Compression of high-energy laser pulses below 5 fs

M. Nisoli, S. De Silvestri, and O. Svelto

Centro di Elettronica Quantistica e Strumentazione Elettronica—Consiglio Nazionale delle Ricerche, Dipartimento di Fisica, Politecnico, Piazza L. da Vinci 32, 20133 Milano, Italy

R. Szipöcs and K. Ferencz

Szilárdtestfizikai Kutatóintézet, Pf. 49, H-1525 Budapest, Hungary

Ch. Spielmann, S. Sartania, and F. Krausz

Abteilung Quantenelektronik und Lasertechnik, Technische Universität Wien, Gusshausstrasse 27, A-1040 Wien, Austria

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High-energy 20-fs pulses generated by a Ti:sapphire laser system were spectrally broadened to more than 250 nm by self-phase modulation in a hollow fiber filled with noble gases and subsequently compressed in a broadband high-throughput dispersive system. Pulses as short as 4.5 fs with energy up to 20-μJ were obtained with krypton, while pulses as short as 5 fs with energy up to 70 μJ were obtained with argon. These pulses are, to our knowledge, the shortest generated to date at multigigawatt peak powers.

Spectral broadening of laser pulses by self-phase modulation (SPM) in a single-mode optical fiber followed by chirp compensation in suitable phase-dispersive elements is a well-established technique for pulse shortening. Pulses down to 6 fs were obtained in 1987 from 50-fs pulses of a mode-locked dye laser,[1] and more recently pulses of ~5 fs were generated from 13-fs pulses of a cavity-dumped Ti:sapphire laser.[2] However, the use of single-mode fibers limits the pulse energy in both cases to a few nanojoules. During the past few years great technological advances have occurred in the field of ultrafast-pulse generation, in particular the development of high-energy solid-state femtosecond lasers. Ti:sapphire amplifiers seeded by 10-fs laser oscillators can now generate pulses of ~20 fs with gigawatt[3,4] or terawatt[5,6] peak power at repetition rates in the kilohertz and 10-Hz regimes, respectively. Recently, a powerful pulse-compression technique based on spectral broadening in a hollow fiber filled with noble gases demonstrated the capability of handling high-energy pulses (submillijoule range).[7] This technique presents the advantages of a guiding element with a large-diameter single mode and of a fast nonlinear medium with a high threshold for multiphoton ionization.

In this Letter we show that combination of the hollow-fiber technique with a broadband, high-throughput dispersive system allows the compression of 20-fs pulses down to a duration as short as 4.5 fs in the pulse-energy range of tens of microjoules, corresponding to multigigawatt peak powers. This result, along with the potential scalability of the system to significantly higher pulse energies, is expected to open up new prospects in high-field light–matter interaction.[8]

We carried out the experiments using a Kerr-lens mode-locked mirror-dispersion-controlled Ti:sapphire oscillator, which provides nearly transform-limited 8-fs pulses.[9] These pulses were amplified at a repetition rate of 1 kHz in a multipass amplifier pumped by the second harmonic of a Q-switched Nd:YLF laser.[3] The output pulses have a duration of 20 fs, energy up to 300 μJ, and a spectrum centered at 780 nm. The pulses were almost transform limited. The amplified pulses were coupled into a 160-μm-diameter, 60-cm-long fused-silica hollow fiber. The fiber was kept straight in a V groove made in an aluminum bar that was placed in a high-pressure chamber with fused-silica windows (1 mm thick) coated for broadband antireflection. The hollow fiber was filled with argon or krypton at different pressures. By properly matching the input beam to the EH_{11} mode of the fiber, we measured an overall fiber transmission of 65%, which is close to the value (~73%) predicted by the theory.[10]

The frequency-broadened pulses emerging from the hollow fiber were compressed by a double pass through two pairs of fused-silica prisms of small apex angle (20°) and by two reflections on a broadband chirped mirror for compensation of quadratic as well as cubic phase distortion. The use of thin prisms with a broadband antireflection coating instead of Brewster-angle prisms allows for a smaller propagation length through glass. This results in a smaller amount of material group delay dispersion and, correspondingly, in a reduction of the negative group delay dispersion required by the prism pairs as well as higher-order dispersion terms. A chirped mirror was introduced to compensate for the cubic phase distortion arising from propagation through the hollow fiber and the prism sequence. This mirror provides at each reflection a negative group delay dispersion with a positive cubic and quartic dispersion. The prism-chirped mirror combination ensures control of second- and third-order dispersion over ~120 THz and provides a high transmission efficiency (>80%) in the range 630–1030 nm.[8]

A typical shape of the spectrum measured at the output of the fiber-compressor system for an argon pressure $p = 3.3$ bars and an input peak power $P_0 = 4$ GW is shown in Fig. 1(a). The shape of the

