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High-brightness terawatt KrF* (248 nm) system

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Abstract. Experimental results that demonstrate a reproducible peak power of 1 TW, with a pulse energy of 250 mJ at a wavelength of ~ 248 nm are presented. The output pulse is focusable down to intensity levels in the 10^{19} W/cm^2 range with improved shot-to-shot energy stability. This system gives a resulting brightness of $\sim 3.3 \times 10^{21}$ W cm^{-2} sr^{-1}.

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The combination of ultrashort pulse generation and chirped pulse amplification (CPA) techniques has been the driving force in the development of laser systems with peak powers at the terawatt level [1–6]. Most high-power systems developed to date work in a spectral region between 0.8 and 1.1 microns, with TW-class lasers being demonstrated in Ti:Sapphire-, Nd:glass- and Cr:LISAF-based systems. A known viable option that does not rely upon solid-state materials operating in the infrared is offered by excimer lasers. The large-gain bandwidth of these systems makes them suitable amplifiers for extending the realm of experimental high-intensity physics into the ultraviolet (UV), and efforts in this direction have been undertaken by a number of groups [7–9].

In this letter we report on the performance of a Ti:Sapphire/KrF* laser system leading to the generation of 250 mJ, 250 fs, low divergence pulses at a wavelength of 248 nm. Figure 1 shows the schematic of the system setup. The initial pulses are obtained by means of a home-built, hard-apertured Kerr lens mode-locked (KLM) Ti:Sapphire oscillator. This laser is optimized for stable operation at a central wavelength of 745 nm using a single-plate birefringent filter with a tuning range of 720–850 nm. The average mode-locked power from this system is 380 mW when pumped by 6.7 W of an all-line Ar-ion laser (Coherent Innova 300). The pulse duration is measured to be 85 fs, assuming a sech^2 envelope shape, at a repetition rate of 86 MHz. The pulse is amplified by the use of the conventional CPA technique. Gold-replica gratings (groove density of 1200 lines/mm) are used in a single-grating stretcher/single-grating compressor combination positioned to give a stretch factor of 500.

After the initial stretching, the infrared pulses are sent through a dye and a triple-pass Ti:Sapphire amplifier. The first stage provides a pulse selection, since the relatively short (~ 100 ps) upper-state lifetime of the dye (LD7OO Perchlorate, concentration 0.01 M in methanol) gives an interval long enough for amplification but shorter than the spacing between successive pulses. The repetition rate of the amplified pulses is determined by the dye pumping rate, which occurs at a frequency of 2 Hz. The dye cell is arranged in a double-pass configuration, as illustrated in Fig. 1. The 523 nm radiation provides longitudinal pumping, and the two passes are counterpropagating with respect to the pump. This arrangement reduces the effect of mode beating in the pump pulse, a process caused by the short upper-state lifetime of the dye, thus reducing fluctuations in the gain. As a consequence, the fluctuations in the energy output are restricted to $\pm 20\%$, a significant improvement in energy pulse distribution from the previous transversely pumped dye laser amplifier, which exhibited energy fluctuations of $\pm 40\%$ [10]. Further improvement is expected by substituting the dye cell with a regenerative amplifier which is presently under design. The dye system provides amplification by a factor of approximately 300. Further amplification is achieved by means of a triple-pass Ti:Sapphire amplifier (gain 1000), which brings the total energy of the single pulse to 1.5 mJ. Both of the described amplification stages are pumped with a frequency-doubled Q-switched Nd:YAG (Continuum NY6O) that furnishes 300 mJ to the triple-pass amplifier with a residual 25 mJ used to pump the dye.

The recompressed pulse duration has been measured to be ~ 90 fs, with a spectrum centered at 745 nm of FWHM $= 10$ nm. This gives a resulting time–bandwidth product of the order of 0.5, which suggests the possible existence of a pedestal quantifiable at less than 10% of the peak intensity. The tripling process would then reduce it to levels where it would not become of consequence in the following ultraviolet amplification stages (i.e. a 10% pedestal would be reduced to $\sim 0.1\%$ in the tripled pulse). Furthermore, the observations on the UV spectra at various points in the

