Generation of sub-20-fs deep-ultraviolet pulses by using chirped-pulse four-wave mixing in CaF_2 plate

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Sub-20-fs deep ultraviolet (DUV) pulses are generated by using nondegenerate, chirped-pulse four-wave mixing of the fundamental and second-harmonic pulses from a commercial Ti:sapphire amplifier in a CaF_2 plate. The energy of the DUV pulses is 3.8 μJ, with a conversion efficiency from total pump energy to DUV of ~3.8%. The DUV pulse is compressed using a pre-chirp, introduced via a fused silica window in the fundamental beam. The central wavelength of the DUV spectrum can be tuned from 257 to 277 nm by adjusting the cross angle between the two pump beams. The spectrum can reach a width of 16.8 nm, which can support a pulse duration of 8.7 fs.

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Ultrashort pulses, especially those with a few-cycle pulse duration and microjoule-level pulse energy in the deep ultraviolet (DUV) range (200–300 nm), are required when investigating ultrafast dynamics in atoms, molecules, clusters, and polymers [1–3], especially in some human health-related molecules such as DNA, RNA, and proteins, which have a large absorption peak in the range of 240–300 nm, and some ultrafast processes with time constant ~100 fs or even faster [3–5]. There are several possible methods of generating ultrashort DUV pulses, such as the frequency doubling of optical parametric amplifier (OPA) [6], four-wave mixing (FWM) in hollow fibers or noble gas-filled cells [7–9], the spectra broadening of the third-harmonic wave of Ti:sapphire lasers in noble gas [10], and direct frequency upconversion in gas cells [11,12]. Using these methods, the shortest pulse is sub-3-fs with a sub-4 fs/0.25 mJ pump laser [11], and the highest level of pulse energy generated is 900 μJ with a pump energy of ~7 mJ [10]. The extremely short pulse or high energy of the pump lasers makes the method for generating such state-of-the-art results not suitable for common applications, such as pump-probe experiment for investigating ultrafast dynamics in biological molecules.

In this Letter, we generated sub-20-fs DUV pulses in a relatively simple way, using the nondegenerate chirped-pulse FWM of the fundamental and second-harmonic (SH) pulses from a commercial Ti:sapphire amplifier in a CaF_2 plate to generate DUV pulses with energy levels of 3.8 μJ. The conversion efficiency from total pump energy (SH + fundamental) to DUV was ~3.8%. The pulses were compressed by a prechirp, introduced through a fused silica window in the fundamental beam. The central wavelength is tunable from 257 to 277 nm by adjusting the cross angle between the two pump beams. The spectra can reach a width of ~16.8 nm, which can support a pulse duration of 8.7 fs.

Figure 1 shows the schematic of the experimental setup. A commercial femtosecond Ti:sapphire regenerative amplifier (RGA) system (Micra+ Legend-USP, Coherent, 1 kHz/50 fs/2.5 mJ) was used as the pump source, and 300 μJ of the total energy was applied in the experiment. About 200 μJ of the pump energy was focused into a 0.2 mm BBO crystal, cut at $\theta = 29°$ for Type I phase matching for the SH generation. The fundamental and SH pulses were focused into the 0.5 mm CaF_2 crystal using concave mirrors. The cross angle between the fundamental and SH beams was set as ~18.7° (in air), which would fulfill the phase-matching condition for the four-wave mixing process: $\omega_{DUV} = 2\omega_{SH} - \omega_F$, $\boldsymbol{k}_{DUV} = 2\boldsymbol{k}_{SH} - \boldsymbol{k}_F$, where ω_X and $\boldsymbol{k}_X$ denote the angular frequencies and wave vectors, respectively, with X corresponding to DUV, SH, and fundamental (F). A fused silica window (pre-chirp in Fig. 1) with a thickness of 12 mm introduced a dispersion of 430 fs^2 to the fundamental pulse. The group delay dispersion (GDD) of the variable neutral density filter, the half-wave plate, and beam splitter were estimated as 100 fs^2, 70 fs^2, and 50 fs^2, respectively. Other optical components in the experimental setup brought dispersions below 20 fs^2. The pulse from the Ti:sapphire RGA was set to be slightly

