Generation of 200-μJ, sub-25-fs deep-UV pulses using a noble-gas-filled hollow fiber

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High-energy 110-fs pulses of a KrF excimer laser system were spectrally broadened by self-phase modulation in a neon-filled hollow fiber and subsequently compressed by a grating pair. In this way, 25-fs pulses with energies as high as 200 μJ were generated at 248 nm. The pulses were characterized by an all-reflective single-shot transient grating frequency-resolved optical gating. © 2009 Optical Society of America

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Several fields of contemporary physics and chemistry, such as attosecond pulse generation or time-resolved studies of atoms, molecules, and solids, can benefit from ultrashort deep-UV (DUV) pulses. This is the reason for the great efforts made recently to develop light sources capable of delivering such pulses. Unfortunately, this task is rather challenging, because only a few exotic materials exist that can be used for light amplification in this wavelength range, but their bandwidth does not support pulses shorter than ~100 fs. Therefore the usual approach has been to extend the operation range of standard near-IR Ti:sapphire laser systems to the UV-DUV spectral range by applying ultrabroadband frequency conversion techniques. In this way sub-20-fs pulses could be generated in the DUV [1–7]; however, the pulse energy has been limited to a few microjoules.

Our approach is to apply a spectral broadening technique to moderately short but energetic pulses, which are directly amplified in the DUV. Theoretically much higher energies can be achieved in this way, because it is not the pulse generation but only the spectral extension, which is governed by a low-efficiency third-order nonlinear process. For the spectral broadening, one of the most efficient methods is to use self-phase modulation (SPM) in noble gases filled into a hollow waveguide [8,9]. The feasibility of our approach has been demonstrated by the generation of sub-20-fs pulses with 20 μJ energy at 248 nm by applying the hollow fiber technique to 100-fs output pulses of a dye-excimer laser system [10]. However, it turned out that owing to the short wavelength a number of issues arise that have to be properly addressed, such as low critical power of self-focusing, severe photoionization, high-*f*-number focusing geometry, and high nonlinear absorption of the window materials.

In this Letter we report on the generation and characterization of energetic sub-25-fs pulses at 248 nm, obtained by a hollow-fiber arrangement particularly optimized for UV operation. The obtained pulse energies of as great as 200 μJ are, to the best of our knowledge, 1 order of magnitude higher than any previously reported value obtained for DUV pulses shorter than 100 fs. This allowed us to perform what we believe for the first time single-shot frequency-resolved optical gating (FROG) measurement on sub-100-fs UV pulses [11].

To prevent self-focusing in the fiber at high pulse energies, only helium and neon were considered as nonlinear media, whose nonlinear refractive index is small. To keep the ionization at a tolerable level, a fiber with large bore diameter was chosen, which yielded relative low peak intensity in the fiber. To compensate the effect of lower peak intensity on the spectral broadening, a long fiber was used, which was permitted by the small waveguide losses owing to the large bore diameter and short wavelength. To avoid severe nonlinear absorption in the windows of the gas chamber, the whole focusing/collimating optics was placed into the chamber.

The scheme of the experimental apparatus is shown in Fig. 1. The horizontally polarized output of a hybrid Ti:sapphire-KrF excimer laser system operating at 248.5 nm was compressed by a single-pass grating compressor to a transform-limited duration of 110 fs. An iris diaphragm transmitted the central circular part of the homogeneous, nearly flat-topped beam that was then seeded into the hollow fiber. Because of the short wavelength and the relatively large bore diameter of the hollow fiber, a large-*f*-number focusing geometry ($f^{\#}\approx 550$) was required for launching the laser beam into the fiber. In our arrangement the focusing was accomplished by a

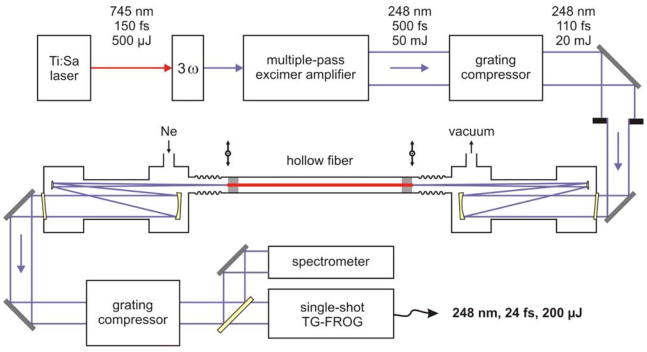

Fig. 1. (Color online) Schematics of the experimental arrangement.

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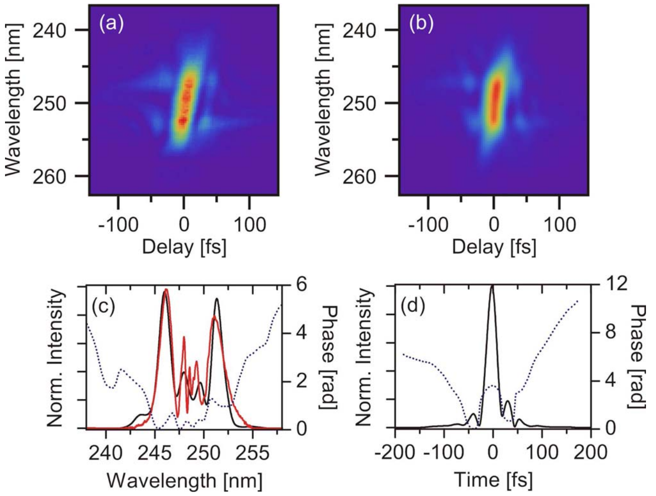

