16-fs, 1-μJ ultraviolet pulses generated by third-harmonic conversion in air

S. Backus, J. Peatross, Z. Zeek, A. Rundquist, G. Taft, M. M. Murnane, and H. C. Kapteyn

Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, Michigan 48109-2099

Received October 30, 1995

We describe a simple method for generating sub-20-fs ultraviolet light pulses with useful average powers, using a kilohertz Ti:sapphire laser system. By focusing a 22-fs, 1-mJ laser pulse in air, we obtain ultraviolet pulses with an energy of 1 μJ and at a wavelength of 266 nm and with an average power of 1 mW. The pulse duration of the ultraviolet pulses was measured to be 16 fs with frequency-resolved optical gating.

We report the generation of ultrashort UV pulses by using nonresonant third-harmonic conversion of a 22-fs driving pulse[1] in a gas. To generate ultrafast UV pulses by harmonic generation it can be preferable to use gases rather than crystals such as β-barium borate. This is so because complications such as finite phase-matching bandwidth and temporal walk-off, which place limits on the UV pulse width, can be avoided if one uses gases. Ringling *et al.*[2] used 800-nm 150-fs laser pulses to generate 180-fs UV pulses with β-barium borate. However, it is unlikely that this approach can be extended to sub-20-fs pulse widths. Excimer lasers also can produce short pulses in the UV, but they are bandwidth limited to pulse durations greater than 100 fs.[3–6] Here we demonstrate the generation of 266-nm light with a measured pulse duration of 16 fs, produced simply by focusing 22-fs laser pulses in air. The 1-mJ laser pulses are converted into 3ω light with an efficiency of $>0.1\%$, giving UV pulses of 1-μJ energy. Higher conversion efficiencies are obtained with argon. At our 1-kHz repetition rates this technique gives an average UV power of 1 mW. Its advantages are the extremely short pulse duration, the relative ease of implementation, and the high repetition rate.

Our research is similar to that of Siders *et al.*,[7] who reported efficiencies as great as 0.01% for third-harmonic conversion of an 800-nm laser pulse in a chamber filled with argon gas. Their 80-fs laser pulses had an energy of 1.5 mJ and were focused with an $f/10$ optic. This corresponds to a nominal (vacuum) peak intensity in the focus of $\approx 10^{16}$ W/cm^2. Such a high-intensity pulse ensures strong ionization breakdown of the gas and hence a radical distortion of the laser beam as it propagates through the focus.[8] When they were collimating the third-harmonic beam they observed a strongly blue-shifted spatial structure with different collimation properties from those of the rest of the third-harmonic beam profile. The bandwidth of this blue-shifted portion was seen to be as much as 10 nm, and they noted that this bandwidth is sufficient to support sub-20-fs pulses under appropriate phase conditions. However, the actual pulse duration was not measured.

The generation and blue shifting of the third harmonic as reported by Siders *et al.* is in qualitative agreement with one-dimensional calculations performed by Vanin *et al.*[9] However, the calculations show a potential conversion efficiency of 1%, whereas the experimentally observed conversion efficiency was 0.01%. They suggested that a larger f-number may improve the conversion efficiency, but to achieve a similar intensity would require more laser energy. In our research we use pulses of approximately the same pulse energy but with much shorter duration. Thus we can implement a longer focusing geometry ($f/30$) without unduly sacrificing peak intensity. This results in an order-of-magnitude increase in conversion efficiency. Because our laser pulses are short, the third-harmonic pulse is naturally also short. In contrast to Siders *et al.*, we do not observe multiple collimations in the UV beam.

The laser pulses were focused with an $R = 60$ cm protected silver mirror and $f/30$ focusing geometry. We determined that the beam focus is within 1.8× of the diffraction limit (in vacuum). On focusing a 1-mJ pulse in air, we observe plasma channels ≈1 cm long, as measured from the recombination light. Figure 1 shows the experimental setup, including the apparatus for measuring the UV pulse duration (described below). The output light was collected and recollimated by an $R = 50$ cm dichroic mirror, which reflected $>99\%$ of the 266-nm light while transmitting ≈90% of the fundamental light. The 3ω light is further selected by three additional mirrors, reducing the fundamental light by a factor of $>10^{-4}$. We verified that the fundamental light did not interfere with our measurements by evacuating the focus and observing that the third-harmonic signal was not present.

