CHINESE OPTICS LETTERS
Vol. 22, No. 5 | May 2024

Generation of 160 nm vacuum ultraviolet light at 82 MHz in a KBBF crystal by the fifth-harmonic of a Ti:sapphire laser

Yun Zhang (张 赟)[1*] and Lirong Wang (汪丽蓉)[2,3]
[1] Department of Engineering Science, The University of Electro-Communications, Tokyo 182-8585, Japan
[2] State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, Taiyuan 030006, China
[3] Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan 030006, China

*Corresponding author: yunzhang@uec.ac.jp

Received November 7, 2023 | Accepted January 18, 2024 | Posted Online May 17, 2024

We report the generation of quasi-cw vacuum ultraviolet (VUV) light at 160 nm with a repetition rate of 82 MHz by two second-harmonic generations and one sum frequency mixing. The VUV laser light is produced as a fifth-harmonic generation of a mode-locked ps Ti:sapphire laser system by successive stages with nonlinear crystals of LBO and KBBF. A stable generation of laser light at 200 nm for more than 6 h is the most important step for obtaining the generation of light at a wavelength of 160 nm.

Keywords: vacuum ultraviolet light; second harmonic generation; sum frequency mixing.
DOI: 10.3788/COL202422.051901

1. Introduction

Coherent radiation in deep ultraviolet (DUV) and the vacuum ultraviolet (VUV) spectral range below 200 nm is of great benefit to both basic science and industrial applications such as angle-resolved photoemission spectroscopy, photolithography, and semiconductor metrology[1-3]. There are several techniques to produce VUV light: synchrotron radiation, gas-discharge sources, excimer lasers, free-electron lasers, and frequency conversion[4-6]. All solid-state VUV lasers generated through frequency conversion, which includes sum frequency mixing (SFM) and second-harmonic generation (SHG), possess many advantages such as high photon flux, narrow bandwidth, good beam quality, and high compactness compared with other VUV sources. However, the generated shortest wavelength strongly relies on the nonlinear crystal. For a long time, the shortest wavelength of SHG has been restricted to the range above 205 nm. With the emergence of the crystal material potassium fluoro-beryllo-borate (KBBF) and rubidium fluoro-beryllo-borate (RBBF), direct SHG to wavelengths below 200 nm has become possible[7,8]. Unfortunately, these crystals have a plate-like form along the z axis, and it is very difficult to grow them thick enough to be cut at the angles needed in phase matching for frequency conversion. A special KBBF prism-coupled device (PCD) makes it possible to realize the phase-matching condition for producing VUV light through frequency conversation[9]. Since the first all-solid state VUV lasers were obtained in KBBF-PCD, serials of VUV lasers based on KBBF-PCD have been reported with a pulse train and continuous wave[10-12]. Recently, lots of remarkable achievements were reported in the frequency doubling of laser light to below 200 nm using KBBF-PCD. For instance, watt-level average power has been obtained with a tunable range of 185–200 nm at a low repetition rate[13]. Meanwhile, a high-average power out in the wide range of 170–232 nm was also realized by fourth-harmonic generation of a high repetition rate Ti:sapphire laser[14]. However, it is a great challenge to generate the radiation below 170 nm due to the fact that the transmission dramatically decreases and the absorption loss strongly increases with decreasing wavelength. In fact, only a few works have been reported on the generation of VUV below 170 nm.

The wavelength can be further shortened when high harmonic generation is employed. Usually, gas nonlinear media are adopted. Recently, it has been extensively employed to generate the frequency comb with intracavity enhancement or extracavity enhancement structures[15-17]. For example, a frequency comb with a wavelength of below 100 nm is obtained by high harmonic generation in Xe gas media. Harmonics up to the 19th order have been observed[18]. A compact and robust system can be realized by high-order harmonic generation of a fiber laser[19,20].

