Sub-10-fs supercontinuum radiation generated by filamentation of few-cycle 800 nm pulses in argon

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Focusing 12 fs pulses of 800 nm with moderate energy (0.35 mJ) into atmospheric-pressure argon (Ar) gives rise to filamentation (self-focusing) and a supercontinuum with a very broad pedestal, extending to 250 nm. According to the present understanding, the short wavelengths are produced by self-phase modulation in the self-steepened trailing edge of the pulse. Pulses in this spectral range might thus be intrinsically short. Indeed we demonstrate this by extracting the light near the end of the filament, terminating self-focusing by a pressure gradient at a pinhole, beyond which the Ar is pumped away. We obtain pulses of 9.7 fs in the region of 290 nm without the necessity of compression.

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If intense pulses are injected into optical materials, the intensity-dependent change of the refractive index leads to self-phase modulation (SPM) and broadening of the spectrum. The resulting radiation is chirped. An important application is pulse shortening: At 800 nm one typically uses an inert gas for the medium and subsequently compresses the pulses by chirped mirrors [1]. It has recently become popular to use instead of gas-filled capillaries [2] for this purpose self-guiding by self-focusing in simple cells filled with atmospheric pressure argon, for example [3–6]. (In fact, the pulses of 10–12.5 fs used in this Letter are made in this way [4].) We found that focusing few-cycle pulses again into argon (Ar) gives rise to a supercontinuum spectrum with a very broad pedestal, reaching in the UV to 250 nm and 200 nm with pulses of 10 and 6 fs, respectively [4,7]. Under conditions of single filamentation (which is simply obtained by limiting the input energy to $\leq$0.35 mJ by an iris), this source of radiation has good pulse-to-pulse stability of the spectrum, the resulting energies, peak powers, pulse shapes, and the beam direction. A particularly attractive feature is the fact that all wavelengths are collimated on the axis, in contrast with the supercontinuum from longer pulses, where the short wavelengths were mostly generated in the form of conical emission. The radiation suggests itself for spectroscopic applications, in particular for time-resolved spectroscopy. The energies are even sufficient for use as a pump in such experiments. We already demonstrated this possibility in [8]: The pulses cut out from the supercontinuum at various UV wavelengths had a duration of 30 fs, after traveling over 2.5 m in air and several millimeters in optics and subsequent (re)compression by a prism pair. It was made probable that this duration was limited by higher-order dispersion. Shorter pulses may hence be expected, if the beam path after the filament is evacuated.

SPM causes a positive chirp: In the rising part of the pulse the wavelength is redshifted, turning to a blueshift beyond the pulse maximum. Hence the short wavelengths are produced in the decaying wing of the pulse [9] and this wing can be self-steepened because the pulse maximum travels more slowly than the temporal wings [10]. Due to the steep trailing edge, the spectrum is asymmetrically broadened, extending much more to shorter wavelengths. In principle, under conditions of self-focusing the negative contribution to the refractive index, induced by the increasing plasma density on the axis, can also induce a blueshift. This time-dependent refractive index has recently been invoked to explain pulse self-compression observed (for the inner part of the beam cross section) at higher energies ($\approx$5 mJ) than used by us ($\leq$1 mJ) [11–14]. At lower energies, this contribution seems to be negligible [9]. Under these conditions, the pulses near 800 nm emerging from the filament are typically longer than before entering the nonlinear medium, probably due to their broad spectrum and the continued propagation in the medium. They can be recompressed by available technology (chirped dielectric mirrors). In contrast, this is not easy with tunable short wavelengths. But it may be supposed that compression is not necessary for them: If they arise from the self-steepened trailing edge, they may be intrinsically short. To profit from this effect, one has to avoid lengthening by possible pulse splitting (that may develop later, after the self-steepening [7]) and by dispersion of transmissive materials. In this Letter, we show that this is indeed possible simply by terminating the filament by a pressure gradient. In this way, any dispersive medium is simultaneously avoided after UV generation.

We use a commercial Ti:sapphire laser system (Spectra-Physics Tsunami and Spitfire), emitting pulses at 800 nm with energy of up to 2 mJ, pulse duration of $\approx$45 fs (FWHM), spectral half-width of 30 nm (14 THz) at a repetition frequency of 1 kHz. The pulses are then broadened to $\approx$80 nm by focusing (f=2 m) into 500 mbar of Ar and compressed to 12.5 fs by subsequent reflection from chirped mirrors

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[Fig. 1(a)]. Part of this radiation (≤200 μJ), taken aside by a beam splitter, is used as a probe in a pump–probe setup for cross correlation with the UV beam; additional chirped mirrors are used to compensate the stretching of these pulses by the dispersion in air. This is also done with the other beam. This other part (≈700 μJ) is limited by a diaphragm D (diameter 5 mm, to avoid multiple filamentation) to ≈350 μJ and refocused (f=1 m) into a second Ar cell [Fig. 1(b)]. In this cell (with variable pressure) filamentation is terminated by the pressure drop (e.g., from 1 bar to 8 mbar) at the exit aperture (diameter 0.6 mm). The radiation passes through three more apertures (0.7, 1, and 2 mm), serving for differential pumping, into the evacuated box B, where it is recollimated and focused by dielectric mirrors through another hole (4 mm) to the detection chamber. On the way it passes through the hole of a mirror, merging in this way with the IR probe beam that is focused by this mirror. The two beams ionize Xe and the ion yield is measured in a time-of-flight mass spectrometer as a function of the probe delay. Spectra are recorded, after deflecting the beam out from box B by a movable mirror, by a calibrated spectrometer after integrating over the beam cross section by an Ulbricht sphere.

