Appl. Phys. B 41, 49–52 (1986)

Applied Physics B Photophysics and Laser Chemistry



# Single Picosecond UV Pulse Generation by Mode-Locking of an Excimer Laser-Pumped Dye Laser

M. Watanabe, A. Endoh, and S. Watanabe

The Institute for Solid State Physics, The University of Tokyo, Roppongi 7-22-1 Minato-ku, Tokyo, 106 Japan

Received 9 April 1986/Accepted 19 May 1986

**Abstract.** A single picosecond ultraviolet pulse has been generated based on mode-locking of a dye laser pumped by a long pulse XeCl laser to serve as the input source for a high-power ps KrF laser system. A short-pulse uv dye laser (BBQ) pumped by an additional XeCl laser was used to selectively amplify a single pulse from a mode-locked pulse train with the pulse separation of 3.2 ns. The amplified single pulse was frequency-doubled to 248 nm with the pulse duration of 20 ps.

**PACS:** 42.55.Mv, 42.55.Hq, 42.60.By

High-power and picosecond (ps) excimer lasers in the ultraviolet (uv) wavelength region have stimulated the applications in various fields, including the generation of coherent extreme-ultraviolet (xuv) light [1–3], multiphoton processes [4], and surface science [5]. High-power ps rare-gas halide laser system is now under construction in our institute [6] for the investigation of nonlinear optics, and solid-state physics in the uv or xuv wavelength regions and for basic researches on x-ray lasers. We have developed a ps pulse generation system to serve as the input source for this laser system. To obtain such uv ps pulses, various methods have been presented until now. Mode-locking of rare-gas halide lasers was investigated [7, 8]. But the pulse duration was limited to a few hundred picoseconds because of the limited number of round trips and the lack of an effective saturable absorber. With the use of stimulated Brillouin scattering in liquid, a pulse duration less than 100 ps was obtained from a XeCl laser [9]. The other approach was a wavelength conversion from dye lasers tuned to the multiple wavelengths of rare-gas halide lasers [10–17]. Picosecond pulses were easily generated from a mode-locked dye laser due to the availability of adequate saturable absorbers and well-established pumping sources including cw lasers. The ps pulses were obtained at the XeCl [11], KrF [13], and ArF [15] wavelengths using mode-locked dye lasers pumped by Ar or frequency-doubled Nd:YAG lasers. In XeCl [12] and KrF [14], sub-picosecond pulse generation using an optical fiber pulse compression was reported recently. But these systems require the complicated synchronism between the ps generator and excimer amplifiers. As for cost and simplicity, the system based on excimer lasers is desirable. Along this line a single ps pulse was obtained by an unique system based on a distributed-feedback laser pumped by a XeCl laser for injection into a KrF amplifier [16]. Recently, a pulse duration of less than 40 ps was generated, using a simple technique of the cavity transient of a quenched dye laser [17], and the pulse was reduced to 9 ps with the saturated amplification of a KrF laser.

In this paper, we report on the generation of a single ps pulse at the KrF laser wavelength from a mode-locked dye laser system pumped by two XeCl lasers with different pulse durations. A single pulse with a pulse duration of 20 ps was obtained at 248 nm. A long-pulse XeCl laser with total pulse duration of more than 200 ns was used to pump a mode-locked dye laser. A short-pulse uv dye laser system pumped by a XeCl laser with a pulse duration of 10 ns (full width at half maximum, FWHM) was used to selectivity amplify a single pulse from the mode-locked pulse train with a pulse separation of 3.2 ns.

