Generation of intense 8-fs pulses at 400 nm

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Frequency-doubled pulses from a sub-40-fs, 1-kHz Ti:sapphire amplifier system are spectrally broadened in an argon-filled hollow waveguide. Compression of the self-phase-modulated pulses is implemented with chirped mirrors and a prism pair, yielding 8-fs, 15-μJ pulses in the violet spectral range. © 1999 Optical Society of America

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Optical pulse compression has been established as a powerful technique for generating intense sub-10-fs pulses in the red and near-infrared spectral range. Self-phase modulation (SPM) in a single-mode fiber followed by propagation through a dispersive delay line consisting of prisms and diffraction gratings was successfully exploited more than a decade ago for production of 6-fs, 1-nJ, 8-kHz pulses at 620 nm.[1] Recently, further pulse shortening was achieved at higher repetion rates and (or) higher pulse energies. Pulses near 800 nm at a 1-MHz repetition rate with durations as short as 4.5 fs and energies up to 6 nJ were obtained by use of a cavity-dumped Ti:sapphire oscillator and an improved dispersive delay line incorporating chirped mirrors and a prism pair.[2] Replacement of the single-mode fiber with a gas-filled hollow waveguide allowed scaling to much higher pulse energies.[3] SPM in a gas-filled hollow fiber followed by dispersive compression in ultrabroadband chirped dielectric mirrors yielded 4.5–5-fs submillijoule pulses with peak powers up to 0.1 TW at a 1-kHz repetition rate.[4,5]

The generation of intense sub-10-fs pulses was recently extended into the 500–700-nm wavelength range by use of a continuum-seeded noncollinear optical parametric amplifier pumped by the second harmonic of a Ti:sapphire laser.[6,7] To our knowledge, these are the shortest wavelengths at which powerful sub-10-fs light pulses had been produced. In the violet–ultraviolet range microjoule-energy pulses of ~20 fs in duration were demonstrated recently.[8–10] In this Letter we report the generation of 15-μJ, 8-fs pulses at a wavelength of approximately 400 nm.

This experiment draws on our previous work, which resulted in the production of powerful 20-fs pulses tunable from 360 to 440 nm by use of a hollow-waveguide quartz-prism compressor seeded with the frequency-doubled output of a 1-kHz femtosecond Ti:sapphire amplifier system.[10] Pulse shortening was found to be limited by a residual third-order spectral phase left uncompensated in the compressor stage. This system has been improved in several respects. First, shorter frequency-doubled seed pulses with a duration of 29 fs are used. Second, SPM is carefully optimized in the hollow waveguide. Last but not least, the quartz-prism compressor is supplemented with specially designed chirped mirrors that allow reduction of undesirable third-order dispersion (TOD) in the compression system.

Powerful ultrashort pulses at 800 nm are generated with a home-built Ti:sapphire laser system that utilizes chirped-pulse amplification.[11] The seed pulses, as generated by a 15-fs chirped-mirror-compensated Ti:sapphire oscillator,[12] are stretched by a factor of 10,000 in an all-reflective pulse stretcher[13] capable of controlling higher-order dispersion. A lamp-pumped Q-switched intracavity-frequency-doubled Nd:YLF laser delivering an average output power of 23 W at 532 nm pumps a 10-round-trip regenerative Ti:sapphire amplifier and a two-pass Ti:sapphire power amplifier at a repetition rate of 1 kHz. The 2.4-mJ amplified pulses, with a spectrum peaking at 800 nm, are recompressed with a 50%-transmitting reflective grating pair to a pulse duration of 38 fs.

Efficient frequency doubling of downcollimated 1-mJ 38-fs fundamental pulses is achieved in 100-μm-thick β-barium borate (BBO), resulting in violet pulses with energies up to 400 μJ and a spectral width of 8 nm [see Fig. 1(a)]. Changing the distance of the compressor gratings in the amplifier system slightly introduces a small negative chirp in the fundamental that precompensates for dispersion in the doubling crystal and allows us to achieve high conversion efficiencies, up to 40%. Self-diffraction (SD) third-order autocorrelation measurements of the violet pulses in a 100-μm-thick potassium dihydrogen phosphate (KDP)

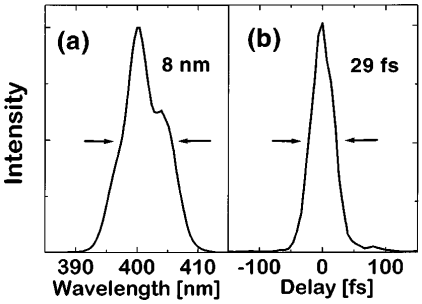

Fig. 1. (a) Spectrum and (b) SD autocorrelation trace of the output of the BBO crystal. A correlation width of 39 fs corresponds to a pulse width of 29 fs.

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crystal [Fig. 1(b)] reveal a significantly shorter pulse duration [29 fs (FWHM)] for the second harmonic than for the fundamental.