In frequency conversion, the convenient way is SHG or cascaded SHG. This is because, in SHG, only one input light beam is required. However, the phase matching condition limits the generated light wavelength. Based on Sellmeier equations, it was predicted that VUV light at a wavelength of 161 nm is limited by direct SHG with KBBF, and 167 nm light is produced by eighth-harmonic generation[21,22]. To produce shorter wavelength light with higher photon energy, the SFG becomes important[23,24]. In SFG, two beams with different wavelengths are required, and both spatial matching and temporal matching between two beams also become necessary. Although several experiments have demonstrated that generation of a wavelength below 150 nm is possible by SFG, the generation of such light with a high repetition rate is necessary to achieve high resolution and high signal-to-noise ratio in the application of photoelectron spectroscopy[25,26]. It is also required for investigating dispersion relations of refractive indices of KBBF at the short wavelength side.

In this paper, we report the generation of quasi-cw VUV light at 160 nm with a repetition rate of 82 MHz by cascaded SHG and one SFG. The 160 nm laser light is produced as a fifth-harmonic generation of a mode-locked ps Ti:sapphire laser system. The two SHGs provide 7.5 mW output at 200 nm with a stability of more than 6 hours. We check the generated 160 nm light with a lock-in amplifier technique. Several nW with a photon number per longitudinal mode per second of about 10^5 is estimated using the measured fifth-harmonic signal.

2. Experimental Setup

The schematic of our experimental setup is shown in Fig. 1. The fundamental light source is a mode-locked Ti:sapphire laser (Spectra-Physics, Tsunami) with a maximum power of 1.4 W, a pulse duration of 2.0 ps, a wavelength of 800 nm, and a repetition rate of 82 MHz, which is pumped by a frequency-doubled $Nd:YVO_4$ laser (Coherent VerdiV10). The output beam of a Ti:sapphire laser was split by a beam splitter (70:30). The 70% fundamental light power was used for the fourth-harmonic generation (FHG), and the rest was employed for the fifth-harmonic generation. For generating the second-harmonic light (SH, 2ω), we employed a cavity enhanced SHG, in which an LBO crystal is chosen as nonlinear media because of its low dispersion and small walk-off angle. In the first stage SHG, more than 600 mW of output at 400 nm was obtained with an input power of 1.2 W. The details of our first stage have been reported in our previous papers[27,28]. In the second stage SHG, we generated the laser light at 200 nm by single-pass configuration SHG, in which a KBBF-PCD is employed. The polarization of light is rotated to meet the phase-matching condition with a half-wave plate (not shown in Fig. 1). The KBBF-PCD consists of a KBBF with a thickness of 2.3 mm and a pair of fused-silica prisms with an apex angle of 55 degrees. In the third stage, an SFG is used to produce fifth-harmonic light (5ω) with fundamental light (ω) and the generated fourth-harmonic light (4ω). A KBBF-PCD, which is placed in a vacuum chamber to avoid absorption of VUV radiation by oxygen gas, was employed. When the fundamental wavelength is 800 nm, the type I phase-matching angle of the SFG process in KBBF crystal is estimated to be around 57 degrees[29,30].

Based on the previous reports, the generated VUV light at 160 nm with power of several nW was estimated[25,30]. To detect such a low signal with high signal-to-noise ratio, a lock-in amplifier is employed. To do this, a chopper wheel is inserted in the fundamental beam to modulate it. For the detector, a phototube (Hamamatsu R1187, response spectrum 115–200 nm) is used. The lock-in amplifier measured only modulated signals of produced light at a wavelength of 160 nm. The SFM occurs only when the fundamental pulse train and fourth-harmonic pulse train simultaneously arrive at the KBBF crystal. Thus, a movable prism was employed for temporally delaying the fundamental light, and the output of the lock-in amplifier was recorded when the prism was linearly scanned with a motor. To check the spatial and temporal mode-matching between the fundamental light and fourth-harmonic light, we check the interference between two SH lights, in which one is the unconverted SH from KBBF and the other one is generated by fundamental light with another LBO crystal inserted in the fundamental light beam. A visibility of more than 60% was observed.

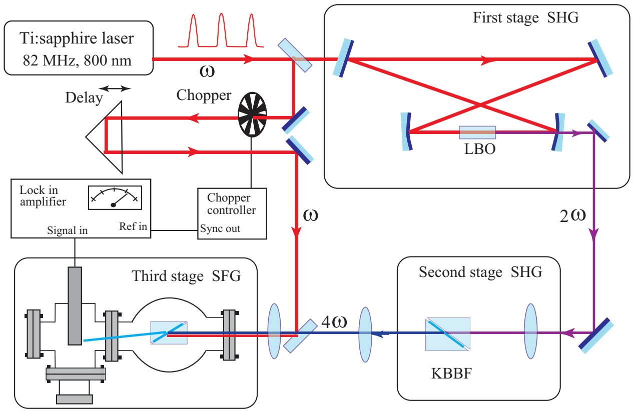

Fig. 1. Schematic of experimental setup.