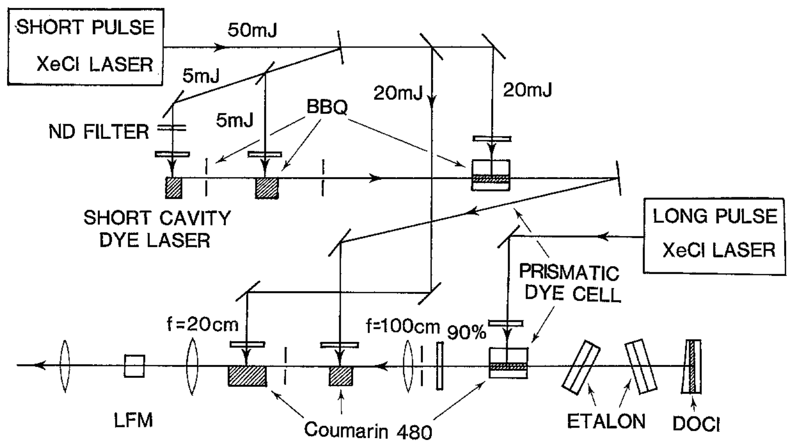


Fig. 1. Experimental setup of the excimer-laser-pumped dye-laser system

## Experiments and Discussions

A schematic of the total system is shown in Fig. 1. Mode-locking of a dye laser pumped by a long pulse XeCl laser was described in [18]. For single-pulse amplification from the mode-locked pulse train, stable and low-jitter operation of the two XeCl lasers is required. Especially in the long-pulse XeCl laser, the high repetition capability is desirable for the alignment of the system. For this purpose, a magnetic switch and a thyratron were used rather than a rail-gap and a spark gap, which were used in the previous experiment [18]. The circulation fan and cooling fin were added in the laser head for high-repetition operation. As a result of these improvements, the fluctuation of the output energy was within $\pm 3\,\%$, and the pulse shape was well reproduced in every shot. The jitter at the build up of the pulse was measured to be within a few nanoseconds. The pulse of 220 ns in the total duration having 240 mJ energy was produced at the appropriate condition. The typical optical pulse shape is shown in Fig. 2. The repetition rate was limited to 10 Hz by the capacity of the power supply. The detailed characteristics will be reported in [19].

Mode-locking was performed passively with the combination of Coumarin 480 as a laser dye and DOCI (3,3″ diethyloxacarbocyanine iodide) as a saturable absorber. The cavity for mode-locking was set to 46 cm in length with a 90% reflectance output coupler and a saturable absorber cell of 0.5 mm thickness in contact with the total reflector. A prismatic dye cell [20] with a bore of 1.5 mm in diameter and an active length of 20 mm was used to improve the spatial beam quality. The experiment of mode-locking was carried out with the concentrations of $4.5 \times 10^{-3}$ M methanolic solution of Coumarin 480 and $1.5 \times 10^{-4}$ M methanolic solution of DOCI, respectively. Two air-gap etalons with different free spectral ranges (17 nm and 19 nm, respectively) were used to tune the wavelength to 497 nm. Figure 3 shows the oscillogram of the mode-locked pulse train. It shows the rapid reduction of the background within 10 ns from the start of the pulse train. The improvement due to the prismatic cell compared with the previous paper was not observed about the pulse duration, but a beam divergence of less than 1 mrad was observed with good spatial symmetry. More than 30 pulses were observed during 100 ns, and the total energy was 50 μJ.

Selection of a single pulse from a pulse train becomes easier if the pulse separation is increased. Therefore we investigated the cavity length dependence of the evolution of the mode-locked pulse train. When the cavity was lengthened to 100 cm with a similar cavity arrangement (Fig. 1), the evolution of the pulse train was quite incomplete. The quality of the pulse train was found to deteriorate considerably when the cavity was lengthened beyond 60 cm, resulting in a slow reduction of the background and, in some cases, a