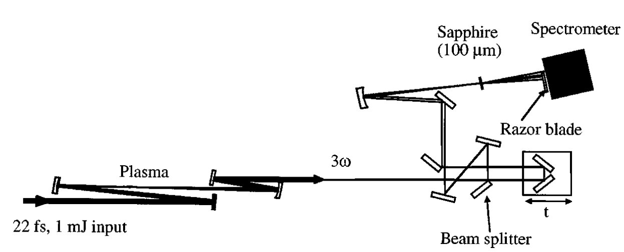

Fig. 1. Schematic of the third-harmonic generation setup and the SD frequency-resolved optical gating pulse-measurement apparatus.

0146-9592/96/090665-03$10.00/0

The laser pulses used in these experiments have a bandwidth of 40 nm, centered at 800 nm, with a near-transform-limited pulse duration of 22 fs. However, we achieve 40% higher output power in the third harmonic (1 μJ) when the excitation pulse is negatively chirped to $\approx$40 fs. The laser pulse emerges from the interaction region also with a negative chirp but with slightly shorter duration. This case corresponds to the maximum 3ω output power and the shortest measured third-harmonic pulse duration of 16 fs. On the other hand, if the excitation pulse is positively chirped to $\approx$40 fs, the output third-harmonic power decreases by 40% relative to the case of a 22-fs excitation pulse. Similarly, a 40-fs transform-limited pulse does not produce as much third-harmonic light as does a negatively chirped 40-fs pulse. We speculate that the dispersion arising from the combination of a rapidly changing plasma index or self-phase modulation and negative dispersion in the plasma favors third-harmonic generation from a negatively chirped 40-fs pulse. We are performing further research to enable us to understand this in detail. The bandwidth of the third harmonic is 6 nm, centered at 266 nm, for the case of the 40-fs negatively chirped excitation pulse, as shown in Fig. 3 below. A measurement of the UV spectrum produced by a transform-limited 40-fs pulse showed a width of $\sim$3 nm. Therefore, with broader-bandwidth excitation pulses, we can generate the shortest UV pulses.

When lower-f-number focusing optics (e.g., $f5$–$f20$) are used the output power of the third harmonic is reduced. However, under such conditions, light at the fundamental wavelength is dramatically blue shifted (many visible colors) and scattered into a cone angle much wider than that of the initial f-number. The strongest third-harmonic output occurred for $f/30$ focusing for our laser pulse energy. At larger f-numbers the third-harmonic signal is also reduced, and the blue shifting and scattering of the laser pulse are reduced.

A variety of schemes have been used to measure pulse durations in the UV.[4–6,10–12] They rely on two-photon absorption and other nonlinear processes. We have used the technique of frequency-resolved optical gating[13] (FROG) to characterize our UV pulses. At the 1-μJ level there is ample energy to do a scanning self-diffraction (SD) FROG measurement. The SD technique retrieves pulse shape information from the diffracted beam scattered by the crossing of two identical beams in a material. This technique is related to the technique used by Heist and Kleinschmidt,[4] who measured subpicosecond UV pulses by the deflection of one beam by another. SD FROG does not require second-harmonic conversion of the UV pulse and thus avoids the problems of working with vacuum-UV light.

