A picosecond widely tunable deep-ultraviolet laser for angle-resolved photoemission spectroscopy*

Zhang Feng-Feng(张丰丰)[a)c)], Yang Feng(杨 峰)[a)], Zhang Shen-Jin(张申金)[a)†], Xu Zhi(徐 志)[a)c)], Wang Zhi-Min(王志敏)[a)], Xu Feng-Liang(许凤良)[a)], Peng Qin-Jun(彭钦军)[a)], Zhang Jing-Yuan(张景园)[a)], Wang Xiao-Yang(王晓洋)[b)], Chen Chuang-Tian(陈创天)[b)], and Xu Zu-Yan(许祖彦)[a)]

[a)] *Research Center for Laser Physics and Technology, Key Laboratory of Functional Crystal and Laser Technology, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing* 100190, *China*

[b)] *Beijing Center for Crystal R & D, Key Laboratory of Functional Crystal and Laser Technology, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing* 100190, *China*

[c)] *Graduate University of Chinese Academy of Sciences, Beijing* 100190, *China*

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We develop a picosecond widely tunable laser in a deep-ultraviolet region from 175 nm to 210 nm, generated by two stages of frequency doubling of a 80-MHz mode-locked picosecond Ti:sapphire laser. A β-BaB_2O_4 walk-off compensation configuration and a $KBe_2BO_3F_2$ prism-coupled device are adopted for the generation of second harmonic and fourth harmonics, respectively. The highest power is 3.72 mW at 193 nm, and the fluctuation at 2.85 mW in 130 min is less than $\pm 2\%$.

Keywords: tunable laser, deep ultraviolet, walk-off compensation, KBBF crystal

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1. Introduction

Deep-ultraviolet (DUV) lasers are very useful for laser spectroscopy, Raman spectroscopy, angle-resolved photoemission spectroscopy (ARPES), and so on.[1–6] Due to the merits of high energy resolution, bulk sensitivity, etc., DUV laser-based ARPESs have been fabricated and attracted the attention of scientists, what is more, some new phenomena have been observed.[7–9] So far, the reported DUV solid state lasers for ARPESs[3–5] are at fixed wavelengths, which restricts the capability of ARPESs for studying the electronic structure normal to the layer. An ARPES equipped with a tunable DUV laser makes it possible to clarify the final state effect of the photoelectron,[10,11] the overall band structure and the three-dimensional dispersion curve in the momentum space,[12] so a tunable DUV laser is more preferable for ARPES.

For the generation of tunable DUV laser, frequency doubling in nonlinear optical crystal is a simple and convenient method which is important for practical applications. Compared with other nonlinear optical crystals, such as β-BaB_2O_4 (BBO) and LBO, the $KBe_2BO_3F_2$ (KBBF) crystal has demonstrated the potential for producing tunable DUV lasers by using the frequency doubling method.[13–17] These previous reports, however, focused on the fourth harmonic generation (FHG) of the nanosecond (ns) and femtosecond (fs) light sources. The disadvantage of an ns DUV source is the low repetition rate, while the bandwidth of a fs DUV source is sometimes too broad for some applications. For the applications of a DUV laser in ARPES, a narrow bandwidth is crucial for reducing the space charge effect and improving the energy resolution of ARPES so that the continuous wave (CW) or quasi-continuous wave (QCW) lasers are often used.[5,18,19] The repetition rate of ps lasers can be as high as $\sim$ 100 MHz, which is higher than that of an ns laser ($\sim$ kHz); on the other hand, the bandwidth of a ps laser is narrower than that of an fs laser. In view of these two advantages, a ps tunable DUV light source with high repetition rate is more desirable for the ARPES, and high energy resolution and low space charge effect can be achieved.

In this paper, we present a ps DUV laser tunable from 175 nm to 210 nm produced by the FHG from a ps 80-MHz Ti:sapphire laser using a BBO walk-off compensation (WOC) configuration for second harmonic generation (SHG) and a KBBF prism-coupled device (KBBF-PCD) for FHG. The maximum output power of 3.72 mW is obtained at 193 nm, and the average power is more than 0.15 mW from 175 nm to 210 nm by adopting a KBBF thickness of 1.45 mm. The fluctuation of output power at 2.85 mW of 193 nm in 130 min is less than $\pm 2\%$. To our knowledge, it is the first time that the ps tunable DUV laser has been developed for ARPES.

