Amplification of ultrashort pulses in krypton fluoride at 248 nm

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Ultrashort pulses were produced at 248 nm by using a transverse-discharge KrF* excimer laser as a high-energy amplifier system. Input pulses for the amplifier system were obtained by upconverting the output of a mode-locked visible dye laser to the ultraviolet by using nonlinear crystals. Pulses of up to 20 mJ in energy with 10–30-psec duration were obtained at 10 pulses/sec. The output pulse width was characterized by using an electronic autocorrelator.

The rare-gas halogen excimer-laser systems have long been recognized as attractive media for the production of high-power picosecond pulses in the ultraviolet region of the spectrum. Of particular interest are the XeF*, XeCl*, KrF*, and ArF* excimers, which emit at 351, 308, 248, and 193 nm, respectively. An early attempt by Christensen *et al.*[1] to produce short pulses utilized active mode locking of the XeF* laser and achieved minimum pulse widths of 1–2 nsec. Tomov *et al.*[2] used a XeF* laser as an amplifier of light pulses produced by harmonic generation of the output of a mode-locked Nd:glass laser. This resulted in laser pulses of 200-psec duration. These techniques, as well as others, were later extended to XeCl* in several laboratories.[3–7] In the case of KrF* at 248 nm, only passive mode locking has been explored.[8,9] Here again, the pulse width obtained was slightly under 2 nsec because of the low number of round trips (3 or 4) available during the approximately 10-nsec duration of the discharge.

In the present work, we have used standard nonlinear optical techniques to upconvert the output of a well-mode-locked dye laser to 248 nm for amplification in KrF*. Similar work is under way[10] to amplify ultrashort pulses in ArF* at 193 nm. A block diagram of our apparatus is shown in Fig. 1. The ultrashort pulses were produced by a synchronously pumped mode-locked dye laser similar to the system described by Wokaun *et al.*[11] This system consisted of an actively mode-locked, *Q*-switched Nd:YAG oscillator, which produced a train of 70-psec, 1.06-μm pulses with an interpulse separation of 8 nsec, and an overall *Q*-switched envelope of approximately 200 nsec. After the beam was amplified and frequency doubled in KDP, the 532-nm pulse train pumped the dye-laser oscillator (DCM dye). A single pulse at approximately 648 nm was then amplified and frequency doubled, and the 324-nm radiation that resulted was summed with a single 70-psec, 1.06-μm pulse in KDP to produce the desired 248-nm wavelength. The amplified 648-nm laser pulse length was measured by background-free second-harmonic generation,[12] yielding a second-order autocorrelation FWHM of 20 psec. Assuming a Gaussian pulse shape, this corresponds to an actual FWHM pulse width of 14 psec.[12] The 248-nm pulse may have been somewhat shorter because of the quadratic intensity dependence of second-harmonic generation. Up to 20 μJ of energy per pulse was available at 248 nm. The bandwidth at 248 nm was inferred from measurements of the 648-nm linewidth by observation of the interference fringes in transmission through a high-finesse étalon. The extra contribution to the 248-nm linewidth from the 1.06-μm beam was negligible since the Nd:YAG laser produced transform-limited 70-psec pulses. It was found that the bandwidth for each 648-nm laser pulse was essentially equal to the Fourier-transform limit (1 cm^{-1}); however, the shot-to-shot frequency jitter was as high as 10 cm^{-1}. This jitter could be eliminated by reconfiguring the dye-laser oscillator cavity at the expense of an increase in pulse width to about 30 psec.

Amplification of this laser pulse occurred in the 85-cm-long discharge region of a Lambda Physics EMG 200 excimer laser with the mirrors removed and the windows tilted by approximately 20° to eliminate feedback. However, even with the complete absence of optical feedback, the gain was sufficiently high that approximately 60 mJ of amplified spontaneous emission (ASE) was emitted from each end of the amplifier. A 1:4 cylindrical telescope was used to match the input beam to the 6-mm × 30-mm cross section of the discharge. Reflection losses here typically reduced the pulse energy at the amplifier input to 10–12 μJ. The

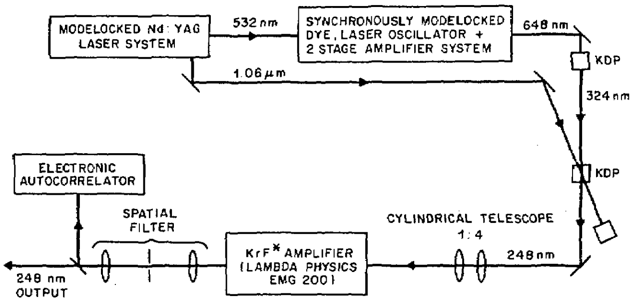

Fig. 1. Block diagram of laser system for production of ultrashort ultraviolet pulses and amplification by a KrF* excimer laser.