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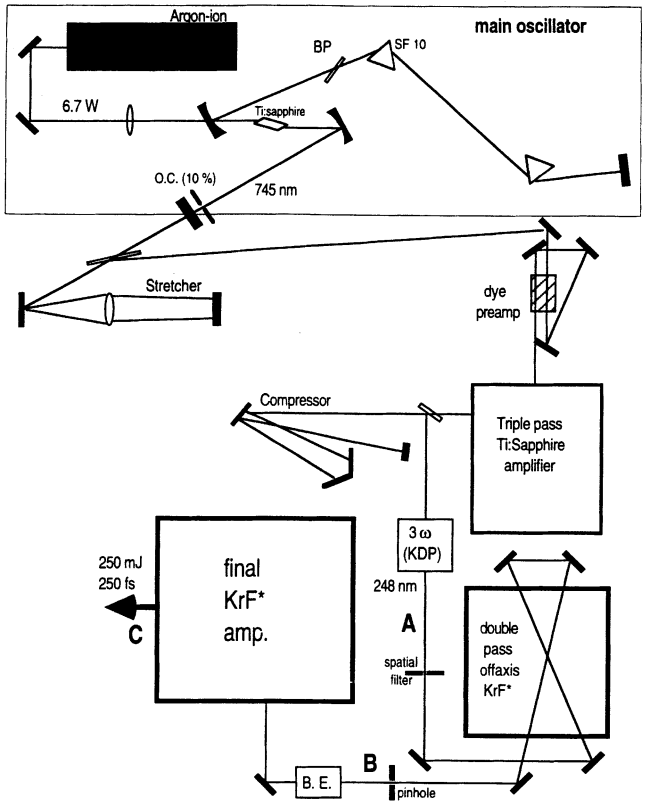

Fig. 1. Schematic of the terawatt Ti:Sapphire/KrF* laser system (O.C.–output coupler, B.E.–beam expander)

chain and the measurements on the pulses (compounded by experimental evidence using the beam) after the amplification stages suggest the absence of any significant temporal structure. Frequency conversion takes place at this point, and the pulses are sent, unfocused, through two KDP crystals of thickness 3 mm and 1 mm, respectively, for doubling of the Ti:Sapphire radiation (47.4° cut, type I interaction) and mixing (82.1° cut, type I interaction) of the produced 372 nm radiation with the residual 745 nm fundamental. A delay stage is included in order to optimize the spatial overlap between the fundamental and the second-harmonic radiation in the mixing crystal [7]. The collinear geometry and the thicknesses of the KDP crystals used in the tripling scheme cause pulse broadening due to group velocity mismatch (GVM) between the fundamental and the second/third harmonic. The calculated increase in pulse duration, assuming a 90-fs input at the tripler, anticipates an output duration of ~ 140 fs, whereas the recorded spectrum at the output of the tripler (point A, Fig. 1) exhibits a bandwidth (FWHM) of ~ 0.91 nm which could support a transform-limited pulse of duration ~ 70 fs, assuming a sech^2 envelope shape for the pulse. It is, nevertheless, important to have a high conversion efficiency in the frequency-tripling process to ensure maximum energy transfer into the first UV amplifier and thus a good contrast ratio between the signal and the amplified spontaneous emission (ASE) in the amplifier. Typical output energies from the tripler are 15 μJ at 248 nm for a 1 mJ, 745 nm input, which are sent to seed the excimer preamplifier. To ensure good beam quality, the beam is spatially filtered in vacuum after the tripler by focusing it into a 50-μm pinhole, which reduces the seed energy to 12.5 μJ.

The first UV amplification stage is performed in a KrF* excimer (Lambda Physik 201 MSC EMG) arranged in a double-pass off-axis geometry [11]. Both the electric excitation circuit and the laser chamber are slightly modified to meet the requirements of off-axis amplification. The device is operated with a 22-kV discharge voltage and a pressure of 2000 mbar (80 mbar F_2, 120 mbar Kr, 1800 mbar Ne). The off-axis angles in the two paths are designed to achieve optimum amplification over the whole beam dimension, compensating for any inhomogenieties in the gain profile of the excimer to eliminate, for instance, the presence of hot spots in the beam. This design ensures optimum performance in the subsequent amplification stage, where a good input spatial profile enables efficient energy extraction from the final amplifier with good contrast and, as a consequence, an output beam of excellent focusability and homogeneity [12].