Figure 1 shows the setup for the SD FROG measurement, which is similar to that of a standard autocorrelator. The beam is split by being spatially chopped in half with the edge of a mirror. This avoids the temporal broadening owing to dispersion that could be introduced by a conventional beam splitter. At a wavelength of 266 nm the 16-fs pulse can be dramatically broadened by propagation through even 1 mm of material. The two beams in the FROG setup are delayed relative to each other and then focused by an $R = 30$ cm mirror with a crossing angle of 2 deg. We used a 100-μm-thick sapphire plate as the nonlinear medium for the SD FROG measurement. When the two pulses are crossed in the sapphire, a nonlinear grating is induced in the material as a result of the optical Kerr effect. This grating scatters a small fraction of each beam at twice the original crossing angle. The self-diffracted beam can then be separated easily from the two original beams and the spectrum of the SD beam monitored as a function of relative delay between the two pulses. The data can be displayed as a two-dimensional plot, which is then used to reconstruct the electric field of the pulse. A FROG trace corresponding to a 16-fs UV pulse is shown in Fig. 2(a). The spectrometer camera integrated $\approx$100 pulses for each spectral trace, corresponding to a given time delay. The pulse-to-pulse laser energy fluctuation is $\sim$2%.

Figure 2(b) shows the pulse temporal profile retrieved by the SD FROG deconvolution algorithm. Figure 3 shows both the measured and the retrieved spectra of the pulse. The 6-nm spectral width can in principle support a pulse as short as 12 fs, in con-

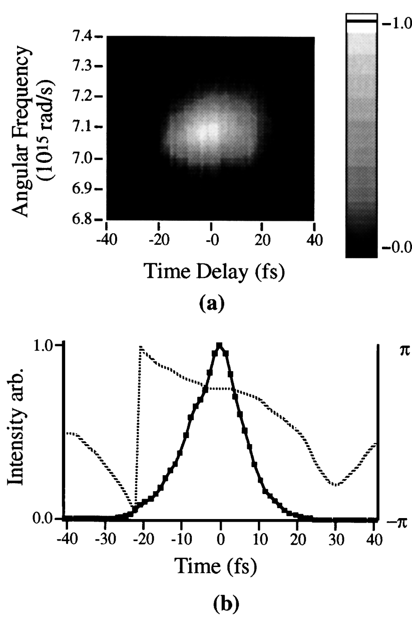

Fig. 2. (a) Self-diffraction scanning FROG trace of a 16-fs 266-nm pulse. (b) Temporal profile of the pulse retrieved by the FROG algorithm. The solid curve shows the intensity amplitude, and the dashed curve shows the phase.

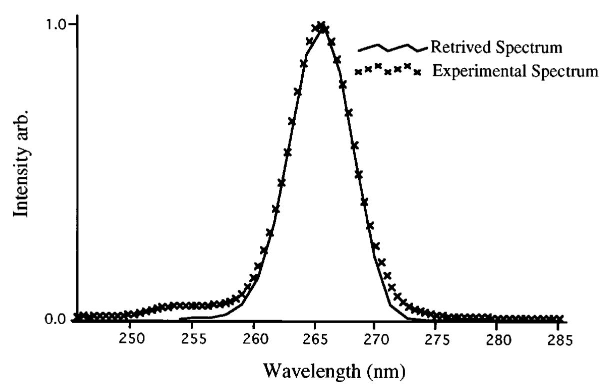

Fig. 3. Measured (crosses) and retrieved (line) spectra of a 16-fs, 266-nm pulse.

trast to the 16-fs pulse width measured. However, from Fig. 2(b) we can see that the phase of the electric field varies in time across the pulse, which would indicate some residual chirp. This residual chirp can be explained by the dispersion introduced by the air in the beam path between the generation region and the FROG measurement apparatus. We placed the FROG setup as close as possible to the generation region, but the beam path in air was still ≈ 1 m. When we increased this beam path, the duration of the pulse increased. Computer simulations show that a 12-fs UV pulse propagation through 1 m of air will disperse to ≈ 16 fs, in agreement with our experimental measurements. Therefore, with proper dispersion compensation, pulses as short as 12-fs in the UV might be measured.