For spectral broadening, violet pulses of 300-μJ energy are focused with a concave mirror with radius $R = 1$ m into a hollow waveguide with a bore diameter of 220 μm and filled with argon at pressure $p = 0.4 - 0.6$ bars (1 bar = 10,000 Pa). The choice of argon as the nonlinear medium is a good compromise between a moderate ionization threshold and moderate nonlinearity, whereas in the case of helium or neon the ionization properties are better but the nonlinearity is much smaller. Even at these moderate gas pressures the intense light induces strong spectral broadening, as shown in Figs. 2 and 3. We estimate the calculated optimum fiber length L_{opt} for generating a spectrally broadened pulse with a nearly quadratic spectral phase (i.e., linear chirp in the time domain) to be 40 cm for $\lambda_0 = 400$ nm, using $L_{\text{opt}} = (6L_{\text{nl}}L_D)^{0.5}$,[14] where $L_{\text{nl}} = 1/\gamma\, P_0 = 1.3$ cm and $L_D = {T_0}^2/\beta_2 = 1.9$ m. Here $P_0 = 26$ GW is the peak power of the seed pulses, and $T_0 = 48$ fs is defined by $P(T_0) = P_0/e$. Further, $\gamma = n_2\omega_0/cA_{\text{eff}} = 3.01 \times 10^{-11}$ W^{-1}cm^{-1} is the nonlinear coefficient, $n_2 = 3.10 \times 10^{-19}$ cm^2 W^{-1} is the nonlinear refractive index at $p = 0.6$ bars,[15] A_{eff} is the effective cross-sectional area of the hollow waveguide, and $\beta_2 = 12.41$ fs^2 cm^{-1} is the group-velocity dispersion, including contributions from the waveguide and the argon gas.[16] To introduce some discrimination against higher-order transverse propagation modes, which may also be excited to some extent owing to incomplete matching of the free-propagating input beam to the fundamental EH_{11} mode of the waveguide, we use a 70-cm-long waveguide. The output pulse energy delivered in the fundamental mode amounts to approximately 80 μJ, implying a throughput of 26%. The low throughput is assigned to a low coupling efficiency owing to the elliptic beam profile of the second-harmonic pulse, preventing good mode matching to the EH_{11} mode simultaneously in both transverse directions.

The fiber output is collimated with a concave aluminum mirror with $R = 2$ m and reflected six times off chirped multilayer mirrors consisting of 42 alternating layers of SiO_2 and Ta_2O_5. The mirrors exhibit high reflectivity from 330 to 460 nm and are expected to introduce a nearly constant group-delay dispersion (GDD) of approximately -20 fs^2 from 350 to 430 nm. We show a theoretical estimate of the GDD and the TOD in Fig. 2. Employing these chirped mirrors with negligible TOD and minimizing the optical path length in the prisms allows us to reduce the prism apex distance of 45 cm, compared with 130 cm in our previous experiment.[10] Although the dispersion characteristics of the mirrors have still to be measured, we deduce from the shorter prism distance that the mirrors introduce the amount of GDD per bounce that was predicted. The exclusive use of chirped mirrors for pulse compression is currently prevented by the low dispersion and the high loss (10%) that they introduce per bounce. The origin of this anomalously high reflection loss under exposure to high peak intensities remains to be found.

Figure 3 summarizes the results of a SD frequency-resolved optical gating (FROG) measurement[17] of compressed pulses generated at an argon pressure of $p = 0.4$ bars. Figures 3(a) and 3(b) depict the spectrum and the measured SD correlation (filled circles) of the compressed pulses, respectively. The measured SD FROG trace is shown in Fig. 3(c). Figure 3(d) plots the retrieved intensity (solid curve) and phase (dashed curve) of the pulse, yielding a pulse duration (FWHM) of 13 fs and a small amount of residual phase at the pulse center.

The shortest pulse was generated by an increase in pressure of 0.6 bars, which led to further spectral broadening, as revealed by Fig. 4(a). A slight readjustment of the amount of glass introduced by the first prism proved necessary for optimum pulse compression. The SD trace of the compressed pulses [Fig. 4(b)] results in a third-order autocorrelation width of less than 11 fs. We recorded this trace with the stringent requirement of acquisition by use of only those pulses for which the energy and bandwidth fluctuations were less than ±5% of the average value. This excludes any erroneous broadening of the signals owing to intensity fluctuations of the seed pulse. Unfortunately, however, we have not yet implemented these stringent requirements in the acquisition of FROG traces. To determine the deconvolution factor

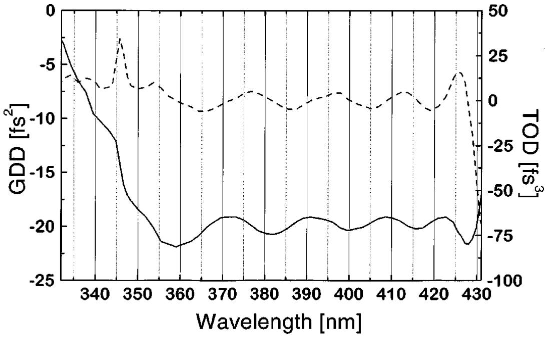

Fig. 2. Theoretical estimate of the GDD (solid curve) and the TOD (dashed curve) properties of the chirped mirrors.