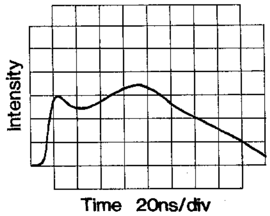


Fig. 2. Typical optical output pulse of the long-pulse XeCl laser

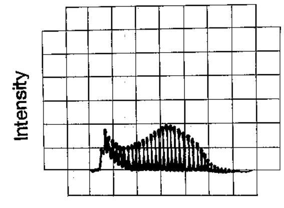


Fig. 3. Mode-locked pulse train at 497 nm with Coumarin 480 as a laser dye and DOCI as a saturable absorber

fault of mode-locking. This result may be explained partly by the relation between the gain recovery time of Coumarin 480 and the round-trip time of the cavity. The gain of Coumarin 480 was recovered before the pulse returned to the gain medium in the cavity when the cavity was longer than 60 cm. Complete modulation was obtained even in the 110 cm-cavity when mode-locking was carried out by adding an acousto-optic modulator in the cavity. However, the pulse duration was not reduced below 100 ps because the number of round trips was limited to ten.

A single pulse among the pulse train was selectively amplified using a two-stage Coumarin 480 amplifier pumped by a conventional short-pulse XeCl laser. The pulse duration of this XeCl laser was 10 ns (FWHM) with an energy of 50 mJ. 10 ns pulses were stably produced with an energy fluctuation of $\pm 2\%$. The temporal jitter was measured to be less than 1 ns. The pulse duration of 10 ns is still too long compared with the pulse separation of 3.2 ns to select a single pulse. Then a uv dye oscillator-amplifier system was employed to reduce the pumping pulse duration, as shown in Fig. 1. The selected single pulse from this first Coumarin 480 amplifier was again amplified in the second Coumarin 480 amplifier directly pumped by a fraction of the short-pulse XeCl laser radiation. The optical beam from the oscillator was softly collimated by a 100 cm focal length lens into the two Coumarin 480 amplifiers, both of which contained $4.5 \times 10^{-3}$ M methanolic solution of Coumarin 480.

The cascade pumping was accomplished by using a short-cavity oscillator and two amplifiers. BBQ (4,4‴ di(2-butyloctoxy)-p-quater-phenyl) in cyclohexane was used for this purpose. The concentrations of BBQ were $1.5 \times 10^{-3}$ M for the short-cavity oscillator and the first amplifier, and $1.0 \times 10^{-4}$ M for the second amplifier, respectively. The short cavity was formed by the dye-cell walls (external dye cell length: 6.5 mm, wall thickness: 1.25 mm). The mirror reflected a small fraction (5 mJ) of the XeCl laser output to pump the short-cavity oscillator and its first BBQ amplifier. The remainder of the XeCl laser beam was split into two parts, one for the second BBQ amplifier and the other for the second Coumarin 480 amplifier. Neutral density filters placed in front of the cylindrical lens were used to adjust the pumping power. When the short-cavity BBQ oscillator was pumped well above the threshold, the pulse shape was almost identical to the pumping pulse with a duration of 10 ns (FWHM). However, when the pumping power was lowered near the lasing threshold, the output pulse was shortened to 3 ns (FWHM). This pulse was amplified to 4 mJ and then directed to the first Coumarin 480 amplifier by two mirrors, which also gave an appropriate timing between the first and second Coumarin 480 amplifiers.

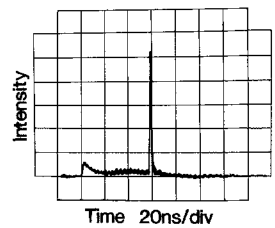


Fig. 4. Selectivity amplified pulse at 497 nm from the first Coumarin 480 amplifier

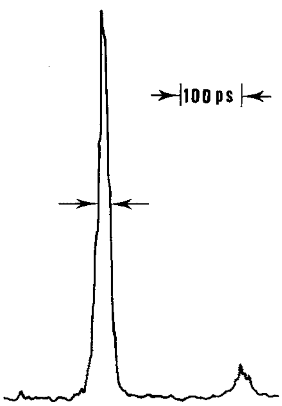


Fig. 5. Streak recording of the pulse form at 248 nm, showing the pulse duration of 20 ps

Figure 4 shows the pulse train from the first Coumarin 480 amplifier. A single pulse of the later part of the pulse train was selectively amplified by a factor of 30. The amplified single pulse was frequency-doubled to 248 nm in an angle-tuned LFM (lithium formate monohydrate) crystal.