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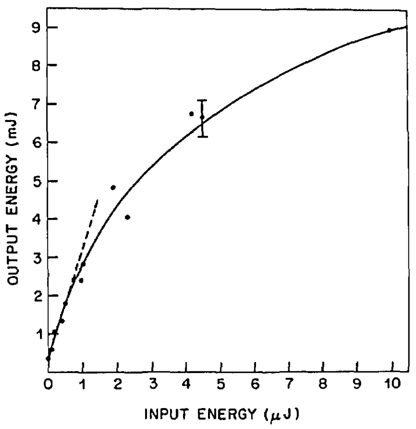

Fig. 2. Input–output characteristics of the KrF* amplifier. The points are the measured data, and the solid line is a fit to a standard two-level model. The small-signal gain (slope of the dotted line) is 3500.

discharge was approximately 20 nsec in duration. Relative timing between the input pulse and the discharge was not critical; a 2-nsec timing jitter was tolerated.

In Fig. 2, the input–output characteristics of the amplifier are shown together with a fit to the usual Franz–Nodvik formula[13] for a two-level system:

$$E_0 = E_{\mathrm{sat}} \ln|1 + \exp(g_0 l)[\exp(E_i/E_{\mathrm{sat}}) - 1]|,$$

where E_0 represents the output energy density and E_i represents the input energy density. The best fit corresponds to a small-signal gain $\exp(g_0 l)$ of 3500 and a saturation-energy density E_{sat} of 2.1 mJ/cm^2. For this measurement, the input wavelength was tuned to the peak of the KrF* gain curve at 248.5 nm. The highest amplified energy observed was 20 mJ, obtained with approximately 20 μJ of input energy.

It is of interest to compare these values with those obtained in the amplification of ~2-nsec pulses in a 45-cm-long KrF* discharge.[9] In that experiment, a saturation-energy density E_{sat} of 3.2 mJ/cm^2 was obtained. One cause for this disagreement is the vastly different time scales of the two experiments. The gain-saturation-recovery dynamics in XeCl* show substantial gain recovery in 40 psec.[4] This effect may also be present in KrF*, which would tend to reduce the measured value for E_{sat} and the small-signal gain for pulses substantially shorter than the fast-recovery time constant.

Gain recovery in XeCl* has been attributed to a combination of rotational relaxation in the upper laser level (B) and vibrational relaxation and dissociation in the weakly bound lower laser level (X).[4] In KrF*, the X state is unbound and dissociates in a time substantially shorter than 1 psec, but it is reasonable to assume that rotational relaxation of the B state occurs on a similar time scale to that of XeCl*. This may then result in a reduction of the accessible rotational population for our 10–30-psec narrow-band pulse. Population transfer to the B state from the nearly degenerate C state on a time scale longer than 30 psec could also contribute to the discrepancy between our values for E_{sat} and those measured with 2-nsec pulses. Finally, there is an additional complication in this experiment from the spatial nonuniformities of the input pulse derived from the dye-laser system. This also tends to decrease the measured value for E_{sat}.

The small-signal gain for 2-nsec pulses obtained in Ref. 9 was 650 in a 45-cm discharge, giving a gain constant of 0.145 cm^{-1}. In order to compare this value with the one obtained for our system, the differences in excited-state densities between the two systems must be estimated. When operated with a standard flat–flat optical resonator, the amplifier used in Ref. 9 was reported to produce 200-mJ pulses. In the same configuration, the amplifier used in this work produced 1000-mJ pulses. If we now normalize to the volumes of the two amplifiers (45 cm × 1 cm^2 for Ref. 9, compared to 85 cm × 1.8 cm^2 for this work), and take into account the difference in small-signal gain that is due to the incomplete rotational relaxation discussed above, an expected gain constant of 0.14 cm^{-14} is obtained for our amplifier. This corresponds to a total gain of e^{12} = 1.7 × 10^5. The measured gain, obtained from the data of Fig. 2, is only 3500. The large discrepancy is most likely caused by saturation of the gain by the strong ASE output, which has a gain constant (undiminished by incomplete rotational relaxation) of 0.21 cm^{-1} and a total gain some 400 times larger than the gain for the input pulse. The analogous phenomenon in dye amplifiers for ultrashort pulses has been analyzed.[14] In obtaining the data of Fig. 2, the strong ASE background was suppressed with a simple spatial filter consisting of a 500-mm focal-length lens and a 0.5-mm aperture. This reduced the background to 0.4 mJ, which appears in Fig. 2 as the nonzero intercept.