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spectrum is much more uniform than that previously obtained with longer pulses (140 fs), for which a pure SPM spectrum was observed.[7] This indicates that, at this shorter pulse duration, gas dispersion, besides SPM, also plays an important role. The relative weights of SPM and dispersion can be evaluated by use of characteristic parameters such as the nonlinear length L_{NL} and the dispersion length L_D, defined as[11] $L_{\mathrm{NL}} = 1/\gamma P_0$ and $L_D = T_0^2/|\beta_2|$, where T_0 is the half-width (at the $1/e$ intensity point) of the pulse and $\beta_2 = \mathrm{d}^2\beta/\mathrm{d}\omega^2$ is the group-velocity dispersion of the fiber filled with gas. The nonlinear coefficient γ is given by $\gamma = n_2\omega_0/cA_{\mathrm{eff}}$, where n_2 is the nonlinear index coefficient of the gas ($n = n_0 + n_2 I$, where I is the field intensity), ω_0 is the laser central frequency, c is the speed of light in vacuum, and A_{eff} is the mode effective area. For best pulse compression, an optimum fiber length L_{opt} exists, and its value is well approximated by $L_{\mathrm{opt}} \approx (6L_{\mathrm{NL}}L_D)^{1/2}$.[12] Assuming that, for argon, $n_2/p = 9.8 \times 10^{-24}\ \mathrm{m}^2/\mathrm{W\,bars}$ (Ref. 13), one obtains $L_{\mathrm{NL}} \approx 0.92$ cm. On considering the contributions to second-order dispersion from both gas[14] and waveguide,[10] one obtains a value of $\beta_2 \approx 40\ \mathrm{fs}^2/\mathrm{m}$, which gives $L_D \approx 320$ cm. Then, the optimum length turns out to be $L_{\mathrm{opt}} \approx 42$ cm, somewhat shorter than that used in the experiments. However, if one takes into account the peak-power reduction during propagation, which tends to increase L_{NL}, the length of the fiber is not too far from optimum.

We sent the self-phase-modulated output to the compressor and then measured the compressed pulses by an interferometric autocorrelator with silver mirrors and a very thin (15-μm) β-$\mathrm{BaB_2O_4}$ crystal. Figure 1(b) shows the measured second-harmonic interferometric autocorrelation trace obtained when separation between the two couples of prisms was set at 2 m. The chirped mirror was used for folding the prismatic delay line. The time delay in the autocorrelator was calibrated with an He–Ne laser. To evaluate the pulse duration, we took the inverse Fourier transform of the spectrum of Fig. 1(a) and assumed, as a free parameter, some residual cubic phase distortion, $(\mathrm{d}^3\phi/\mathrm{d}\omega^3)$. A good fit to the experimental data was then obtained with a pulse duration (FWHM) of 5.3 fs and $|\mathrm{d}^3\phi/\mathrm{d}\omega^3| = 20\ \mathrm{fs}^3$. The precision of this evaluation is mainly affected by possible errors in the measured spectral shape (the spectrograph was calibrated with a standard tungsten lamp) and in the assumed spectral phase; we expect to introduce errors of less than ±10%. The minimum pulse duration, as calculated on assuming optimum phase-distortion compensation, was 5.2 fs. Therefore the pulses can be considered to be almost transform limited. The results shown in Fig. 1 refer to the case in which output pulse energy from the compressor was 40 μJ, corresponding to an input energy to the fiber of 80 μJ. By increasing the input pulse energy to 140 μJ, we generated pulses as short as 5 fs, with an energy of 70 μJ.

A typical broadened spectrum measured for a krypton pressure $p = 2.1$ bars and an input peak power $P_0 = 2$ GW is shown in Fig. 2(a). Under these conditions L_{NL} and L_D for krypton almost match those for argon, since the increased cubic nonlinearity[13] and dispersion[14] for krypton are balanced by the reduction in input peak power and pressure. The shape of the krypton spectrum is more uniform than that of argon and presents a somewhat greater extension. The different spectral-broadening features of krypton confirm previous observations performed with longer pulse duration (140 fs).[7] By best compression of the pulse whose spectrum is shown in Fig. 2(a), we measured the interferometric autocorrelation trace of Fig. 2(b). From this trace, a pulse duration of 4.5 fs (FWHM) was evaluated, assuming a residual cubic phase distortion $|\mathrm{d}^3\phi/\mathrm{d}\omega^3| = 10\ \mathrm{fs}^3$. Pulse energy after compression was 20 μJ. These pulses represent the shortest generated to date at the tens-of-microjoules energy level. The minimum pulse duration, estimated from the spectrum shown in Fig. 2(b) was 4.3 fs; therefore, the pulses are almost transform limited. On increasing the input peak power to 4 GW and decreasing the pressure to 1.1 bars to maintain constant L_{NL}, slightly longer pulses (5.3 fs) with twice as much energy (40 μJ) were obtained.

