has been suggested[8] that multiphoton absorption in the gain medium could mask an increase in E_{sat} as the peak power is increased with decreasing pulsewidth. This explanation seems unlikely, however, since the effect of nonlinear absorption must exactly compensate an increase in E_{sat} to account for our observation of no detectable change in E_{sat} within the experimental error.

Recall that previously reported values of E_{sat} are obtained from Eq. (1), which is derived by explicitly excluding nonsaturable linear absorption. To evaluate the importance of nonsaturable loss on gain measurements in XeCl, we measured the small-signal linear absorption coefficient, α, as a function of wavelength, λ, for a neon-based gas mixture. These measurements are plotted in Figure 2, and show that $\alpha = 0.014$ cm^{-1} for $\lambda > 313$ nm. There is an apparent decrease in absorption at shorter wavelengths, which is of course due to the onset of gain as the wavelength approaches 308 nm. We therefore assume that the nonsaturable loss is constant across the gain profile and equal to the off-band value of 0.014 cm^{-1}. Since the small-signal gain is 0.195 cm^{-1}, $\alpha = g_0/14$.

With nonsaturable absorption included, pulse propagation in the amplifier is described by the equation[10]

$$\frac{dE}{dx} + \alpha E + g_0 E_{sat}\left[\exp(-E/E_{sat}) - 1\right] = 0 \quad (2)$$

where E(x) is the energy fluence at position x in the amplifier. Equation 2 can be numerically integrated over the amplifier length to give E_{out} as a function of E_{in}. To determine the effect of nonsaturable absorption on our measurements of E_{sat}, Equation 2 is integrated using E_{sat} and g_0 as fitting parameters with the measured value of α substituted as a nonadjustable constant. The quality of the fits to the saturation data is unaffected by the inclusion of nonsaturable loss. We find that E_{sat} increases to 1.35 mJ/cm^2 for 160-fs and 3-ps pulses, and increases to 3.0 mJ/cm^2 for 600-ps pulses. Although the inclusion of nonsaturable loss does increase E_{sat} in all pulsewidth regimes, the values of E_{sat} in the intermediate and bandwidth-limited regimes are still identical.

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2. KINETICS AND GAS CHEMISTRY OF VERY LARGE E-BEAM PUMPED KrF LASERS

2.1. The Effect of ASE on the Performance of Large KrF Amplifiers

The design of large KrF laser systems for breakeven inertial confinement fusion (ICF) is tending toward the use of a small number of very large final amplifiers. These amplifiers would have lengths of several meters with apertures of 1 m x 2 m. In amplifiers of this size, amplified spontaneous emission (ASE) competes with efficient energy extraction, even for relatively low small-signal gains (< 2%/cm). To study the effect of volumetric ASE on such large amplifiers, a new computer model has been developed at Los Alamos National Laboratory[11]. Parameters in this model are the small signal gain, gain-to-loss ratio, saturation intensity, wall reflectivity and amplifier dimensions. To benchmark this model, measurements were made of small-signal gain in the Large Aperture Module (LAM) of the Aurora KrF laser system[12]. This double-sided e-beam pumped device is presently the largest excimer amplifier in operation and has a gain length of 2 m and an aperture of 1 m x 1 m.

Gain measurements were made under three different conditions inside the amplifier. The first consisted of the normal, fully pumped, unrestricted volume. Measured small-signal gains in this mode were ≈1.6%/cm for a gas pressure of 700 Torr, and F_2 concentration of 0.3% and a diode voltage of 650 kV. The average pump power for these conditions was ≈125 kW/cm^3. Previous measurements under similar conditions on smaller amplifiers in the Aurora chain, where ASE is not significant, typically showed a gain to pump power ratio of ≈2.0%/cm per 100 kW/cm^3. Thus, the small-signal gain in the LAM in the absence of ASE is estimated to be ≈2.5%/cm.

The second set of measurements were made in a restricted volume using two horizontal plates mounted 20 cm apart in the center of the amplifier. This should reduce the amount of ASE and, hence, increase the measured gain. Under the same pumping conditions, measured small signal gains in this smaller volume were ≈2.1%/cm – a significant increase over the 1.6%/cm in the larger volume.