Figure 2 shows the spectra before (= spectrum of the probe) and after the second cell filled with 950 mbar of Ar. The termination pinhole is placed at a distance of 106 cm from the focusing mirror, i.e., 6 cm after the geometrical focus or 2–3 cm before the normal end of the filament. The inset indicates the spectrum reflected by dielectric mirrors that cut out the UV part for determination of the pulse duration. This cut can be fitted to a Gaussian of maximum 290 nm (4.28 eV) and half-width 32.8 nm (117 THz), which corresponds to a transform-limited duration of 4 fs. From integrating the spectra one obtains an energy of 250 nJ of this cut, which is 7.3×10^{-4} of the

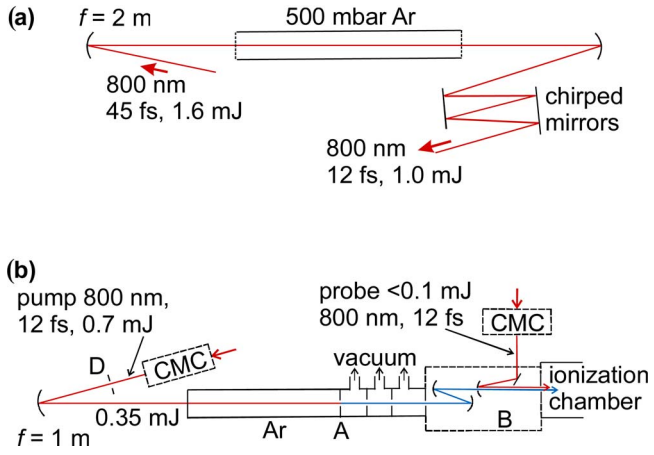

Fig. 1. (Color online) Setup (a) for generation of the 12 fs pulses, (b) for generation of the supercontinuum and the short UV pulses, also indicating the arrangement for measuring the cross correlation between the pulses. CMC is a chirped-mirror compressor as in (a), D is a diaphragm of variable diameter, A is the first of four apertures (needed for differential pumping), and B is an evacuated box with mirrors for collimation, focusing, and merging the beams.

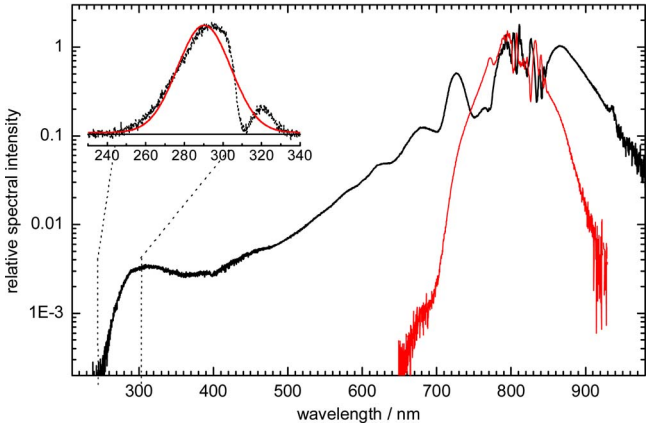

Fig. 2. (Color online) Full spectrum after and before the (second) Ar cell [shown in Fig. 1(b)]. The inset shows the spectrum near 270 nm cut out by the dielectric mirrors (nominal range indicated by dotted lines).

input of 0.35 mJ. In the ionization chamber, we only use ≈30 nJ. The loss is due to the apertures, which are not yet optimized for maximum optical transmission.

Figure 3 shows the cross correlation (Xe^+ yield versus UV-IR delay) of this UV part with the fundamental (IR) pulses (800 nm, 1.55 eV). The latter have a duration of 12.5 fs, determined by a commercial autocorrelator, and an energy of 10–30 μJ (intensity $\approx10^{13}$ W cm^{-2}). The half-width of the Xe^+ signal is 10.9 fs. This corresponds to a UV pulse duration of τ_{UV}=9.7 fs, assuming that ionization is made by 1 UV + 6 IR photons; i.e., that the cross-correlation function contains the first power of the UV pulse and the sixth power of the IR pulse. Indeed experiments with variation of the UV energy confirm that only one UV photon is involved in the ionization.

This corresponds to 2.4 times the transform-limited duration. If the pulse is linearly chirped, this implies a group delay dispersion (GDD) of +12.6 fs^2. (The sign is inferred from pulse lengthening by inserted thin quartz plates of known GDD.) It is conceivable that such a small GDD is due to the dielectric mirrors. A small contribution is also expected from the last few centimeters in the filament (0.82 fs^2/cm in 1 bar Ar).