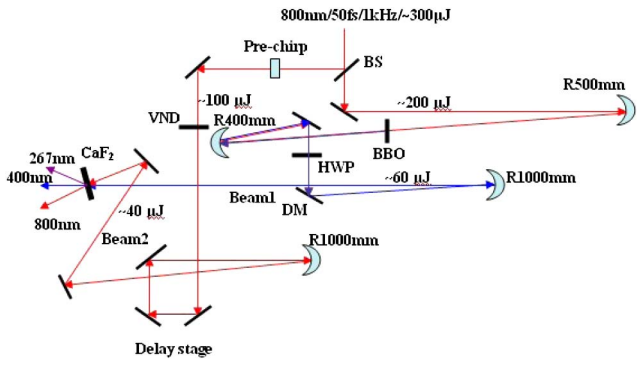

Fig. 1. Schematic of the experimental setup. BS, beam splitter; VND, variable neutral density filter; HWP, half-wave plate; DM, dichroic mirror.

negatively chirped (−100 – 0 fs^2). As a result, the GDDs of the fundamental and SH pulses were ~500 fs^2 and ~100 fs^2, respectively. The DUV pulse was generated by FWM when the two pump pulses were temporally overlapped. The DUV pulse was characterized by a dispersion-free self-diffraction frequency-resolved optical gating (SD-FROG) system with a 100 μm thick sapphire plate for generating the SD signal.

The chirped-pulse FWM process has been used to generate ultrashort visible [13] or DUV [7] pulses in previous researches. The principle of compression of the DUV pulse generated by using chirped FWM in this experiment can be understood as follows. The electric fields of the SH and fundamental beams can be expressed as

$$E_{\mathrm{SH}}(t) \propto \exp\{i\omega_{\mathrm{SH}}t + \phi_{\mathrm{SH}}(t)\}, \quad (1)$$

$$E_F(t) \propto \exp\{i\omega_F t + \phi_F(t)\}. \quad (2)$$

The field of DUV can be written as

$$E_{\mathrm{DUV}}(t) \propto \exp\{i(2\omega_{\mathrm{SH}} - \omega_F)t + 2\phi_{\mathrm{SH}}(t) - \phi_F(t)\}. \quad (3)$$

Then,

$$\begin{aligned}\partial^2\phi_{\mathrm{DUV}}(t)/\partial t^2 &= \partial^2(2\phi_{\mathrm{SH}}(t) - \phi_F(t))/\partial t^2 \\ &= 2\partial^2\phi_{\mathrm{SH}}(t)/\partial t^2 - \partial^2\phi_F(t)/\partial t^2. \quad (4)\end{aligned}$$

Both fundamental and SH pulses are positively chirped in the experiment and have the relation $\partial^2\phi_F(t)/\partial t^2 > 2\partial^2\phi_{\mathrm{SH}}(t)/\partial t^2 > 0$. As a result, $\partial^2\phi_{\mathrm{DUV}}(t)/\partial t^2 < 0$ is satisfied, which means the DUV pulse is negatively chirped. This negative chirp would compensate for the dispersion of air from the output end to the sapphire plate in SD-FROG, and the sapphire plate itself.