3. Stable Generation of Fourth Harmonics at 200 nm

For generation of light at 200 nm, a single-pass configuration SHG is employed. The phase-matching angle of KBBF in type-I SHG depends on the fundamental wavelength. To obtain a perfect phase-matching angle, the KBBF PCD was mounted on a rotating stage. Hence, the incident angle can be adjusted by rotating the stage. The generated DUV at 200 nm and fundamental UV wave can be separated through the rear prism based on the different refractive indices. A thermal power meter was used to measure the power of the produced 200 nm light. Figure 2 gives the measured output powers for 200 nm at different input powers of 400 nm. A maximum output power of about 21 mW was obtained at the input power of 580 mW. This corresponds to a conversion efficiency of 3.6%. The curve in Fig. 2 was well fitted by $P_{\mathrm{out}} = E_{\mathrm{NL}} P_{\mathrm{in}}^2$ with an $E_{\mathrm{NL}} = 0.06\ \mathrm{W}^{-1}$. This E_{NL} is much better than the previous experimental reports and indicates that an optimal performance is obtained[27]. It also shows that the output power can be further improved by increasing the input power. Unfortunately, the high output power was hard to maintain, and it dropped significantly as the pumping

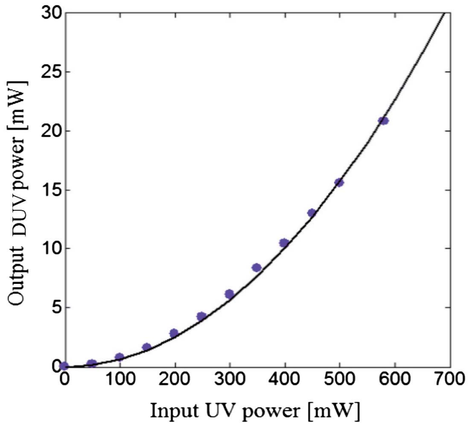

Fig. 2. FH output power by a single-pass frequency doubling configuration with KBBF-PCD. The filled circles are experimental data, and the curve is best fit with $P_{\mathrm{out}} = E_{\mathrm{NL}}P_{\mathrm{in}}^2$, giving $E_{\mathrm{NL}} = 0.06$.

continued increment. This phenomenon was also observed in our previous reports[27,28]. Hence, stable operation of the system is more attractive.

Recently, stable operation of the frequency conversion system with KBBF-PCD was extensively investigated, and some reasons of instability were summarized. One reason considered is a heating up of crystal, which induces phase mismatching by generated harmonic light. Usually, it can be avoided by actively controlling the temperature of crystal. Unfortunately, no significant improvement was achieved even when the temperature of crystal was controlled for the pulse laser[31–33]. But temperature control is very important when a continuous wave laser light is employed for frequency conversion[12]. Another reason considered is the destruction of crystal by strong input and output light. Hence, the system was desired to operate at low input power, and some experimental results show that a stability of over 1 h was obtained with a low input power[33,34]. It was also demonstrated that another effective method to obtain stable operation is using a thinner KBBF[32]. All these methods are at the cost of conversion efficiency. It seems more challenging to keep the high power and high stability operation of the system. To obtain a long-term stable operation and a reasonable power for sum frequency mixing, the input power of 400 nm light was reduced to 400 mW. Figure 3 gives one case of our long-term operation of FH generation for single-pass configuration KBBF-PCD. It indicates an average power of 7.5 mW (corresponding to a conversion efficiency of 1.9%) and peak-to-peak fluctuations of 4.8% in 6 h. To our best knowledge, this is the most stable operation for generation of VUV light. This stability is convenient for the next step of generation of 160 nm light.