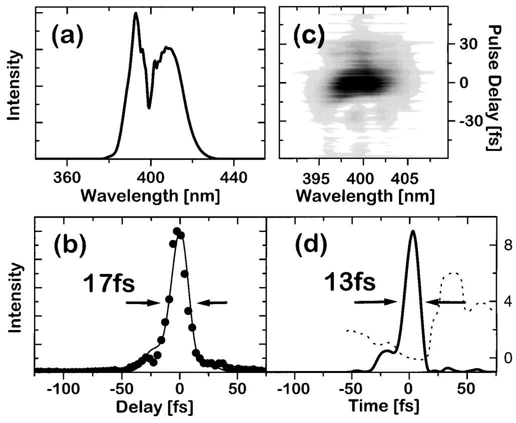

Fig. 3. (a) Spectrum, (b) SD autocorrelation, (c) SD FROG, and (d) retrieved pulse intensity and phase of compressed blue pulses obtained with 0.4-bar Ar in the hollow fiber. A calculated SD trace [solid curve in (b)] compares well with the experimental values (filled circles).

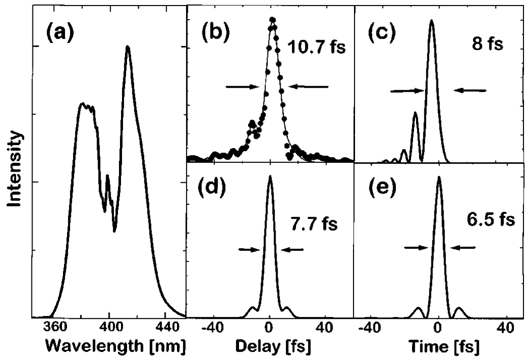

Fig. 4. SD autocorrelation [filled circles in (b)] of compressed blue pulses obtained with 0.6-bar Ar in the hollow fiber with the spectrum given in (a). The solid curves in (b) and (c) are the calculated SD signal- and pulse-intensity profiles, respectively, as described in the text. The Fourier-transform-limited SD signal and the temporal intensity profile are shown in (d) and (e), respectively.

and the residual higher-order phase, we can analyze the compressed pulses only by a fit of the spectral phase,

$$\Phi(\omega) = \Phi_0 + \left.\frac{d\Phi}{d\omega}\right|_{\omega_0}(\omega - \omega_0) + \frac{1}{2}\left.\frac{d^2\Phi}{d\omega^2}\right|_{\omega_0} \times (\omega - \omega_0)^2 + \frac{1}{6}\left.\frac{d^3\Phi}{d\omega^3}\right|_{\omega_0}(\omega - \omega_0)^3 + \dots,$$

where the second- and third-order Taylor expansion coefficients are to be chosen to provide the best fit to the measured SD autocorrelation trace in Fig. 4(b). This fit is obtained with

$$\left.\frac{d^2\Phi}{d\omega^2}\right|_{\omega_0} = 0\ \text{fs}^2, \qquad \left.\frac{d^3\Phi}{d\omega^3}\right|_{\omega_0} = -70\ \text{fs}^3.$$

The corresponding pulse intensity envelope and the third-order autocorrelation trace are shown by the solid curves in Figs. 4(c) and 4(b), respectively. The small deviations between the experimental and the calculated traces may be due to higher-order phase terms. It is, however, clear that the remaining spectral cubic phase dominates the pulse characteristics. This analysis results in a pulse duration of 8 fs and a deconvolution factor of 1.35. These values are smaller than those obtained with second-harmonic autocorrelation because of the higher-order nonlinearity [$\chi^{(3)}$] that is responsible for self-diffraction. The pulse energy after compression is 15 μJ, giving rise to a peak power of approximately 2 GW. Owing to the diffraction-limited nature of the waveguide output, these pulses are expected to be focusable to intensities greater than 10^{16} W/cm^2 with $f/3$ optics.

The intensity envelope and the corresponding SD autocorrelation trace of a transform-limited signal with the spectrum shown in Fig. 4(a) are depicted in Figs. 4(e) and 4(d), respectively. The presence of small satellites relates to the modulated structure of the spectrum. Comparison of Figs. 4(c) and 4(e) implies that the compressed 8-fs violet pulses are 1.25 times Fourier limited.

We have demonstrated the generation of sub-10-fs gigawatt peak-power pulses in the violet spectral range for what we believe to be the first time. The performance of the system is currently limited by the loss and dispersion characteristics of the available chirped mirrors, which have been used in this wavelength range for what is to our knowledge the first time. The hollow-waveguide technique employed for SPM is not restricted to the pulse energies and spectral bandwidths demonstrated in this Letter. In fact, we recently achieved spectral broadening to bandwidths in excess of 70 nm near 400 nm with this same setup. Hence progress in short-wavelength chirped-mirror technology holds promise for the generation of intense 5-fs-scale light pulses in the violet–blue spectral range.

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