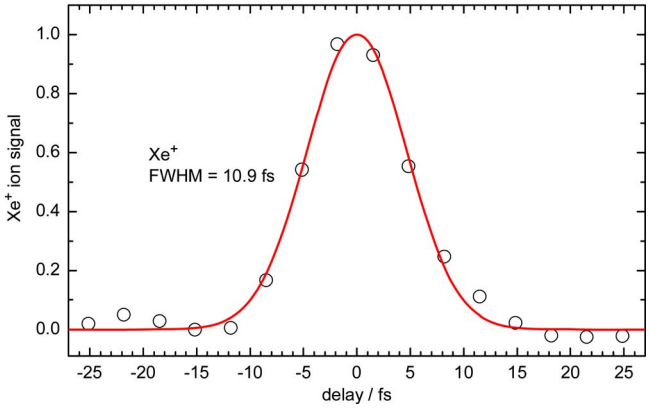

Fig. 3. (Color online) Cross correlation of the UV and IR pulses, resulting from ionization of Xe by 1 UV + 6 IR photons.

To check to what extent the third-harmonic (270 nm) contributes to the supercontinuum, we varied the Ar pressure (Fig. 4). Below 750 mbar a separate maximum (270 nm, half-width 15 nm or 63 THz at ≤500 mbar) can be seen, but is covered at higher pressure by the rapidly growing supercontinuum. Its contribution to the 250–300 nm window can be estimated to be ≤5% with pressures ≥900 mbar, if the termination aperture is at 106 cm from the $f=1$ m mirror (as in the figures). With earlier termination, the isolated third harmonic also persists to higher pressures, and the continuum already ends at longer wavelengths; there is even no detectable supercontinuum but a third-harmonic at up to 1 bar, if the aperture is placed into the geometric focus (100 cm from the mirror) or slightly before (98 cm, which is the waist location of the filament [4]). The increased importance of the third-harmonic as compared with the case without termination is remarkable. Further propagation in Ar hence seems to largely reconvert this harmonic to the fundamental. (This idea also applies to Fig. 3 of [4], where termination is done by a very small pinhole but propagation continues in Ar.) In fact, this has been predicted and confirmed for longer pulses by Aközbek *et al.* [15] and Chin *et al.* [16]. Continued propagation in the nonlinear medium reduces by phase mismatch the third-harmonic power by an order of magnitude and—due to a phase-coupling effect—leaves it constant thereafter.

To conclude: after spectral broadening by SPM in a gas, pulses in the region of 800 nm are usually compressed by chirped mirrors; this also compensates the pulse stretching by the continued propagation in a dispersive medium. We found that pulses from the UV part of the supercontinuum are intrinsically short, shorter than the original pulse, if the filamentation is terminated by a pressure gradient. This confirms the idea that the short wavelengths are generated in the self-steepened trailing edge of the pulse. According to calculations, self-steepening can even lead to an optical shock, i.e., to a decay within 1–3 fs [7], so that even shorter UV pulses can possibly be extracted. Since all the blueshifted part is produced in the trailing edge (the third harmonic is negligible, as stated above), we can expect that all the short-wavelength supercontinuum (or at least any cut from it with similar bandwidth) is intrinsically short, shorter than the pump pulse. With the full spectral width, these pulses would then be far from transform limited. It would be desirable to find out whether they have a regular chirp that would allow further compression. However, the pulses demonstrated here already present an attractive, rugged source of tunable sub-10-fs pump (or probe) pulses for transient spectroscopy; tuning from the deep UV to the visible can be done, e.g., by choosing the appropriate dielectric mirrors. Furthermore it seems probable that shorter pulse durations in the UV can also simply be obtained by using shorter (e.g., 5–6 fs) pump pulses, which can be obtained [3,5,6] by adding a second compression step of the kind in Fig. 1(a). This method of shortening the UV pulses, which only uses available technology, is at present certainly simpler than the development of compression techniques for the UV pulses.

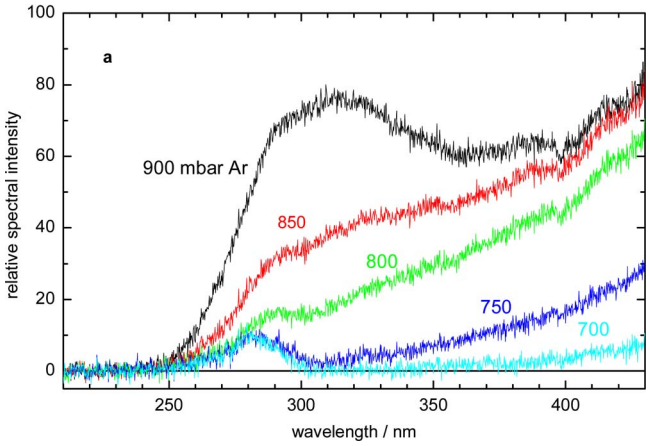

Fig. 4. (Color online) Evolution of the short-wavelength part of the spectra on raising the Ar pressure in the cell with termination of the filament [Fig. 1(b)].

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