4. Generation Vacuum Ultraviolet Light at 160 nm

The generated 4ω beam is recollimated after the KBBF-PCD and combined with the remaining fundamental beam (about 350 mW) to be focused onto another KBBF-PCD, which is placed in a vacuum chamber by a lens with a focal length

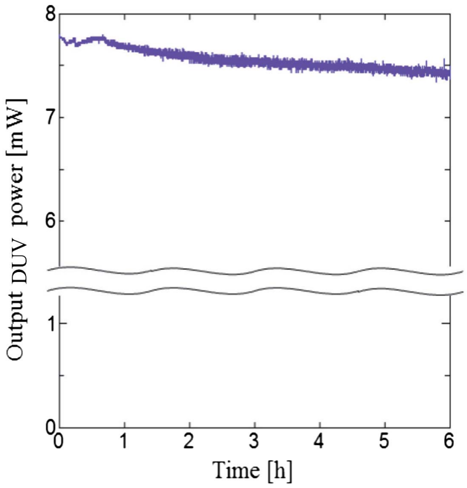

Fig. 3. Long-term measurement of the generated FH power at an incident UV power of 400 mW.

of 200 mm. The chamber keeps a pressure of 10^{-3} Pa during the experiment. To detect the fifth-harmonic signal, the output of the lock-in amplifier was recorded when the delay of the fundamental beam is scanned. A clear peak, which indicates the generation of 5ω, is observed as shown in Fig. 4. The peak has a full width at half-maximum of about 600 μm corresponding to the pulse width of fundamental light. The angle of the KBBF crystal and focusing of the incident beam were optimized by maximizing the output signal. The maximum generated power at 160 nm is estimated to be about 2 nW using the provided values of the phototube sensitivity. The conversion efficiency seems rather poor compared to recent work[30] since a higher repetition rate laser is employed in the present experiment. The power of generation may be improved by employing low repetition lasers, which have a strong peak power. This is most probably due to the absorption in the KBBF crystal, which strongly depends on the quality of the crystal at the low wavelength range. Further improvement is expected by employing a KBBF crystal with the new method growth.

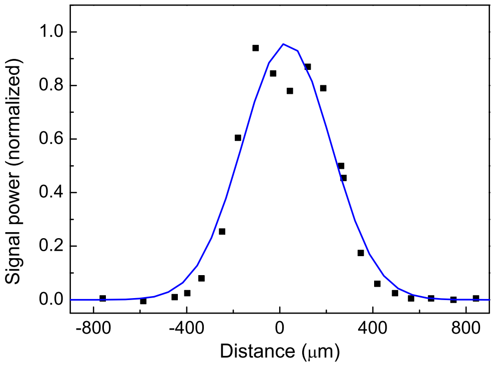

Fig. 4. The output signal of lock-in amplifier depends on the optical delay of the fundamental light.

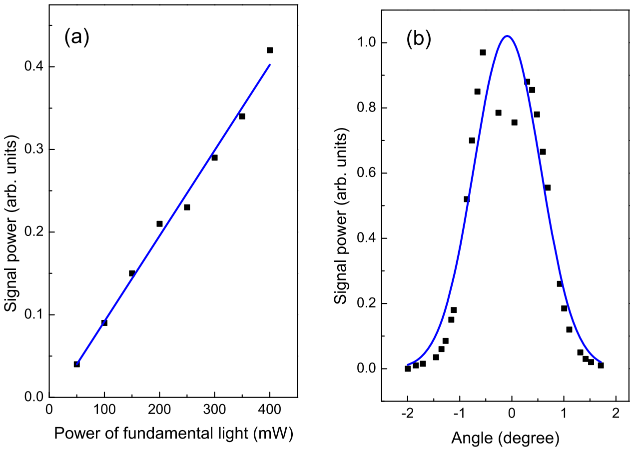

Fig. 5. The 5ω signal measured by changing parameters. (a) The 5ω signal plotted against the power of fundamental light. (b) The 5ω signal versus the crystal-angle.

To further confirm the generation, the strength of the achieved 5ω signal was also measured by changing power of the fundamental beam and the KBBF angle. Figure 5(a) gives the measured lock-in amplifier output when the fundamental power was changed. It indicates that the 5ω signal scales linearly to the fundamental power as expected. Figure 5(b) shows the obtained 5ω signal strength as a function of the incident KBBF angle. The squares give the measured data while the curve shows a theoretical fitting obtained by introducing a mismatching parameter. The results indicate that a full width at half-maximum (FWHM) of the temperature tolerance is about 2°C.