The pulse energy of the DUV pulse was measured as 3.8 μJ with the pump energy of 60 μJ for the SH beam and 40 μJ for the fundamental beam. The output energy became saturated according to the energy measurement with different pump levels, and the CaF_2 crystal would be damaged by heat within several minutes of running at this pump energy rate. The energy saturation may be due to the two-photon absorption of the DUV pulse in the CaF_2 crystal. To enable it to run over a long period, the energy of both pump pulses was set as 40 μJ, resulting in a DUV pulse energy of ~3.1 μJ. By adjusting the cross angle between the fundamental and the SH beams, the spectra [Fig. 2(a)] of DUV pulses were tunable, with a tunable range of the central wavelength from 257 to 277 nm, and a full spectra range from 255 to 295 nm. The extension of this spectral range is limited by the spectra width of the two pump pulses. The spectrum of the DUV pulse with the highest energy [the green line in Fig. 2(a)] was centered at 265 nm, with a width of ~5.7 nm. The widest spectrum, ~16.8 nm, was obtained for the cross angle of ~16.6° [Fig. 2(a)], which was quite different from the phase-matching angle (~18.7°) for the FWM process of the center wavelengths (800 and 400 nm) of the two pump pulses. This small cross angle enabled the red part of the SH spectrum and the blue part of the fundamental spectrum, such as 405 and 765 nm, to fulfill the phase-matching condition and, as a result,

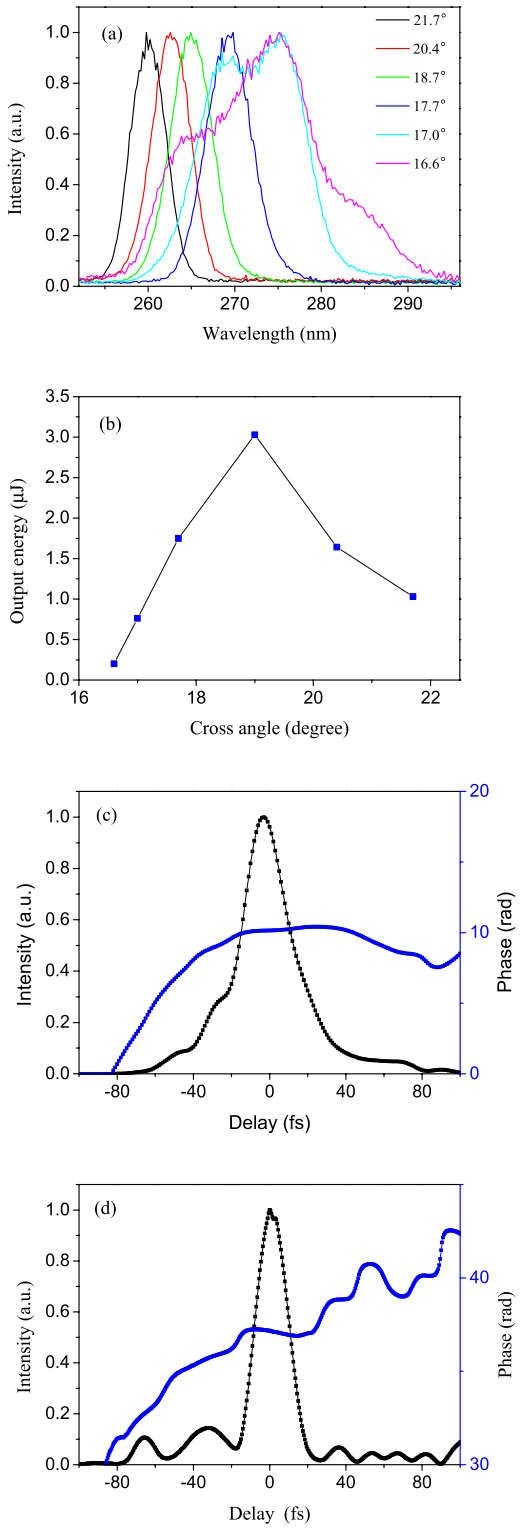

Fig. 2. Spectral and temporal characteristics of the generated DUV pulse: (a) spectra and (b) pulse energy of the DUV pulse with the different cross angles of the two pump beams. Retrieved intensity profiles of (c) 28.9 fs and (d) 18.7 fs DUV pulses, corresponding to the spectra in green (18.7° cross angle) and cyan (17.0° cross angle) in (a), respectively.

