High-energy picosecond near-vacuum ultraviolet pulses generated by sum-frequency mixing of an amplified Ti:sapphire laser

Jiangfeng Zhu,[1] Weijun Ling,[2] Zhaohua Wang,[1] Peng Wang,[1] Jinghua Sun,[1] Zhiyi Wei,[1,*] Dacheng Zhang,[3] Xinwen Ma,[3] and Wenlong Zhan[3]

[1]Institute of Physics, Chinese Academy of Sciences, Beijing National Laboratory for Condensed Matter Physics, Beijing, 100080, China

[2]College of Mathematics Physics and Information Science, Tianshui Normal University, Tianshui, 741001, China

[3]Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China

*Corresponding author: wzhy@aphy.iphy.ac.cn

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We demonstrate high-energy picosecond near-vacuum ultraviolet laser pulse generation. Frequency quadrupling is achieved by noncollinear sum-frequency mixing of the fundamental and the third harmonic of a two-stage Ti:sapphire amplifier in β-BaB_2O_4 crystal. UV pulses with energies of ~10 mJ tunable from 195 to 210 nm at a 10 Hz repetition rate are obtained.

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1. Introduction

Intense laser pulses in the UV and vacuum-ultraviolet (VUV) spectral range are of great interest in laser resonant ionization of heavy ions, micromaching, photolithography, photobiology, and time-resolved spectroscopy [1,2]. However, there are only a few excimer lasers that can produce fundamental wavelengths in this spectral range, for example, ArF (193 nm), KrF (248 nm), XeCl (308 nm), and XeF (351 nm), but all these lasers work at a discrete wavelength without tunability. Recently, with the rapid progress of Ti:sapphire laser technology, newly developed Ti:sapphire amplifier systems based on chirped-pulse amplification techniques have turned out to be reliable laser sources for the generation of picosecond and femtosecond pulses widely tunable in the visible and near-infrared spectral range. Phase-matched nonlinear frequency conversions including second-harmonic generation (SHG) and sum-frequency mixing (SFM) are promising methods for the conversion of ultrashort pulses from visible and near-infrared to blue and UV, even VUV. The development of highly efficient nonlinear optical crystals especially for the UV region makes these nonlinear frequency conversions a reality. Of these crystals, β-BaB_2O_4 (BBO) [3] and LiB_3O_5 (LBO) [4] are of great importance for producing UV pulses due to their excellent optical and mechanical quality, high nonlinear coefficient, and broad transparent range down to less than 200 nm. The combination of the well-established Ti:sapphire laser systems and the outstanding nonlinear crystals should produce reliable, compact, tunable near-VUV laser pulses. Nanosecond laser systems, which are based on coherent mixing of the fundamental, second, and third harmonic radiation of a Ti:sapphire laser in BBO crystal can produce tunable wavelengths down to 188.5 nm [5,6]. This is just the transparency cutoff of BBO crystal (189–3500 nm at room temperature). Shorter wavelength generation at 186.0 nm is demonstrated by cooling BBO to 91 K, with the absorption spectra for BBO at a lower limit of 187 nm at this temperature [7]. Because LBO extends its transparency range to as low as 160 nm, it is a promising choice for the nonlinear frequency conversion process in the wavelength range below 189 nm. Seifert *et al.* [8] demonstrated femtosecond VUV pulses in the

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range of 172.7–187 nm by SFM of the Ti:sapphire's fourth harmonic and a parametrically generated infrared pulse in LBO.

However, the energy of UV and VUV in most previous experiments is of the order of nanojoules [8] and microjoules [9] due to the high repetition rate of the pump Ti:sapphire amplifier, typically a kilohertz regenerative amplifier that can deliver millijoule-level pulse energy. This greatly limits the application in many fields that need larger energy. In this paper, we present high-energy SHG and sequential harmonic generation (SFM of a fundamental and its second and third harmonics) of amplified Ti:sapphire laser pulses with a pulse duration of 60 ps. Efficient generation of tunable near-VUV (190–210 nm) pulses is carried out in BBO crystal arranged in a noncollinear SFM scheme. Single pulses with energy higher than 100 mJ at the second harmonic (SH) and tens of millijoules at the third and fourth harmonics (TH and FH) are achieved when the fundamental Ti:sapphire laser delivers pulse energy up to 360 mJ.