5. Conclusion

In conclusion, we have demonstrated the generation of quasi-cw vacuum ultraviolet light at 160 nm with a repetition rate of 82 MHz by two SHGs and one SFM. More than 6 h of stable operation of generating 200 nm light at power of 7.5 mW was obtained. The generated coherent quasi-cw VUV can be used in many applications, such as the study of high-temperature superconductivity with angle-resolved photoelectron spectroscopy.

References

1. J. Meng, G. Liu, W. Zhang, *et al.*, "Coexistence of Fermi arcs and Fermi pockets in a high-T_c copper oxide superconductor," Nature **462**, 335 (2009).
2. Z. Xie, S. He, C. Chen, *et al.*, "Orbital-selective spin texture and its manipulation in a topological insulator," Nat. Commun. **5**, 3382 (2014).
3. T. Kiss, F. Kanetaka, T. Yokoya, *et al.*, "Photoemission spectroscopic evidence of gap anisotropy in an f-electron superconductor," Phys. Rev. Lett. **94**, 057001 (2005).
4. L. Kang and Z. S. Lin, "Deep-ultraviolet nonlinear optical crystals: concept development and materials discovery," Light Sci. Appl. **11**, 201 (2022).
5. C. P. Christensen, R. W. Waynant, and B. J. Feldman, "High efficiency microwave discharge XeCl laser," Appl. Phys. Lett. **46**, 321 (1985).
6. L. V. Dao, K. A. Tran, and P. Hannaford, "Cascaded four-wave mixing in the XUV region," Opt. Lett. **43**, 134 (2018).
7. C. T. Chen, Z. Y. Xu, D. Deng, *et al.*, "The vacuum ultraviolet phase-matching characteristics of nonlinear optical $KBe_2BO_3F_2$ crystal," Appl. Phys. Lett. **68**, 2930 (1996).
8. L. J. Liu, L. Zhao, X. J. Zhou, *et al.*, "Recent progress in the development of $KBe_2BO_3F_2$: a deep-UV nonlinear optical crystal," Appl. Phys. B **128**, 17 (2022).
9. T. Togashi, T. Kanai, T. Sekikawa, *et al.*, "Generation of vacuum-ultraviolet light by an optically contacted, prism-coupled $KBe_2BO_3F_2$ crystal," Opt. Lett. **28**, 254 (2003).
10. C. Chen, G. Wang, X. Wang, *et al.*, "Improved Sellmeier equations and phase-matching characteristics in deep-ultraviolet region of $KBe_2BO_3F_2$ Crystal," IEEE J. Quantum Electron. **44**, 617 (2008).
11. C. T. Chen, G. L. Wang, X. Y. Wang, *et al.*, "Deep-UV nonlinear optical crystal $KBe_2BO_3F_2$-discovery, growth, optical properties and applications," Appl. Phys. B **97**, 9 (2009).
12. M. Scholz, D. Opalevs, P. Leisching, *et al.*, "A bright continuous-wave laser source at 193 nm," Appl. Phys. Lett. **103**, 051114 (2013).
13. T. Kanai, X. Wang, S. Adachi, *et al.*, "Watt-level tunable deep ultraviolet light source by a KBBF prism-coupled device," Opt. Express **17**, 8696 (2009).
14. X. Zhang, Z. M. Wang, G. L. Wang, *et al.*, "Widely tunable and high-average-power fourth-harmonic generation of a Ti:sapphire laser with a $KBe_2BO_3F_2$ prism-coupled device," Opt. Lett. **34**, 1342 (2009).
15. J. Fischer, J. Drs, F. Labaye, *et al.*, "Efficient XUV-light out-coupling of intracavity high harmonics by a coated grazing-incidence plate," Opt. Express **30**, 30969 (2022).
16. K. Wakui, K. Hayasaka, and T. Ido, "Generation of vacuum ultraviolet radiation by intracavity high-harmonic generation toward state detection of single trapped ions," Appl. Phys. B **117**, 957 (2014).