The duration of the 248 nm pulse after the double-pass excimer is obtained through a two-photon fluorescence (TPF) measurement which relies on the two-photon induced F-center formation in a fused silica window. A small piece (thickness ~ 2 mm) of fused silica is positioned so that two counterpropagating pulses, which are replicas of one another, overlap inside the silica plate. Imaging of the resulting two-photon fluorescence excited by these short pulses requires high resolution and very accurate focusing inside the crystal. The fluorescence images were formed by a long working distance microscope, on a CCD camera (CID 2710): the resolution obtained, calibrated with a USAF test pattern, is 2.4 microns per pixel. After the first UV amplifier, the pulse duration is found to be 160 fs (Fig. 2), with an energy of 500 μJ. The spectrum (Fig. 3) taken at this point (point B, Fig. 1), shows a slight narrowing compared with the one taken after the tripler (point A, Fig. 1). The measured pulse duration concurs with the fact that the first amplification stage does not cause broadening of the pulse, and confirms that the walkoff between the fundamental and higher-frequency pulses due to GVM in the tripler is the main cause of temporal broadening of the pulse. Similar measurements were performed using barium fluoride as the two-photon fluorescent medium. The fluorescence signals were found to be significantly weaker, and it was not possible to locate the focus at the required magnification, thereby preventing accurate measurements of the sub-200 fs pulses.

The final amplification is obtained by means of a large aperture (10 cm) excimer amplifier [12]. The beam is focused and sent through a second pinhole in order to filter out any ASE leaking from the first excimer, with a measured energy contrast ratio between the ASE and the amplified UV beam greater than three orders of magnitude. The beam is then directed through a telescope beam expander before entering the amplifier. This power amplifier is operated at relatively low pressure and low gain in order to reduce wave-front distorsions and ASE. Produced at a repetition rate of 0.4 Hz, the pulses exhibit an average energy of 250 mJ (peak values above 400 mJ have been observed with the different dye preamplifier setup, at the expense of shot-to-shot stability), and their duration was measured, giving a value of 250 fs (Fig. 2). The spectrum, taken at point C (Fig. 1), retains the same characteristics, with a resulting bandwidth of 1.1 nm (FWHM) (Fig. 3) and, once again, shows no evidence of self-phase modulation. The slight shift in the central wavelength and the broadening in the spectrum are attributed to gain saturation effects. The same effects are also the probable cause of temporal broadening, since cal-

culations on the linear dispersion taking place in the CaF_2 and MgF_2 windows rule out linear dispersion as a temporal broadening mechanism, thus leading to the conclusion of the possible presence of high-order chirp in the final pulse.

The average energy of 250 mJ ± 20% is observed consistently throughout the daily operation of the system. This is a sensible improvement over the previously detected values, which exhibited fluctuations of the order of 85% with an average value of 240 mJ. The spectral features (0.95 ± 0.15 nm detected in points A, B, C, Fig. 3) are consistent as well, and no drift has been detected if not due to the degradation of the excimer gas. The superior beam quality allows reproducible focusability which has been measured to be two times the diffraction limit (a few microns) leading to very desirable experimental conditions. Assuming a top-hat beam, the resulting brightness is found to be $\sim 3.3 \times 10^{21}\,\mathrm{W\,cm^{-2}\,sr^{-1}}$.

In conclusion, a femtosecond laser system that routinely operates at the terawatt power level in the UV region of the spectrum (248 nm) has been demonstrated. The pulse is found to exhibit no evidence of self-phase modulation throughout the amplifier chain and has a high energy stability. The homogeneity of the beam profile allows efficient amplification with superior focusability. These are the essential conditions that permit the investigation of physical interactions of materials with reproducible ultraintense, ultrashort pulses at high energy levels. Further improvements are expected by eliminating the remaining cause of energy fluctuation, namely the dye preamplifier, which would lead to even more desirable performance in the ultraviolet.