2. Experimental Setup and Results

The experimental setup is schematically shown in Fig. 1. The picosecond fundamental pulses are produced by a Ti:sapphire laser system including a picosecond mode-locked oscillator, a regenerative preamplifier, and a multipass master amplifier. The oscillator is a picosecond version of a Tsunami laser (Spectra–Physics, Mountain View, California, USA) that delivers average power as high as 1 W (4.5 W, 532 nm pumping) at a 80 MHz repetition rate and a pulse duration of 60 ps. Wavelength tunability from 715 to 855 nm is achieved by adjusting the birefringent filter in the cavity. After a pulse picker with a Pockels cell, the selected pulses are synchronized with the pumping lasers and amplified to 3 mJ in a Ti:sapphire regenerative amplifier pumped by a 60 mJ, 532 nm Nd:YAG laser at a 10 Hz repetition rate (Quanta Ray, Spectra–Physics). Further amplification is carried out by a multipass Ti:sapphire amplifier that is pumped by two Nd:YAG lasers (Quanta Ray, Spectra–Physics). After six passes through the amplifier, amplified pulse energy as high as 600 mJ is available with 1.6 J of pump energy, which corresponds to an extraction efficiency of 37.5%. Unfortunately, the laser beam quality is rather bad due to the self-focusing effect and wavefront aberration from the thermal effect in the Ti:sapphire crystal. The bad beam quality remarkably reduces the following frequency conversion efficiency. Therefore a compromise between the achievable pulse energy and beam quality is made by a five-pass configuration using a vacuum spatial filter in the third pass. This setup delivers a maximum energy of ~360 mJ and with good beam quality.

The nonlinear frequency conversion begins with frequency doubling of the fundamental pulses. LBO crystal is the best crystal for doubling the frequency to near 800 nm because it has large angle acceptance and small walk-off, and better beam quality can be achieved from this crystal. Moreover, since the pump energy in our experiment is large, the nonlinear crystal must have a high damage threshold. LBO meets this requirement very well. In our experiment, the LBO crystal can sustain pulse intensity up to at least 10 GW/cm^2 at a pulse duration of ~60 ps. The fundamental pulses have a diameter of 9 mm, which corresponds to an intensity of 9 GW/cm^2, which is sufficient to double the frequency, so the laser beam is directed through the LBO crystal without focusing. The LBO crystal in the experiment is cut for type I noncritical phase matching in the X–Y plane ($\theta = 90°, \varphi = 31.7°$) with a size of 10 mm × 10 mm × 7 mm. Both sides of the crystal are antireflection coated for central wavelengths of 800 and 400 nm. The crystal is mounted on a precise rotation stage and phase matching is achieved by rotating the horizontal angle of the crystal. Figure 2 shows the SHG efficiency versus the incident intensity of the fundamental pulse. SH energy as high as 135 mJ is obtained with a pump energy of 360 mJ, which corresponds to a conversion efficiency of 37.5%. We can see from Fig. 2 that the SHG efficiency has a tendency toward saturation. Efficiency up to 40% may be achieved when using a higher pump fundamental energy.

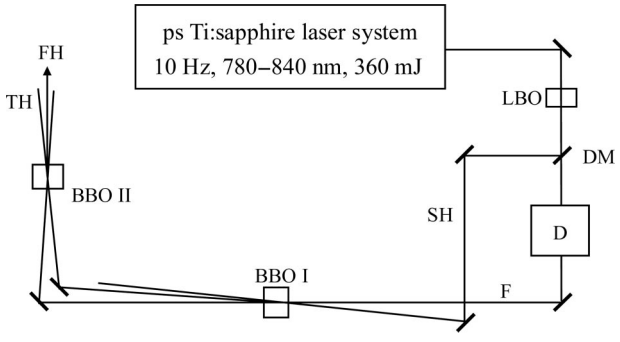

Fig. 1. Experimental layout of the Ti:sapphire amplifier and three-stage harmonic generators. F, fundamental; SH, TH, and FH, second-, third-, and fourth-harmonic generation, respectively; DM, dichroic mirror; D, delay line.

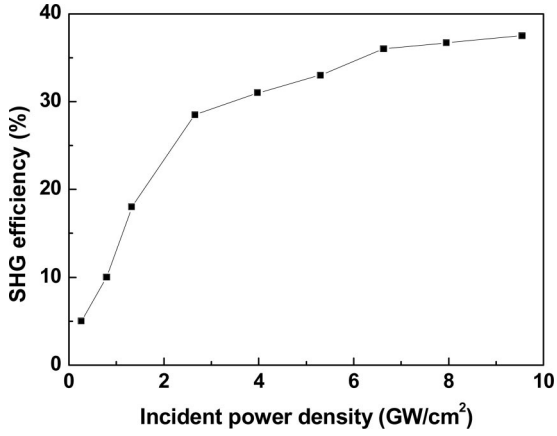

Fig. 2. SHG conversion efficiency versus incident laser intensity.

Direct frequency doubling of the SH pulses from the Ti:sapphire laser by BBO crystal is only available down to 205 nm, at which wavelength the essential nonlinearity vanishes. Wavelengths less than 205 nm by direct SHG are available by using another notable nonlinear crystal $KBe_2BO_3F_2$ (KBBF) produced by Chen *et al.* [10,11], which has a high birefringence and a transparency range down to 155 nm. However, because of its platelike nature, it is difficult to grow a KBBF crystal thicker than 1 mm, greatly limiting its applications in VUV optics. To solve these problems, sequential SFM of the fundamental wavelength and its harmonics of the Ti:sapphire laser by BBO is proposed. It allows phase matching down to the transparency cutoff wavelength of the BBO crystal and with a larger nonlinearity.