17. J. Seres, E. Seres, E. Cespedes, *et al.*, "Nonperturbative generation of harmonics by nanometer-scale localized electronic states on the surface of bulk materials and nano-films," Optics **4**, 246 (2023).
18. J. Zhang, L. Q. Hua, Z. Chen, *et al.*, "Extreme ultraviolet frequency comb with more than 100 μW average power below 100 nm," Chin. Phys. Lett. **37**, 124203 (2020).
19. C. Zhang, P. Li, J. Jiang, *et al.*, "Tunable VUV frequency comb for ^{229m}Th nuclear spectroscopy," Opt. Lett. **47**, 5591 (2022).
20. A. Ozawa, Z. G. Zhao, M. Kuwata-Gonokami, *et al.*, "High average power coherent VUV generation at 10 MHz repetition frequency by intracavity high harmonic generation," Opt. Express **23**, 15107 (2015).
21. J. J. Li, F. F. Zhang, Z. M. Wang, *et al.*, "High-energy single-frequency 167 nm deep ultraviolet laser," Opt. Lett. **43**, 2563 (2018).
22. S. B. Dai, N. Zong, F. Yang, *et al.*, "167.75-nm vacuum-ultraviolet ps laser by eighth-harmonic generation of a 1342-nm Nd:YVO_4 amplifier in KBBF," Opt. Lett. **40**, 3268 (2015).
23. Y. Li, N. Zong, S. Dai, *et al.*, "Experimental and theoretical investigations on high power vacuum-ultraviolet laser at 165 nm by eighth-harmonic generation in KBBF," Opt. Laser Technol. **120**, 105756 (2019).
24. Z. Zhang, H. Han, G. Zhao, *et al.*, "Simulations and experiments toward continuous wave 167 nm laser generation for ARPES with high energy resolution," Front. Phys. **10**, 870339 (2022).
25. T. Kanai, T. Kanda, T. Sekikawa, *et al.*, "Generation of vacuum-ultraviolet light by an optically contacted, prism-coupled $KBe_2BO_3F_2$ crystal," J. Opt. Soc. Am. B **21**, 370 (2004).
26. Y. Nomura, Y. Ito, A. Ozawa, *et al.*, "Coherent quasi-cw 153 nm light source at 33 MHz repetition rate," Opt. Lett. **36**, 1758 (2011).
27. Y. Zhang, Y. Sato, N. Watanabe, *et al.*, "Generation of quasi-continuous-wave vacuum-ultraviolet coherent light by fourth-harmonic of a Ti:sapphire laser with KBBF crystal," Opt. Express **17**, 8119 (2009).
28. Y. Zhang, R. Hoshi, D. Yamaguchi, *et al.*, "Generation of quasi-cw deep ultraviolet light below 200 nm by an external cavity with a Brewster-input KBBF prism coupling device," Opt. Commun. **295**, 176 (2013).
29. R. K. Li, L. R. Wang, X. Y. Wang, *et al.*, "Dispersion relations of refractive indices suitable for $KBe_2BO_3F_2$ crystal deep-ultraviolet applications," Appl. Opt. **55**, 10423 (2016).
30. T. Nakazato, I. Ito, Y. Kobayashi, *et al.*, "Phase-matched frequency conversion below 150 nm in $KBe_2BO_3F_2$," Opt. Express **24**, 17149 (2016).

31. X. Zhou, D. Yoshitomi, Y. Kobayashi, *et al.*, "1 W average-power 100 MHz repetition-rate 259 nm femtosecond deep ultraviolet pulse generation from ytterbium fiber amplifier," Opt. Lett. **35**, 1713 (2010).
32. X. Zhang, L. R. Wang, X. Y. Wang, *et al.*, "High-power sixth-harmonic generation of an Nd:YAG laser with $KBe_2BO_3F_2$ prism-coupled devices," Opt. Commun. **285**, 4519 (2012).
33. L. R. Wang, N. X. Zhai, L. J. Liu, *et al.*, "High-average-power 266 nm generation with a $KBe_2BO_3F_2$ prism-coupled device," Opt. Express **22**, 27086 (2014).
34. Z. M. Wang, J. Y. Zhang, F. Yang, *et al.*, "Stable operation of 4 mW nanoseconds radiation at 177.3 nm by second harmonic generation in $KBe_2BO_3F_2$ crystals," Opt. Express **17**, 20021 (2009).