extended the red part of the DUV spectrum (275.4 nm was generated due to an FWM of 405 and 765 nm). This widest spectrum can support pulse duration of ~8.7 fs, calculated by the inverse Fourier transform of the

spectrum. The DUV pulse with the widest spectrum was not recorded because of its relatively low energy. The pulse energy was reduced by 60% along the path from the output surface to the sapphire plate for SD signal generation in the SD-FROG, due to the low reflectivity of the aluminum mirrors used in the experiment. As shown in Fig. 2(b), the pulse energy is >200 nJ for DUV pulses with different central wavelengths and spectra shapes [Fig. 2(a)]. The vibration of the DUV energy is <2% during the measurement. The pulse energy is sensitive to the cross angle of the two pump beams due to the phase-matching conditions for the FWM. It is possible to achieve a DUV pulse with a broad spectrum and with high energy if a pre-angle-chirp is introduced to the pump beams, enabling most of the spectra of the two pump pulses to fulfill the phase-matching conditions in the FWM process.

Figures 2(c) and 2(d) show the retrieved temporal profile and phase of the DUV pulses with the cross angles of the two pump beams at ~18.7° and ~17.0°, respectively. The measured pulse width is 28.9 fs for the DUV pulse with a spectral width of 5.7 nm and a pulse energy of 3.1 μJ [the green line in Fig. 2(a)]. The DUV pulse with the spectral width of 13.4 nm and pulse energy of 0.76 μJ [cyan line in Fig. 2(a)] has a width of 18.7 fs. The FROG errors in the two measurements were smaller than 0.01. The pulse was optimized by adjusting the distance between the output surface and the sapphire plate in the SD-FROG, which brought unneglected positive dispersion to the DUV pulse [7]. The dispersion of the pump laser was also slightly adjusted. The weaker basal and parasitic pulses near the main pulses in Figs. 2(c) and 2(d) are induced by the high-order dispersion from the pump laser pulse, which has a similar temporal profile to the DUV pulse. Angular dispersion induced in the FWM process also prevents the compression of DUV pulses. Proper pre-angular chirp of the pump pulses may reduce the angular chirp, as used in a previous experiment for OPA pulse generation [14].

The generation of sub-20-fs DUV pulses using nondegenerate chirped-pulse FWM of the fundamental and SH pulses, produced by a commercial Ti:sapphire amplifier in a bulk material appears more compact and cheaper than the frequency doubling of OPA [6] or FWM with OPA [15] because of the high cost and large setup for an OPA system. It also consumes less pump energy and has a higher conversion efficiency than FWM in hollow fiber or noble gas-filled cells [7–9]. The pump/output energy is <0.3 mJ/3.8 μJ in this Letter, and 1.2 mJ/0.3 μJ, 0.8 mJ/1 μJ, and 2 mJ/2 μJ in Refs. 7 through 9, respectively. The DUV spectrum generated is also wide enough to support a pulse width of sub-10-fs. The method used in this Letter may make the DUV laser with sub-20-fs pulse duration and microjoule-level energy a commonly usable laser source, and furthermore it may help to promote the investigation of ultrafast dynamics in biological molecules and basic organic molecules. There are several problems with this method, such as the angular dispersion of the DUV pulse, but this may be solved by careful adjustment of the pre-angular chirp of the pump beams.

In conclusion, sub-20-fs DUV pulses are generated using nondegenerate, chirped-pulse FWM of the fundamental and SH pulses, from a commercial Ti:sapphire amplifier in bulk material. The pulse energy reaches 3.8 μJ, and the central wavelength and spectral width are tunable. The width of the widest spectrum is 16.8 nm, which supports pulse duration of 8.7 fs. This simple and low-cost way allows generating several-microjoule, sub-20-fs DUV pulses. In future studies, we will focus on the sub-10-fs DUV pulse generation by introducing a pre-angular chirp to pump beams and carefully managing the GDD and high-order dispersion.

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