An important characteristic of the output is the pulse width, which we would like to keep as short as possible. However, under conditions of fairly strong gain saturation, the possibility arises of pulse-shape distortion by the amplifier. In this respect, amplification of ultrashort pulses in the 10–20-nsec-duration excimer discharges is closely analogous to the amplification of ultrashort dye-laser pulses in dye amplifiers pumped by the second harmonic of a Q-switched Nd:YAG laser, for which it has been shown[14] that the output pulse length depends critically on the input pulse shape and intensity. In strongly saturated amplifiers, it is in general difficult to keep the output pulse widths from broadening.[14] For this reason, it is extremely useful to have some means of monitoring the output pulse width. The usual technique of autocorrelation by second-harmonic generation is not applicable to these ultraviolet laser pulses since there exists no nonlinear crystal capable of generating the second harmonic of 248 nm. Recently, ultraviolet short-pulse-width measurement by multiphoton-ionization autocorrelation was demonstrated.[15]

We have developed a general method for measuring ultraviolet laser pulse widths that is based on the electronic autocorrelator of Auston.[16] Briefly, two fast photoconducting switches are connected in series with a voltage bias. A modification of the usual variable-delay interferometer is used in which one beam is sent to each of the photoconductors. It can be shown[16] that,

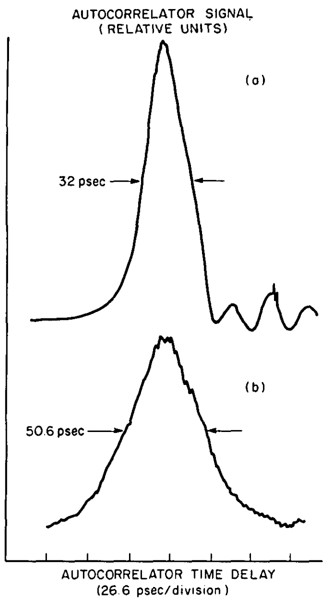

Fig. 3. Measurement of the ultraviolet laser pulse width using an electronic autocorrelator. (a) Impulse response of the device, as measured with 2-psec dye-laser pulses. The subsidiary peaks are due to electronic reflections. (b) Amplified 248-nm pulse. After appropriate deconvolution of the device impulse response, assuming a Gaussian pulse shape, the 248-nm pulse width obtained is 28 psec.

if the pulse duration of the input laser is longer than the response time of the photoconducting switches, the amount of charge flowing through the two photoconductors as the relative time delay is varied is proportional to the second-order autocorrelation function of the laser pulse, convolved with the impulse-response function of the switches. The response time of these devices can be somewhat shorter than 10 psec.[17] In Fig. 3 we show some results obtained by using an electronic autocorrelator to characterize the amplified ultraviolet laser pulse. Figure 3(a) shows a measurement of the basic response of this particular device. This trace was taken using pulses from a cw mode-locked dye laser of ~2-psec duration and shows an impulse response of 32 psec. Figure 3(b) shows the results obtained using the amplified 248-nm laser pulses as input. In this case, the dye laser producing the input to the amplifier was generating 30-psec pulses, as measured by second-harmonic generation. The amplifier output was 10 mJ, and the beam was heavily attenuated before input to the photoconductors. The output pulse width, after deconvolution of the device impulse response, and assuming a Gaussian pulse shape, was 28 psec. This procedure was checked by comparing an electronic autocorrelation of the 30-psec dye-laser pulse with the result obtained by second-harmonic generation. The error in the measurement was estimated to be 20%. Thus, to within this accuracy, no pulse broadening was observed. As was mentioned above, the dye-laser system may be configured to produce 14-psec, 648-nm pulses. In this configuration, the amplified 248-nm pulse width has not been measured, but this should be possible in the near future by using an electronic autocorrelator with 10-psec impulse response.

In conclusion, a system has been constructed for the generation of ultrashort ultraviolet laser pulses in the 15–30-psec range, with peak powers of over 1 GW. Future development will concentrate on multiple passes through the excimer amplifier. In the regime of strong saturation, it may be possible to extract up to 100 mJ of energy from this amplifier module. Careful pulse shaping will undoubtedly be required to accomplish this without significant pulse broadening.[14]

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