As the laser pulse energy is reduced from 1 mJ to 120 μJ the UV output power decreases. For excitation energies of <120 μJ the third-harmonic generation is below our detection limit of ≈ 10 nJ/pulse. The cutoff of the UV signal is abrupt and sensitive to small changes in the laser pulse energy. This apparent threshold also corresponds to the disappearance of visible recombination light in the focus. The correlation between the recombination light and the third-harmonic generation could indicate that phase-matching symmetry breaking occurs during ionization and thus enhances the harmonic generation. Alternatively, it might also indicate that atoms emit third-harmonic radiation most strongly during ionization. It should be noted that in the perturbative limit the net third-harmonic emission from a Gaussian laser focused into a gas cell is zero, owing to macroscopic phase-matching symmetries.[14,15] It may be that the plasma breakdown in the focus plays an important role in breaking the symmetry and thus permits more efficient harmonic generation.

We also examined harmonic generation in argon. For these experiments we placed the focusing optics in a vacuum chamber that was backfilled with argon. We observed a conversion efficiency approximately twice that of air. We also noticed that the conversion efficiency for either gas did not change significantly as the pressure was varied from 1 atm to $\sim 1/3$ atm. At even lower pressures, the third-harmonic generation decreased monotonically with pressure, until at ~ 1 Torr the signal dropped below our detection level of 10 nJ.

We plan to attempt to increase the conversion efficiency by refocusing the fundamental and third-harmonic light through a series of focii, with appropriate relative phase. We will also study fifth- and seventh-harmonic generation, using ultrashort pulses, to generate sub-20-fs light in the vacuum UV. Calculations show[16] that this process should be efficient and should lead to similarly short pulse widths.

In conclusion, we have demonstrated the generation of 12–16-fs UV pulses with microjoule energies and at a repetition rate of 1 kHz. These pulses are to our knowledge the shortest UV pulses generated to date. The experimental setup used is simple, and the pulse shapes were characterized by the technique of self-diffraction (SD) FROG. This research can be extended to generate sub-20-fs pulses throughout the UV and the VUV for application experiments.

The authors gratefully acknowledge funding by the U.S. Department of Energy. H. Kapteyn acknowledges support from a Sloan Foundation Fellowship.

References

1. S. Backus, J. Peatross, C. P. Huang, M. M. Murnane, and H. C. Kapteyn, Opt. Lett. **20,** 2000 (1995).
2. J. Ringling, O. Kittelmann, F. Noack, G. Korn, and J. Squier, Opt. Lett. **18,** 2035 (1993).
3. J. Ringling, O. Kittelman, F. Noack, U. Stamm, J. Kleinschmidt, and F. Voss, Opt. Lett. **19,** 1639 (1994).
4. P. Heist and J. Kleinschmidt, Opt. Lett. **19,** 1961 (1994).
5. A. Tunnermann, H. Eichmann, R. Henking, K. Mossavi, and B. Wellegehaussen, Opt. Lett. **16,** 402 (1991).
6. P. Simon, H. Gerhardt, and S. Szatmari, Meas. Sci. Technol. **1,** 637 (1990).
7. C. W. Siders, N. C. Turner III, M. C. Downer, A. Babine, A. Stepanov, and A. M. Sergeev, J. Opt. Soc. Am. B **12,** 330 (1996).
8. S. C. Rae, Opt. Commun. **104,** 330 (1994).
9. E. V. Vanin, F. P. Schafer, and J. Jethwa, Rev. Sci. Instrum. **61,** 998 (1990).
10. Y. Takagi, Appl. Opt. **33,** 6328 (1994).
11. S. Szatmari, F. P. Schafer, and J. Jethwa, Rev. Sci. Instrum. **61,** 998 (1990).
12. H. Nishioka, M. Ishiguro, T. Kawasumi, K. Ueda, and H. Takuma, Opt. Lett. **18,** 45 (1993).
13. K. W. DeLong, R. Trebino, and D. J. Kane, J. Opt. Soc. Am. B **11,** 1595 (1994).
14. J. F. Ward and G. H. C. New, Phys. Rev. **185,** 57 (1969).
15. G. C. Bjorklund, IEEE J. Quantum Electron. **QE-11,** 287 (1975).
16. I. P. Christov, Department of Physics, Sofia University, BG-1126 Sofia, Bulgaria (personal communication, 1995).