Tunable sub-40 fs ultraviolet pulses from non-collinear four-wave mixing in CaF_2 crystal

Yongliang Jiang[1,2*], Baozhen Zhao[1,2], Keiichi Sueda[2],
Noriaki Miyanaga[2], and T. Kobayashi[1,2,3,4]

[1] *International Cooperative Research Project, Japan Science and Technology Agency,*
4-1-8 Honcho, Kawaguchi, Saitama 332-0012, Japan
[2] *Institute of Laser Engineering, Osaka University, Yamadakami 2-6, Suita 565-0871,*
Ibaraki 567-0047, Japan
[3] *Department of Applied Physics and Chemistry and Institute for Laser Science, University of*
Electro-Communications, Chofugaoka 1-5-1, Chofu, Tokyo 182-8585, Japan
[4] *Department of Electrophysics, National Chiao Tung University, 1001 Ta Hsueh*
Road, Hsinchu 300, Taiwan
[*] *Corresponding author: jiangyl@ile.osaka-u.ac.jp*

Received December 15, 2010; accepted June 29, 2011; posted online June 29, 2011

Pulses with a tunable central wavelength from 278–310 nm reaching <27 fs duration and >1 μJ energy are generated by non-collinear four-wave mixing in a 0.5-mm CaF_2 crystal.

OCIS codes: 140.3610, 190.4223, 320.7110.
doi: 10.3788/COL201109.S10604.

The development of femtochemistry in recent years has enabled us to study and even control the ultrafast dynamics of molecules. Ultrashort ultraviolet (UV) laser sources are essentially needed for the study of dynamics and reactions in various organic molecules of biological importance, such as amino acids, proteins, and DNA, which have UV transitions. The well-established optical parametric amplification technique is not suitable for this region because of the high group-velocity mismatch and the strong absorption of the pump, etc. Many efforts have been made to generate ultrashort UV pulses with frequency up-conversion techniques, such as sum frequency mixing and second harmonic generation (SHG)[1,2]. Another effective method is ultrashort four-wave mixing (FWM) in noble gases[3,4], which usually requires a complicated vacuum system. Ultrafast non-collinear FWM in transparent bulk media is comparatively a compact method, which also has the advantage of tunable output wavelength. This method has been adopted to generate sub-20 fs visible pulses[5,6]. In this letter, we demonstrate the generation of tunable sub-40 fs UV pulses by non-collinear FWM in an isotropic CaF_2 crystal pumped with 120-fs pulses at 393 nm.

As shown in Fig. 1, part of the energy from a commercial 1-kHz chirped pulse amphifier (CPA) pulse laser with 120 fs duration and 1.6 mJ energy at 785 nm was selected and focused on a sapphire crystal to generate a broadband white light seed. After spectral shaping with a filter and temporal shaping with a grating pair, the seed was amplified by a two-stage nonlinear optical power amplifier NOPA system in the spectral region of 500–800 nm. A prism pair was adopted to compress the amplified broadband pulses; the resulting pulse duration was ~10 fs with energy of 7.5 μJ. The few-cycle visible pulses were focused on a 0.5-mm-thick CaF_2 crystal pumped with SHG of another part of the energy from the CPA laser. A concave mirror with a focus length of 10 cm was used to collimate the generated UV energy ($\omega_{\text{UV}} = \omega_{393\text{ nm}} + \omega_{393\text{ nm}} - \omega_{\text{NOPA}}$).

The output UV spectra are shown in Fig. 2(a). The angle between the 393 nm pump laser and the few-cycle visible laser was changed to 8.3°–15.3°, and the corresponding output central wavelength changed from 310 to 278 nm. The full-width at half-maximum (FWHM) of the output spectral bandwidth decreased from 8 to 3 nm with the increase in the phase match angle, which can support sub-20 fs pulses for the long wavelength side and sub-40 fs for the short wavelength side.

The output energies for different central wavelengths are analyzed in Fig. 2(b). The highest output pulse energy of >1 μJ can be achieved with 120 μJ pump energy when the central wavelength is around 300 nm. The low conversion efficiency is mainly caused by two reasons. First, there is a temporal mismatch between the 120 fs pump pulse and the few-cycle seed. Second, only a small part of the broadband seed energy is involved in the FWM process because of the limited phase matching angle.

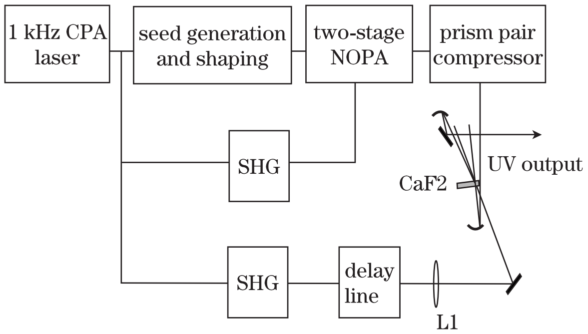

Fig. 1. Schematic of the non-collinear FWM system.

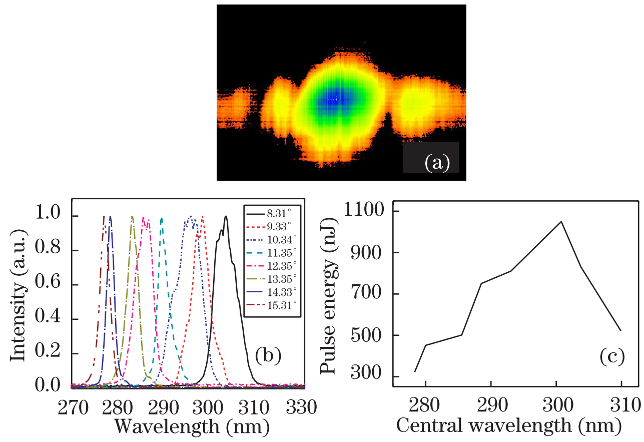

Fig. 2. (a) XFROG trace for 26.7 fs pulses centered at 301 nm; (b) output UV spectra with different match angles, and (c) output pulse energies for different central wavelengths measured in the CaF_2 crystal.

The output UV pulse duration was measured with a homemade DFG-XFROG, where a fraction of the energy from the CPA laser was used as reference. The output pulse width can be optimized by adjusting the insert depth of the prism pair, and no additional UV region compressor is required. Pulse duration of 26.7 fs was observed with the central wavelength of 301 nm with a slightly negative chirped broadband visible pulse, whereas 35.5 fs was observed for the output laser pulse centered at 286 nm.

In conclusion, we have generated sub-40 fs UV pulses with non-collinear FWM in a 0.5-mm-thick CaF_2 crystal without additional compressor. The output central wavelength can be tuned from 278 to 310 nm.

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