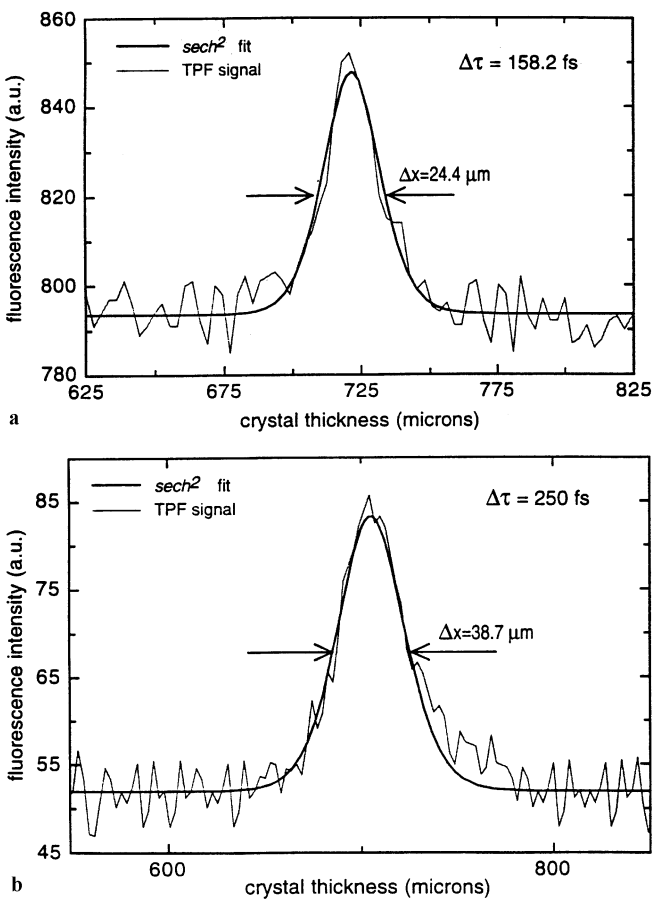

Fig. 2. Measurements of the pulse durations at the outputs of the double-pass KrF^* amplifier (point B, Fig. 1) and of the large aperture KrF^* amplifier (point C, Fig. 1). Both fits assume a sech^2 envelope shape for the pulses for the temporal deconvolution

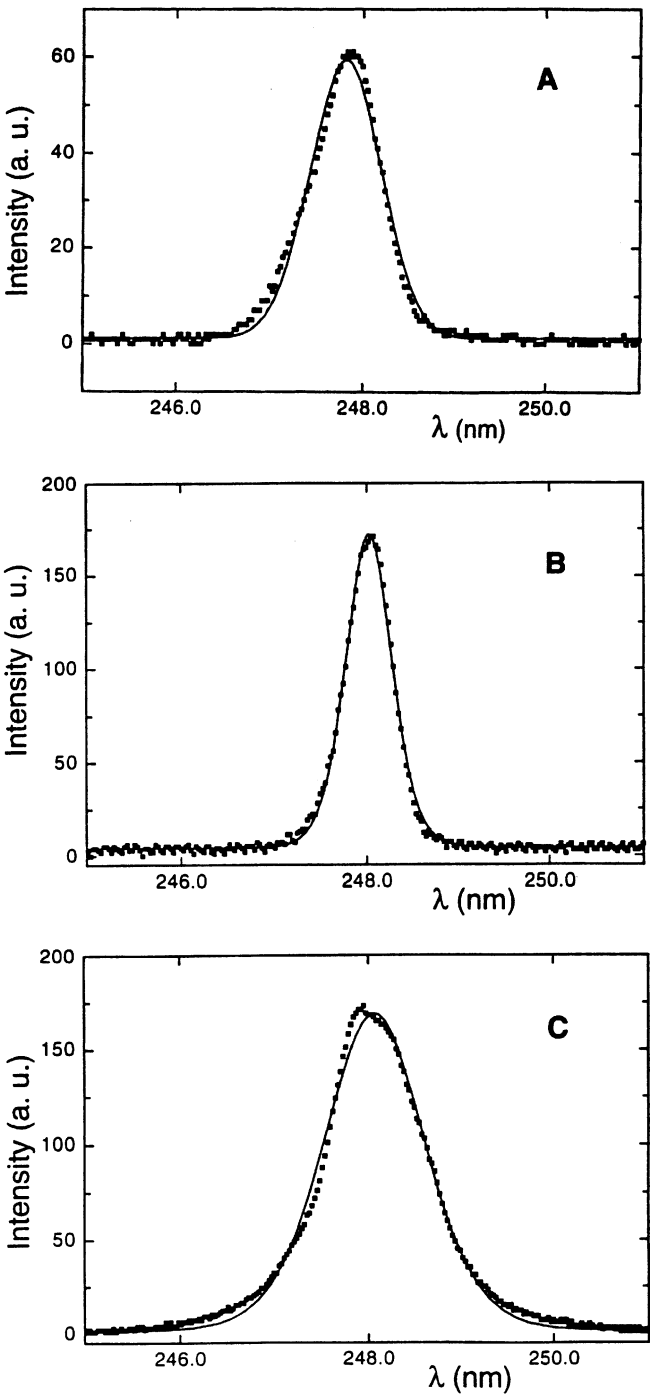

Fig. 3. Spectral evolution of the pulse throughout the system after the frequency tripler (point A, Fig. 1), the double-pass KrF^* amplifier (point B, Fig. 1), and the large aperture KrF^* amplifier (point C, Fig. 1). The spectral widths are fitted to a sech^2 profile in the first case and to a Lorentzian profile for cases **b** and **c**. The values of the widths at FWHM are evaluated to be, respectively 0.91, 0.74, and 1.1 nm

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