2. Experimental setup

The schematic layout shown in Fig. 1 is used for the generation of the ps tunable DUV laser. The fundamental laser is a linearly polarized, mode-locked Ti:sapphire laser with a tun-

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†Corresponding author. E-mail: zhangshenjin@163.com

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able range from 700 nm to 840 nm, pulse width of < 2 ps [full width at half maximum (FWHM)], repetition rate of 80 MHz (Tsunami, Spectra-Physics). The output powers in the whole range exceed 2 W, and the highest output power can be 2.8 W at 772 nm.

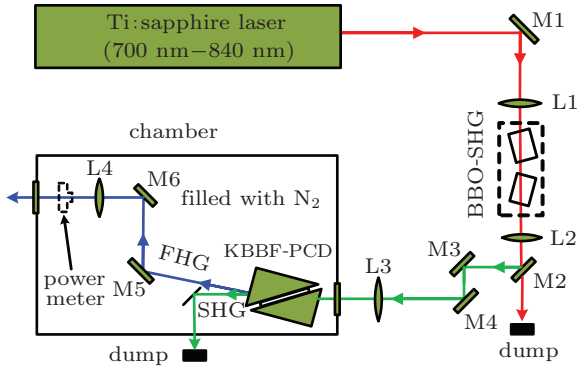


Fig. 1. (color online) Schematic layout of ps tunable DUV laser.

Firstly, the SHG from the Ti:sapphire laser was used to produce tunable light from 350 nm to 420 nm. In order to obtain a better output of second harmonic, a WOC configuration composed of two type-I phase-matched BBO crystals was adopted. The dimensions of both BBO crystals were 8 mm×4 mm×6 mm with the 8 mm side used as the tuning side. The BBO crystals were cut at $\theta = 30.8°$, which is approximately a mean value of the calculated phase-matching angles of 33.7° and 27.9° for frequency doubling at 700 nm and 840 nm. The fundamental laser was focused onto the WOC configuration by a lens (L1) with a focal length of 200 mm, and the focus spot located in the middle of the two closer BBO crystals. Then, the generated tunable output from 350 nm to 420 nm was collimated by a lens (L2) with a focal length of 200 mm, and separated from the pump light by three dichroic splitting mirrors (M2, M3, M4). The output power of this tunable light was measured by a power meter (30A-SH, Ophir. Inc).

Then, the tunable second harmonic (SH) light was focused into a KBBF prism-coupled device (KBBF-PCD) through a plane-convex lens (L3) with a focal length of 300 mm, and type-I phase-matching condition was satisfied for the generation of the tunable DUV light by frequency-doubling. The KBBF crystal was sandwiched between an SiO_2 prism and a CaF_2 prism, and the apex angle of each prism was 60°, corresponding to the phase matching angle of frequency doubling from 374 nm to 187 nm under the condition of normal incidence. The dimension of the KBBF crystal was 6 mm×20 mm×1.45 mm (where 1.45 mm was the thickness along the z axis). The KBBF-PCD was held on a rotator with an angular resolution of < 0.2 mrad to adjust its phase-matching angles for different wavelengths. Due to the strong absorption of DUV light in air (especially below 185 nm), the generation and propagation of the tunable DUV light were enclosed in a hermetical chamber filled with pure N_2 ($> 99.999\%$) at 1 atm (1 atm = 1.01325×10^5 Pa). Two mirrors (M5, M6) were used for aligning the generated tunable DUV laser, and a beam dump was used for collecting the residual radiation from 350 nm to 420 nm. Finally, the diverging tunable DUV light was collimated by a CaF_2 lens (L4) with a focal length of 1000 mm. The output power of the tunable DUV laser source was measured by a power meter (LP-3A, Physcience Opto-Electronics).

3. Results and discussion

By adopting the BBO WOC configuration, an SHG highest power of ~ 1.1 W is obtained at the fundamental wavelength of 772 nm, corresponding to a conversion efficiency of 40%. Moreover, the beam quality of SHG is improved and the walking of the beam direction due to frequency-tuning can be compensated, which is beneficial to the FHG. At 386-nm output power of 1.1 W, the typical two-dimensional profile of SHG beam is measured by a beam quality analyzer (M^2–200, Spiricon Inc.) as shown in Fig. 2. The beam quality factor M^2 values of the horizontal axis and the vertical axis are $M_x^2 = 1.80$ and $M_y^2 = 1.10$, respectively. It can be seen that the beam quality in the y direction is better than that in the x direction. The main reason is the difference in acceptance angle for frequency doubling between the x direction and the y direction and the imperfect compensation of walk off.