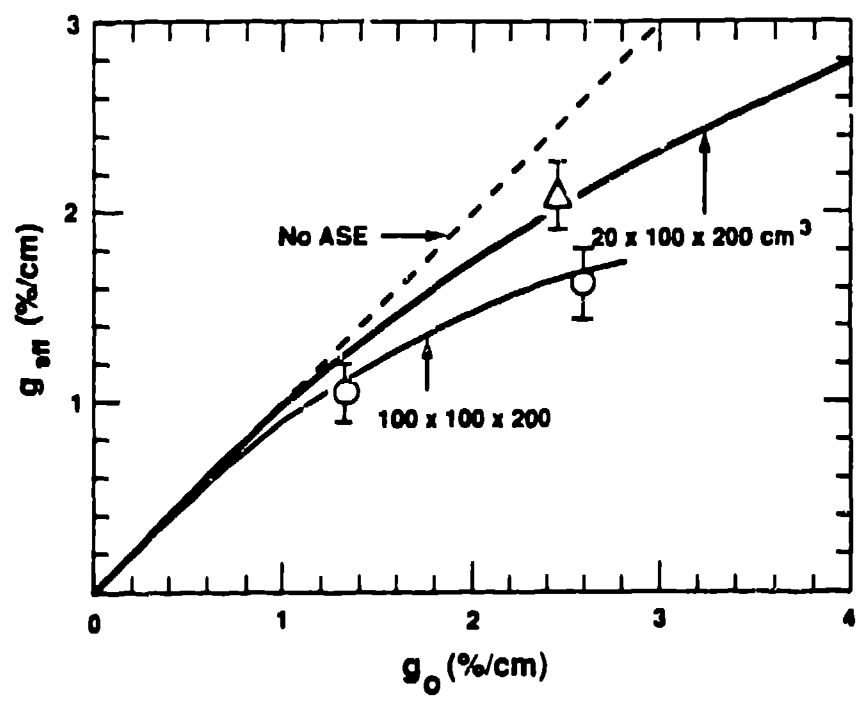

Fig. 2. Computer calculations of the reduction in effective gain caused by ASE in large KrF amplifiers.

The last set of measurements were taken with copper mesh screens installed immediately after the electron beam foils. These screens intercepted ~50% of the electrons and consequently cut the pump power inside the amplifier in half. Since ASE is nonlinear, it should decrease more than a factor of two and, thus, the measured small signal gain with the screens should be more than half the gain without the screens. Measured small signal gains using these screens were ~1.0%/cm, in agreement with the above analysis.

Figure 2 shows the computer model prediction of the effect of ASE on LAM performance as a function of g_0 (the small-signal gain that would obtain in the absence of ASE) along with the data from the gain measurements. As can be seen in the figure, the model compares favorably with the measured values. Quantitative measurements of sidelight fluorescence were also in agreement with model predictions[13].

Thus, the basic predictions of the ASE model have been verified, and it can be used with confidence to project the performance of other large KrF amplifiers. In general, it indicates that ASE will exact a penalty of only 15% in very large, well-designed KrF amplifiers.

2.2. Absorption spectroscopy of rare-gas halide trimers

The spectroscopic properties of the rare gas-halide trimers have been under investigation for a number of years now. These molecules are of interest due to their role in the kinetics of rare gas-halide excimer lasers. Since the trimers are formed during three-body collisions involving the excimers, trimer densities can be quite substantial in high pressure, electron-beam-pumped lasers. This can have a deleterious effect on the excimer output energy, because the trimers are predicted to absorb at the excimer output wavelength. However, measurements of the trimer absorption spectra have been hampered by the difficulty in creating the trimers in the absence of other absorbing species This problem has been solved by exciting large concentrations of rare gas-halogen mixtures in a discharge and probing with a broad band lamp.

Transient absorption measurements have been made to determine the absorption spectra of the rare gas-halide trimers. Large densities of absorbers were created in a small discharge laser having MgF_2 windows and a discharge volume of ~24 cm^3. The laser was typically filled with 400 to 500 mB of a rare gas (Ar, Kr or Xe), 5 mB of F_2 or HCl, and helium to a total pressure of 2.5 Bar. Light from a pulsed, xenon flashlamp was collimated by a short focal length fused silica lens, apertured and passed through the discharge before being focused with an achromatic lens onto the 25-μm entrance slit of a 0.25-m spectrometer. The spectrometer dispersed the light onto a gated, intensified, ultraviolet-sensitive diode array, and a 300-groove/mm grating gave a 200-nm range over the active portion of the diode array. A 20-ns gate pulse was applied to the array detector at the peak of the 300-ns lamp pulse. Timing between the lamp, discharge device and high voltage pulser was controlled with a delay generator, and the detection of discharge fluorescence with a photodiode allowed the drift of the thyratron-switched discharge to be observed and corrected for.