The 400 nm SH pulses and the residual fundamental pulses are separated from a dichroic coating mirror. After a variable delay, the two beams are noncollinearly injected into a BBO crystal (type II phase matching: $\theta = 55.5°$, $\varphi = 30°$; dimensions: 10 m $\times$ 10 m $\times$ 9 mm; protect coating) for frequency tripling. The noncollinear phase-matching scheme provides automatic separation of the generated output from the input parent beams. It is unnecessary to use any dispersing optical elements or dichroic mirrors to separate these beams. It is particularly advantageous that dielectric coating in the UV region is much more difficult and the damage threshold is lower than that in the visible and infrared regions. We have compared collinear TH and FH generation with noncollinear schemes. To our disappointment, the conversion efficiency with collinear phase matching is much lower than that of noncollinear phase matching, probably resulting from the difficulty of multilayer UV coating. In our experiment the noncollinear angle between two incident beams is about 6°. The generated TH UV radiation is combined with the fundamental and injected into the second BBO crystal to generate FH radiation. The BBO crystal here has the same dimensions as the former one but is cut at $\theta = 64.8°$, $\varphi = 0°$ for type I phase matching. With the maximum Ti:sapphire laser pumping energy, TH pulses with an energy of 40 mJ and FH pulses with an energy of 10 mJ are achieved, except for the pulse wavelength below 195 nm.

Wavelength tuning is achieved by finely adjusting the birefringent filter in the oscillator and the phase-matching angles of these crystals in Fig. 1. Because the Ti:sapphire laser amplifier system supports only wavelengths between 780 and 840 nm, the generated TH and FH are in the range of 260–280 and 195–210 nm, respectively. In principle, phase-matched SFM down to the transparency cutoff of BBO crystal (188 nm) is possible. Further effort in extending our Ti:sapphire amplifier to wavelengths below 750 nm may reach the VUV cutoff of BBO crystal. Figure 3 shows the FH conversion efficiency as a function of wavelength with the same maximum pumping energy. The conversion efficiency is defined as $\eta = E(4\omega)[E(\omega)E(3\omega)]^{-1/2}$, where $E(\omega)$, $E(3\omega)$, and $E(4\omega)$ are the pulse energy of the fundamental, TH, and FH, respectively. We have achieved conversion efficiency as high as 26.2% at 210 nm with a maximum energy of 16 mJ at the maximum pump energy. The minimum energy we got is 6 mJ at 195 nm. The reason for decreasing efficiency at shorter wavelengths is both the decrease in the effective nonlinear coefficient and the strong absorption of oxygen (4-0 and 5-0 vibronic components of the Schumann–Runge O_2 band) [9].

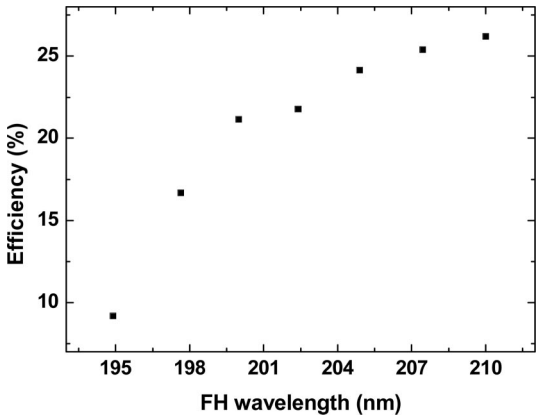

Fig. 3. Conversion efficiency as a function of FH wavelength.

3. Conclusion

In conclusion, by using a noncollinear phase-matching scheme in BBO crystals, we have successfully achieved high-energy picosecond near-VUV radiation tunable from 195 to 210 nm by sum-frequency mixing of the fundamental and the third harmonic of Ti:sapphire laser pulses. With 360 mJ of fundamental pumping energy, SH pulse energy over 135 mJ, TH pulse energy up to 40 mJ, and FH pulse energy as high as 16 mJ are achieved. To our knowledge, this is the highest pulse energy obtained from a picosecond Ti:sapphire laser amplifier in the near-VUV. The use of a noncollinear phase-matching scheme eliminates the difficulty of multilayer UV coating and benefits from accessible conversion efficiency and automatic separation of different beams. The combination of the state-of-the-art Ti:sapphire laser amplifier systems and the highly efficient nonlinear frequency conversion scheme can produce high-energy tunable near-VUV laser pulses for many applications where high peak power and relatively narrow linewidth are required.

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