The output beam was found to be linearly polarized just like the input beam. We tested the spatial coherence of the beam emerging from the fiber by measuring the transverse profile at different distances from the tip of the fiber. The measured beam profiles were compared with the calculated intensity profiles, assuming free-space propagation of a beam with an initial shape equal to that of the EH_{11} mode of the fiber. A typical result obtained at a given distance from the

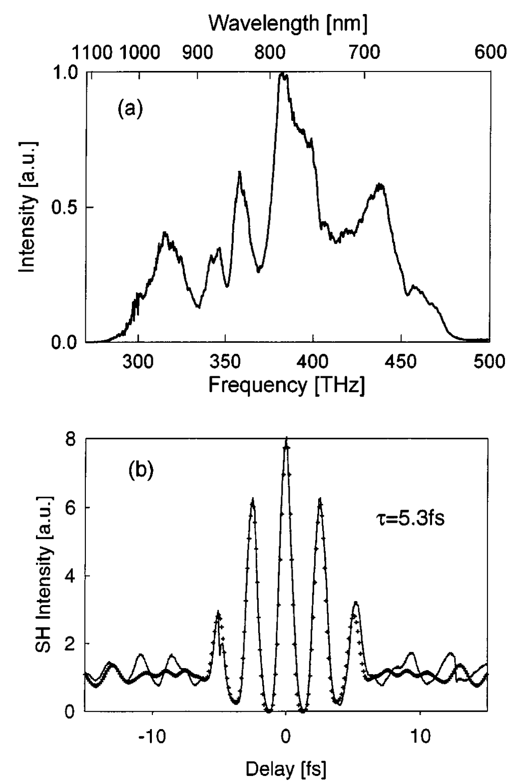

Fig. 1. (a) Spectral broadening in argon at $p = 3.3$ bars and $P_0 = 4$ GW. A low-intensity pedestal (~1% of the peak) extends below 600 nm. (b) Measured (solid curve) and calculated (crosses) autocorrelation trace; an evaluation of pulse duration (FWHM) is also given.

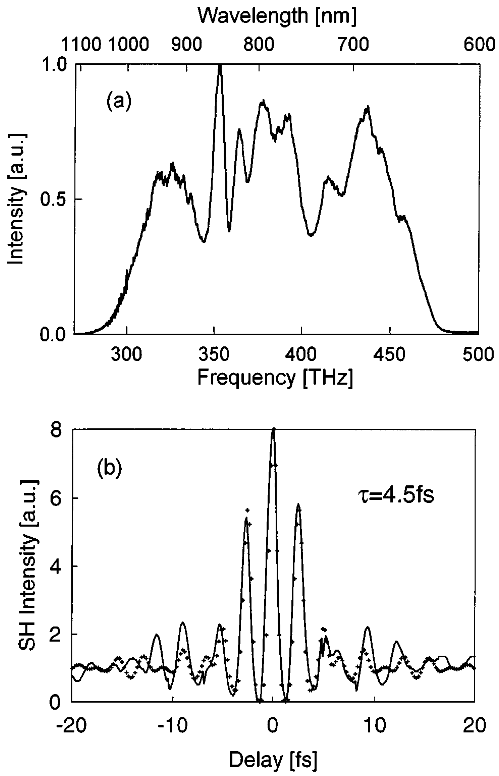

Fig. 2. (a) Spectral broadening in krypton at $p = 2.1$ bars and $P_0 = 2$ GW. A low-intensity pedestal (~1% of the peak) extends below 600 nm. (b) Measured (solid curve) and calculated (crosses) autocorrelation trace; an evaluation of the pulse duration (FWHM) is also given.

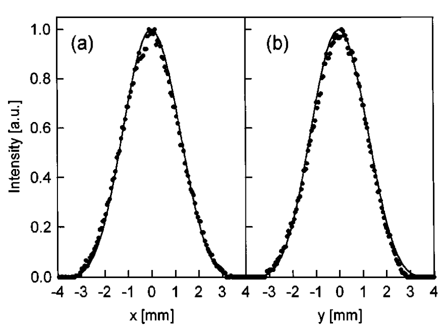

Fig. 3. Measured beam profiles (dots) along orthogonal directions (a) x and (b) y at a distance of 43 cm from the output of fiber. The calculated beam profiles (solid curves) are also shown.

fiber is reported in Fig. 3. Since the measured beam shapes along two orthogonal directions closely follow the calculated curves, the output beam is diffraction limited.

The scalability of the system toward higher pulse energy is an important issue considering the current availability of 20-fs laser pulses with peak powers up to the terawatt level or more. The following two considerations play the most important role: (1) The laser peak power must be smaller than the self-focusing value. This sets a constraint on the type of noble gas to be used and its pressure. (2) The laser peak intensity must be smaller than the multiphoton ionization threshold which applies for the given pulse duration. This represents a constraint on the hollow-fiber diameter and on the type of gas to be used. Since the threshold for multiphoton ionization increases when pulse duration is decreased,[15] we are somewhat confident that the hollow fiber technique can be scaled up to pulse energies of a few millijoules.

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