A detector controller was interfaced to a computer that stored and analyzed the data. Each individual absorption spectrum required three data runs of typically 600 shots per run at a repetition rate of 10 Hz. The first run consisted of acquiring both the lamp light and the discharge fluorescence, and it contained the absorption information. The next two runs collected lamp light and discharge fluorescence separately. Subtraction of background light was automatic. Absorption was calculated by subtracting the discharge fluorescence spectrum from the lamp + discharge spectrum, dividing by the lamp spectrum and taking the natural logarithm of the quotient.

The dependence of the $4^2\Gamma$ absorption for each of the rare gas halide trimers on wavelength is seen in Fig. 3. The spectra were recorded at a delay of 60 ns between the start of the discharge and the end of the gate pulse. Two $4^2\Gamma$ absorption features are evident in the figure. The ($4^2\Gamma \rightarrow 9^2\Gamma$) bands peak at 295, 315 and 340 nm for Ar_2F, Kr_2F and Xe_2Cl, respectively, and each band has a half width of 85 nm. A second band for

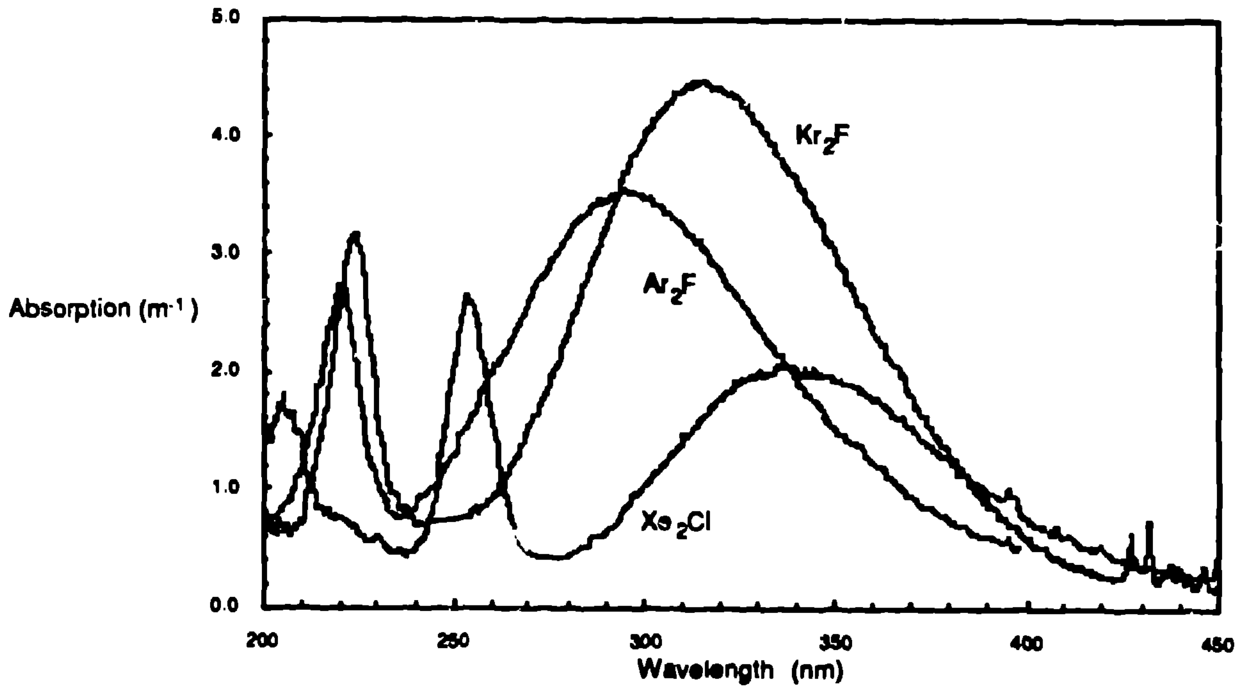

FIG. 3. Absorption spectra of the rare gas halide trimers ($4^2\Gamma$). The ($4^2\Gamma \rightarrow 9^2\Gamma$) absorption bands peak at 295, 315 and 340 nm for Ar_2F, Kr_2F and Xe_2Cl, respectively. The bands peaking at wavelengths below 250 nm are also due to $4^2\Gamma$ absorption. The narrow lines seen above 425 nm are metastable absorption features.

each trimer is also observed at wavelengths less than 250 nm. The time and pressure dependences of the absorption and trimer fluorescence are identical, conclusively identifying the trimer as the absorber.

In summary, continuous measurements of the absorption cross section for the trimers Kr_2F, Ar_2F and Xe_2Cl have been made using a transient absorption set-up. The ($4^2\Gamma \rightarrow 9^2\Gamma$) band has been observed for each of the trimers, as well as another band to the blue that has not been predicted to exist. Experiments are currently in progress that will extend these measurements down to as low as 125 nm.

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