

## Subpicosecond UV Pulse Generation for a Multiterawatt KrF Laser

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**Abstract.** A 180-fs UV pulse has been generated based on a hybrid synchronously pumped mode-locked dye laser for a multiterawatt KrF laser system. The pulse width was measured by the single shot autocorrelation technique with the three-photon fluorescence of the XeF C-A transition. The pulse width broadening due to dispersive media was investigated. The results show that the observed pulse width broadening from 210 fs to 390 fs through the entire system is explained mostly by the linear dispersion of the optical elements for near-transform-limited input pulses.

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A multiterawatt (TW) excimer laser is an attractive light source for multi-photon processes and XUV lasers [1]. Over the past few years, many efforts have been devoted to reducing the pulse width [2-4], and increase the output energy [5, 6]. In our institute, a multiterawatt excimer laser system including wideaperture discharge lasers and an electron-beampumped laser has been completed. A peak power of 1 TW was obtained at XeCl in 310 fs with an amplified spontaneous emission (ASE) less than 2% in energy within the discharge stage [7]. Recently, a 4-TW peak power was obtained in 390 fs with an ASE of 1.8% for KrF by using the entire system [8]. The initial pulse width of 210 fs was broadened to 390 fs through amplification. In this paper, subpicosecond (sub-ps) UV pulse generation and measurement for the multiterawatt KrF system are described with an emphasis on the pulse width broadening due to dispersive media in the system.

A schematic diagram of the generation of 248-nm pulses is shown in Fig. 1. The hybrid synchronously pumped mode-locked dye laser generated sub-ps pulses at 745 nm. The mixed solution of pyridine 2/DDI [9] was used in a single jet as the combination of a gain medium and a saturable absorber. The average power was 30 mW. The frequency-doubled output of a mode-locked cw Nd: YAG laser (Spectra Physics series 3000, PRF 82 MHz) with a fiber-grating

pulse compressor (Spectra Physics 3690) was used to pump the dye laser with a pulse width of 3.5 ps at 530 nm. Autocorrelation measurement showed the pulse width at 745 nm to be 290 fs with some temporal wings present as in Fig. 2a. The spectral width was



Fig. 1. Schematic diagram of the generation of sub-ps pulses at 248 nm



Fig. 2. a Background-free autocorrelation and b corresponding spectrum of 745-nm pulses at the oscillator

2.1 nm, as shown in Fig. 2b. The time-bandwidth product is 0.33, which is very close to the value for a transform-limited sech<sup>2</sup> pulse. Amplification of a single selected pulse in the train was accomplished by a fourstage dye amplifier chain, which was pumped by a 5.5-ns, 75-mJ XeCl laser. In the amplifier chain, LD-700 (Exciton) was used in all stages. Conventional side-pumped cells were used for the first three stages and a prismatic cell with a bore of 2 mm was used for the final stage. The dve concentration was  $1.3 \times 10^{-3}$  M in methanol for the first and second cells. The third and fourth cells were used with the concentration of  $6.5 \times 10^{-4}$  and  $3.5 \times 10^{-4}$  M in methanol, respectively. To reduce ASE, saturable absorbers were inserted between every stage. The solid saturable absorber (Schott glass RG850) was placed between the first and second stages, and HITC saturable absorber dye jets were used between the later stages. The amplified energy was 480 µJ. The total gain through the chain was more than  $10^6$ . The explicit difference in pulse width and spectrum was not observed after the amplifier chain. But the temporal wings almost disappeared during amplification. The autocorrelation trace and spectrum are shown in Fig. 3a and b, respectively. The amplified 745-nm pulse was frequency-doubled in a 1 mm-thick KDP crystal and then sum-frequency



Fig. 3. a Background-free autocorrelation and **b** corresponding spectrum of 745-nm pulses after the dye amplifier chain

mixed with its second harmonic in a 1 mm-thick BBO crystal. The energy of the 248-nm seed pulse was a few microjoules. The UV pulses were amplified by two high repetition rate KrF preamplifiers, which have the same aperture size of  $2 \text{ cm} \times 1 \text{ cm}$ , and active lengths of 30 cm and 60 cm, respectively. First, the pulses were amplified in the 30-cm laser and then in the 60-cm laser with the double-pass configuration employing a vacuum spacial filter between each pass. A typical output energy was 10 mJ with a negligibly small content of an ASE background. The beam divergence was 50 µrad, which is twice the diffraction limit.