After second-harmonic generation, the background of pulse train was not observed on the oscillogram. The pulse duration of 20 ps was measured by a streak camera (Hamamatsu C1370). The pulse energy was approximately 1 μJ. The streak measurement reveals the existence of some subpulses accompanied around the selected pulse in most cases, typically shown in Fig. 5. This is mainly due to the limited number of round trips to suppress subpulses completely. In some cases, double pulses were observed on the oscillogram. This is because the pumping duration for dye amplifiers is close to the pulse separation of the pulse train.

## Conclusion

We have described the single-pulse generation of 20 ps pulses at 248 nm from the excimer laser-pumped dye laser system. The use of a magnetic switch rather than a rail-gap in the 200 ns XeCl laser contributed to low-jitter and high-repetition operation. A single-pulse selection was performed by a BBQ oscillator-amplifier system pumped by 10 ns XeCl laser. Picosecond pulses can be obtained over the wide wavelength region with a little modification of this system.

*Acknowledgement.* The authors wish to acknowledge the assistance of Mr. K. Sajiki in fabrication of excimer lasers.

## References

1. J. Bokor: Opt. Lett. **8**, 217 (1983)
2. J. Reintjes, L.L. Tankersley, R. Christensen: Opt. Commun. **39**, 334 (1981)
3. Ch.K. Rhodes (ed.): *Excimer Lasers*, 2nd edn. (Springer, Berlin, Heidelberg, 1984)
4. K. Boyer, H. Egger, T.S. Luk, H. Pummer, C.K. Rhodes: J. Opt. Soc. Am. B **1**, 3 (1984)
5. P. Haight, J. Bokor, R.H. Storz, J.B. Stark, P.H. Bucksbaum, R.F. Freeman: SPIE **476**, 61 (1984)
6. S. Watanabe, A. Endoh, M. Watanabe: Conference on Lasers and Electro-Optics (June 1984) Anaheim, CA, paper THG2
7. S. Watanabe, M. Watanabe, A. Endoh: Appl. Phys. Lett. **43**, 533 (1983)
8. M. Watanabe, S. Watanabe, A. Endoh: Opt. Lett. **8**, 638 (1983)
9. O.L. Bourne, A.J. Alcock: Opt. Lett. **9**, 411 (1984)
10. M. Maeda, T. Mizunami, A. Sato, O. Uchino, Y. Miyazoe: Appl. Phys. Lett. **36**, 636 (1980)
11. P.B. Corkum, R.S. Taylor: IEEE J. QE-**18**, 1962 (1982)
12. J.H. Glownia, G. Arjavalingan, P.P. Sorokin, J.E. Rothenberg: Opt. Lett. **11**, 79 (1986)
13. P.H. Bucksbaum, J. Bokor, R.H. Storz, J.C. White: Opt. Lett. **7**, 399 (1982)
14. T.S. Luk, A.P. Schwarzenbach, I.A. McIntyre, U. Johann, A. McPherson, K. Boyer, C.K. Rhodes: SPIE **622**, O-E LASE '86, Paper 12
15. H. Egger, T.S. Luk, K. Boyer, D.F. Muller, H. Pummer, T. Srinivasan, K. Rhodes: Appl. Phys. Lett. **41**, 1032 (1982)
16. S. Szatmári, F.P. Schäfer: Opt. Commun. **48**, 279 (1983)
17. B. Burghardt, R. Schwarzwald, Z. Bor, B. Nikolaus: Conference on Lasers and Electro-Optics (June 1985) Baltimore, MD, Paper WR2
18. M. Watanabe, S. Watanabe, A. Endoh: Appl. Phys. Lett. **45**, 929 (1984)
19. S. Watanabe, M. Watanabe, A. Endoh: Rev. Sci. Instrum. (submitted)
20. D.S. Bethune: Appl. Opt. **20**, 1897 (1981)