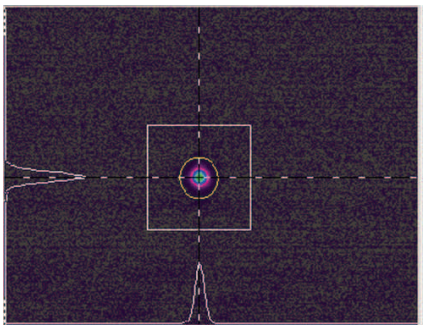


Fig. 2. (color online) Two-dimensional profiles of laser beam at 386 nm.

Figure 3 shows the output powers of SHG and FHG in the whole tunable range. It can be seen that the output powers of the FHG exceed 0.15 mW in the whole tuning range from 175 nm to 210 nm, and the highest power of FHG is 3.72 mW at 193 nm when the pump power is 1.1 W at 386 nm. The lower FHG output powers in the range at longer and shorter wavelengths with respect to 193 nm are mainly attributed to the lower powers of the fundamental and SH light. Besides, at shorter wavelength than 193 nm, the higher absorption losses in KBBF crystal and CaF_2 also result in the reduction of FHG output power.

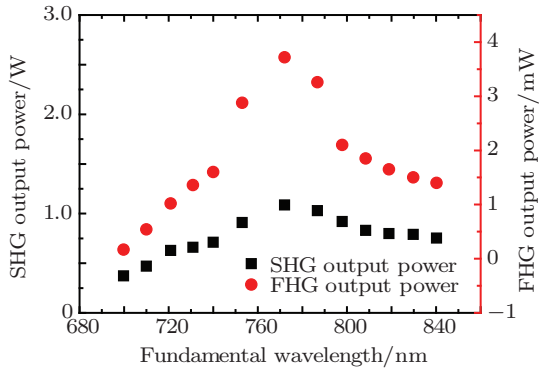


Fig. 3. (color online) Output powers of SHG and FHG each as a function of fundamental wavelength.

In the experiment, the FHG conversion efficiency at 193 nm is about 0.34%, which is lower than the reported results.[20–23] The lower SH peak power density and thinner KBBF crystal are the main reasons for the low FHG conversion efficiency. Therefore, the FHG conversion efficiency can be enhanced by increasing the SH power density through reducing the radius of SH beam in KBBF crystal and adopting the thicker KBBF crystal. It is expected that higher FHG output power can be achieved.

We also test the power stability of the SHG at an output power of 0.96 W and FHG at 2.85 mW in 130 min as shown in Fig. 4. The fluctuations of the output power at 386 nm and 193 nm are less than ±1% and ±2%, respectively.

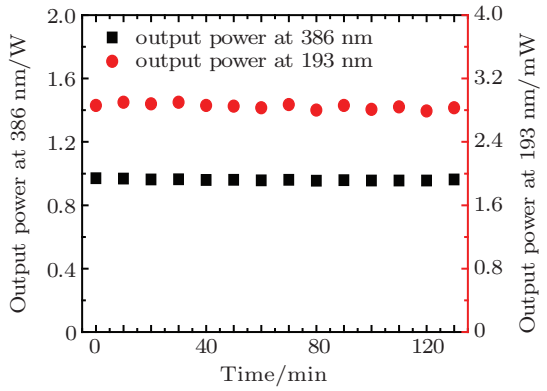


Fig. 4. (color online) Stability measurements at 386 nm and 193 nm, respectively.

4. Conclusion

In this paper, we demonstrate an all-solid-state 80-MHz ps tunable DUV laser with a tuning range from 175 nm to 210 nm. The maximum output power is 3.72 mW at 193 nm and more than 0.15 mW in the whole tuning range. The output fluctuation of the DUV laser is less than ±2% in 130 min. With the ps DUV laser, a photon energy tunable ARPES with high resolution is developed, and it will be an important complement for the former laser-based ARPES.[24] The ps tunable DUV laser-based ARPES will be a more powerful tool for probing the electronic structures of superconductors, topological insulators, semiconductors, and other materials with low work function.

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