The newly devised method based on the XeF C-A transition (480 nm) was used to measure the sub-ps KrF pulses [10]. Autocorrelation traces were obtained in each shot by employing the typical arrangement with the triangular configuration. The pulses were focused by a 60-cm focal-length lens and collided precisely at the focal point in the gas cell. The fluorescence image was enlarged by a factor of 5–10 with a camera lens, and detected by the SIT camera.



Fig. 4. a Autocorrelation trace and b corresponding spectrum of a single 248-nm pulse after the preamplifiers

The pulse width after the preamplifiers was measured to be 180 fs by using this method. The width of autocorrelation trace was divided by 1.29 with regard to three-photon fluorescence by assuming a sech<sup>2</sup> pulse. The single shot autocorrelation trace and spectrum are shown in Fig. 4a and b, respectively. The observed contrast ratio was close to the theoretical value of 10:1.

The pulse was amplified to 1.5 J by a 7 cm  $\times$  7 cm aperture discharge laser and a 23 cm  $\times$  23 cm aperture laser. The pulse width after the final amplifier was measured to be 390 fs when the pulse width at the preamplifier was 210 fs. Self-phase modulation was not observed from the final spectrum. In order to investigate the pulse broadening due to the dispersion of optical element in the KrF system, the change of a pulse width was measured through quartz and CaF<sub>2</sub> blocks as dummy optics. The thicknesses of quartz and CaF<sub>2</sub> were 100 mm and 74 mm, respectively, which almost coincide with those of the entire system. The autocorrelation traces of the pulses before and after the dummy optics were obtained simultaneously by the single shot technique. The experimental arrangement is shown in Fig. 5. The spectra were also monitored



Fig. 5. Schematic of the experimental setup to measure the change of the pulse width



**Fig. 6.** The pulse width after dummy optics versus the input pulse width. The straight line corresponds to the case without broadening. The curve shows the theoretical prediction

before and after the dummy optics simultaneously. The explicit change of a spectral width and shape was not observed between them at an energy of 1 mJ with a cross section of  $2 \text{ cm} \times 1 \text{ cm}$ . The input pulse width was varied by small changes in the adjustment of the hybrid mode-locked dye laser resonator. The pulse width after the dummy optics is plotted versus the input pulse width in Fig. 6. The pulses around 200 fs were almost transform-limited when assuming a sech<sup>2</sup> pulse, while the pulse width around 500 fs was nearly twice the transform-limited value. The time-bandwidth products of all data were within twice those of transformlimited pulses. At the input pulse of 210 fs, the pulse was broadened to 360 fs. This result is close to the pulse width obtained after the final amplifier. The autocorrelation trace and spectrum of the 4-TW system is shown in the previous paper [8]. At the shortest input pulse of 180 fs, the output pulse width was 330 fs. This fact shows that pulse-width broadening was mainly due to the thick optics in the entire system. The pulse-width broadening was previously analyzed in a CPM ring

laser with regards to a frequency sweep and a dispersion by assuming a gaussian pulse shape [11]. As shown in Fig. 6, the theory which neglects frequency sweep is in relatively good agreement with the measurements above an input range of 300 fs to within experimental error, although the input pulses around 500 fs were not transform limited. However, an increase in pulse width predicted by the theory below 300 fs was not observed in this experiment. The reason for this difference is not so clear within the scope of this experiment. This difference would arise from the nature of the input pulse such as a pulse shape and a frequency sweep. But in this experiment the frequency sweep in the input pulse is small because the observed time-bandwidth product is very close to the transformlimited value for a sech<sup>2</sup> pulse below 300 fs. Then, the observed pulse width broadening would be explained by the linear dispersion of the optical elements for near-transform-limited pulses with regards to the fluctuation of the system.

In conclusion, a sub-ps pulse source for the multiterawatt KrF laser system was developed. The pulse width of 180 fs at 248 nm was obtained with a system based on the hybrid synchronously pumped dye laser. Pulse-width broadening through dummy optics was investigated, resulting in good agreement with the observed pulse width increase from 210 fs to 390 fs in the amplification experiment with the full system. The broadening due to the dispersion of the optics will be compensated by using appropriate elements with a negative dispersion such as a prism pair in the early stage.

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