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Generation of sub-17fs vacuum ultraviolet pulses at 133nm using cascaded four-wave mixing through filamentation in Ne.

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The sixth harmonic ($6\omega$, 133 nm) of a Ti:sapphire laser is generated using cascaded four-wave mixing in filamentation propagation through rare gases. The method provides the $6\omega$ pulse energy higher than 5 nJ/pulse at 1 kHz and a pulse duration shorter than 17 fs without dispersion compensation. © 2014 Optical Society of America

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collinearly with precision higher than 1 mrad in our setup. The gas cell has an aluminum pinhole plate 0.5 mm in thickness as the exit for laser pulses. Comparing with our previous study [9], we reduced the thickness of the plate by a factor of three, as laser-drilling of a thinner plate provides a three times larger pinhole diameter (ca. 0.3 mmφ), which results in higher transmission of the laser pulses. The gas cell is connected to a multistage differential pumping system, in which the laser-drilled pinhole, a narrow channel (1.25 mm in diameter and 20 mm in length), and an aperture (3 mmφ) restrict the gas conductance between the cell and an adjacent optics chamber maintained at ~10⁻² Torr [9]. The primary beams of ω and 2ω and the generated 3ω, 4ω, 5ω, and 6ω beams propagate collinearly and pass through the differential pumping system to enter the optics chamber.

Filamentation propagation is induced when an intense laser pulse self-focuses and ionizes a rare gas to create weak plasma. If we neglect optical influence of the plasma, the phase matching angle for noncollinear FWM of 2ω + 2ω − ω → 3ω is predicted to be 1 mrad using the Sellmeier equation for neutral neon gas [11]. As can be seen in Fig. 1, similar calculations predict the phase matching angles for cascaded processes to generate 6ω to be from 1 to 4 mrad. We align the input laser beams collinearly with the precision higher than 1 mrad, so that the noncollinear phase matching is not operative in our experiment.

Figure 2 shows a setup for measurements of the pulse energy and the spectrum of 6ω. In pump–probe experiments, we separate the central and peripheral parts of the output beam using a flat UV-enhanced aluminum mirror with a 3 or 4 mm diameter hole [9]. However, in the measurement of the 6ω pulse energy, we reflected the entire output beam using five dichroic mirrors for 6ω (the specified reflectivity R > 85% for unpolarized light from 130 to 138 nm, Layertec) and sent the purified 6ω beam to a detector chamber housing a calibrated Si photodiode (SXUV, IRD) under vacuum. This optical layout effectively blocks the stray light from the optics chamber; however, a residual ω pulse propagating coaxially with 6ω creates a background signal at the detector. We evaluate this background signal by introducing air (~30 Torr) into the optical path to eliminate 6ω. The true 6ω pulse energy is determined by subtracting the pulse energy of ω from the energy measured under vacuum. The 6ω pulse is not observed when the input ω beam is blocked, indicating that the direct third harmonics generation, 2ω + 2ω + 2ω → 6ω, is negligible.

Figure 2 shows the 6ω pulse energy as a function of Ne gas pressure in the cell. For comparison, the pressure dependence of the 3ω, 4ω, and 5ω pulse energies are also shown in arbitrary units. The 6ω pulse energy rapidly increases with the Ne gas pressure and exhibits its maximum (6 nJ/pulse) at around 200 Torr. Assuming 85% reflectivity of the mirror specified by the manufacturer, the 6ω pulse energy in the cell is estimated to be more than 10 nJ, corresponding to a conversion efficiency of ca. 2 × 10⁻⁴ with respect to the 2ω input pulse energy. The spatial profile of the 6ω pulse examined using a Ce:YAG phosphor plate was round below the gas pressures of ca. 200 Torr.

The 6ω pulse energy declines for Ne gas pressures higher than 200 Torr, even though the 3ω and 4ω pulse energies further increase. Qualitatively, the conversion efficiency increases with gas pressure, while it diminishes due to phase mismatch (Δk) at higher pressures when plasma created in filamentation propagation is neglected. Plasma dispersion will reduce phase mismatch, keeping conversion efficiency higher for low-order har-

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**Fig. 1.** Noncollinear angles expected for cascaded FWM processes to generate 6ω in neutral gas. The calculations were performed using Sellmeier equation given in [11] and the neon gas pressure of 200 Torr.

**Fig. 2.** Schematic diagram of our setup for measurements of the pulse energy and the spectrum of 6ω. D.M. denotes dichroic mirror.

**Fig. 3.** Pressure dependence of the measured pulse energies for 6ω (solid circles). Pressure dependencies of the pulse energies for 3ω (diamonds), 4ω (squares), and 5ω (open circles) reported in [9] are also shown for reference.
The spectrum of the 6ω pulse measured using a VUV monochromator (VM-502, Acton Research) is shown in Fig. 5. The spectrometer has been calibrated using a deuterium lamp. The resolution is estimated as 0.2 nm from a full width at half-maximum (FWHM) of the Lyman-α line (121.5 nm). The 6ω spectrum exhibits a nearly Gaussian profile with a center wavelength at 133 nm. The 5ω component centered at 159 nm is absent, indicating that it is completely attenuated by the dichroic mirrors for 6ω. The observed spectrum shown in Fig. 5 supports a transform-limited pulse width of 6 fs.

Figure 6 shows the cross-correlation trace between the 4ω and the 6ω pulses measured using nonresonant (1 + 1’) two-photon ionization of krypton; the background (time-independent) signals are due to one-color two-photon ionization of 6ω pulses. Contribution of one-color three-photon ionization of krypton with 4ω pulses was negligible. As mentioned earlier, a flat UV-enhanced aluminum mirror with a 3 mm diameter hole is employed for the pump–probe measurements to separate the central and the peripheral parts of the output beams. The central part transmitted through the hole is reflected using five 6ω mirrors and focused by a concave 6ω mirror (r = −1,000 mm) onto an atomic beam of Kr. The peripheral part is reflected using four 4ω mirrors and focused by a concave aluminum mirror (r = −1,000 mm) with a UV-enhanced coating onto the Kr beam in overlap with 6ω. No dispersion compensation is made for 4ω nor 6ω. The Kr⁺ photoion signal is measured using a microchannel plate detector and a single photon counter (SR400, Stanford Research Systems). The observed trace is fitted by a single Gaussian with a FWHM of 17 fs as shown in a solid line in Fig. 6. The FWHM of the cross-correlation is twice larger than 9.2 fs expected from the bandwidths of the 6ω and 4ω pulses. The pulse durations of the harmonics may be shortened by spectral phase transfer technique [12, 13].

In conclusion, we have demonstrated sub-17 fs 6ω pulse generation with a sufficient pulse energy for pump–probe photoelectron spectroscopy. Our filamentation FWM light source can generate simultaneously 3ω, 4ω, 5ω, and 6ω at 1 kHz, enabling a variety of pump + probe schemes such as 3ω + 4ω, 3ω + 5ω, and 3ω + 6ω, etc. Different wavelengths are easily selected using dichroic mirrors. Recently, Shi et al. [14] employed...
$2\omega$ and $3\omega$ of a high-power Ti:sapphire multipass amplifier ($\sim$50 mJ/pulse at 10 Hz) and generated $4\omega$, $6\omega$, $7\omega$ (114 nm), $8\omega$ (100 nm), and $9\omega$ (89 nm) using FWM in Ar. Generation of higher harmonics than the sixth order would also be possible with a 1 kHz system.

References