Fig. 2. (Color online) Single-shot FROG measurement of the compressed pulse. The (a) measured and (b) retrieved FROG traces, the retrieved (c) spectral and (d) temporal profiles are shown. The gray curve in (c) displays the spectrum recorded by an external spectrometer.

reflective Galilean telescope consisting of a negative and a positive spherical mirror, whose separation was slightly larger than the sum of their focal lengths. In this way the required $f \approx 6.6$ m effective focusing could be realized in less than 2 m. The astigmatism introduced by the slight tilt of the focusing mirrors was compensated by tilting the last collimator lens of the laser system. The input beam size was adjusted by the iris diaphragm in order to get clean and stable beam profile at the output of the fiber. The optimum was reached by a beam diameter of 12 mm carrying a pulse energy of 2.9 mJ, which resulted in a focal spot size of 245 μm (full width at $1/e^2$ level), which is $\sim$9% larger than the theoretical value of optimal beam launching. This is most probably due to the finite beam pointing stability, which was about ±10% of the bore diameter of the fiber. As a waveguide a 2-m-long flexible hollow fiber of 320 μm inner diameter was used, which was stretched in order to avoid waveguide losses, critically depending on the straightness of the fiber [12]. The fiber assembly separated the gas chamber into two parts, allowing the use of pressure gradient along the fiber [13]. This was important because of the large Rayleigh length of the focusing ($\sim$20 cm), resulting in a light intensity almost as high as in the fiber over an extended range in front of it. This would lead to self-focusing and beam breakup in the gas, preventing the proper launching of the beam into the fiber. Therefore the chamber in front of the fiber was evacuated, and the noble gas was applied at the output of the fiber, resulting in a gas flow and pressure gradient along the fiber. The collimating optics was matched to the divergence of the beam emerging from the waveguide, which is determined by the bore diameter. In our case the half-angle of divergence of 0.84 mrad [12] required an effective focal length of 8.3 m in order to create a 14 mm diameter collimated beam at the output. All mirrors in the focusing/collimating arrangements were coated by a high-damage-threshold high-reflection coating with a bandwidth of $\sim$20 nm centered at the laser wavelength. Both windows of the 6-m-long vacuum setup were made of 5-mm-thick CaF$_2$. Behind the fiber unit the pulses were compressed by a second single-pass compressor consisting of 1200 lines/mm holographic gratings blazed at 250 nm, having an overall transmission of 52%. The compressed pulses were characterized by an all-reflective single-shot transient grating (TG) FROG device [11].

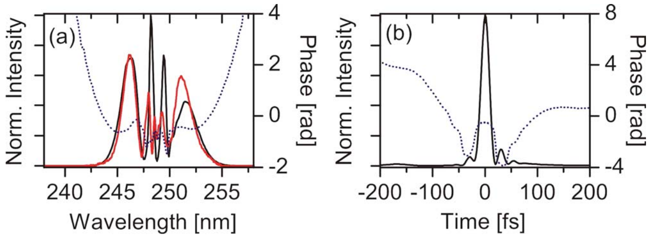

Fig. 3. (Color online) Calculated (a) spectral and (b) temporal profiles. The gray curve in (a) displays the experimentally recorded spectrum.

After testing helium and neon as nonlinear media at different pressures, we found that neon in a 1.7–2 bar pressure range provided the best compromise between spectral bandwidth and pulse energy. Using 2 bar neon at the output side of the fiber the FROG trace shown in Fig. 2 was obtained, yielding a pulse duration of 24.0 fs at a FROG error of 1.1% for the 256 grid size. At this pressure the beam profile was a stable, regular ring system with a clean Gaussian-like core. The pulse energy carried by the central lobe of the profile behind the compressor was 193 μJ corresponding to 371 μJ at the output of the fiber setup, which is only 13% of the input energy. Since the transmission of the evacuated system was $\sim$45%, a loss of 70% was caused by the gas. The loss is attributed to absorption due to multiphoton ionization and to energy leakage from fundamental to higher-order fiber modes due to the defocusing effect of the ionization. It is interesting to note, that in spite of the high absorption, the ionization does not manifest itself in the output spectrum whose shape is characteristic to Kerr-type self-phase modulation [see Fig. 2(c)]. Indeed, by simulating the propagation through the fiber-compressor setup by numerically solving the nonlinear Schrödinger equation, which does not include ionization effects, a remarkably good agreement is achieved between the measured and the calculated spectra, and also between the retrieved pulse shape and the calculated one, for an overall B-integral of about 9 (see Fig. 3).

To understand this result, following [14], let us consider the case when at the entrance of the fiber the light intensity is so high that the ratio of the refractive index changes induced by the Kerr effect and by the ionization is in the range of 5–10. By evaluating the ionization rates according to the general Keldysh [15] or Perelomov–Popov–Terentév theory [16] (which give reliable results even for large values of Keldysh factor, i.e., for low intensity UV light) it turns out, that at the intensities fulfilling the above criterion, the relative ion density and therefore the absorption loss is much higher in the UV than in the IR. Therefore in the UV the intensity will drop faster than in the IR, resulting in a decrease of the ionization-induced phase modulation relative to the Kerr-effect during propagation. Therefore contribution of the ionization to the overall B-integral will be much smaller than that of the Kerr effect.

In conclusion, we generated and characterized 24-fs DUV pulses with energies up to 200 μJ by applying the hollow fiber compression technique to high-energy amplified pulses at 248 nm. It is possible to further increase the energy of the compressed pulse by a factor of 1.5–1.7 by using chirped mirrors instead of the lossy grating compressor, which could also eliminate the slight spatial chirp currently present in the beam. Nevertheless, the current pulse energy is large enough to drive a second stage of spectral broadening, which will enable the generation of few-cycle pulses in the DUV with much higher energies than currently possible.

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