Contents lists available at ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

Full length article

All-solid-state ultraviolet 330 nm laser from frequency-doubling of Nd:YLF red laser in CsB₃O₅

Ming Chen^{a,c}, Zhi-chao Wang^{a,*}, Bao-shan Wang^a, Feng Yang^a, Guo-chun Zhang^b, Shen-jin Zhang^{a,*,1}, Feng-feng Zhang^a, Xiao-wen Zhang^a, Nan Zong^a, Zhi-min Wang^a, Yong Bo^a, Qin-jun Peng^a, Da-fu Cui^a, Yi-cheng Wu^b, Zu-yan Xu^a

^a Research Center for Laser Physics and Technology, Key Lab of Function Crystal and Laser Technology, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China

^b Beijing Center for Crystal Research and Development, Key Lab of Function Crystal and Laser Technology, Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China

ABSTRACT

^c University of Chinese Academy of Sciences, Beijing 100190, China

ARTICLE INFO

Article history: Received 23 September 2015 Received in revised form 18 November 2015 Accepted 14 December 2015 Available online 18 December 2015

Keywords: Nd:YLF crystal SHG UV laser CsB₃O₅

Compact all-solid-state UV lasers have attracted much attention in recent years for their important applications in spectroscopy, bio-analysis, material science, medical surgery and precision manufacturing [1–7]. Most all-solid-state UV lasers are based on frequency conversion from a corresponding infrared laser in nonlinear optical (NLO) crystals. Up to now, as one of the most studied UV lasers, the 355 nm light is commonly generated by frequency tripling of a 1.06 μ m Nd-doped laser in LiB₃O₅ (LBO) crystal [6–9]. Because of the relatively larger effective nonlinear coefficient, high resistance against laser-induced damage and high transparency in the UV region, the CsB₃O₅ (CBO) crystal is also a good choice for high-power UV light generation and several excellent results have been reported, such as the 355 nm laser from third-harmonic generation (THG) of a 1064 nm Nd:YAG laser [10,11] and the 281 nm laser by fourth-harmonic generation (FHG) of a 1123 nm Nd:YAG laser [12]. However, to the best of our knowledge, there is prior no report on UV 330 nm laser from frequency quadrupling of a 1321 nm Nd:YLF laser in CBO crystal.

* Corresponding authors.

E-mail addresses: cationhaigou@163.com (Z.-c. Wang), zhangshenjin@163.com (S.-j. Zhang).

We demonstrate an ultraviolet (UV) 330 nm laser from second-harmonic generation (SHG) of an allsolid-state Nd:YLF red laser in a CsB_3O_5 (CBO) crystal for the first time, to our best knowledge. Under an input power of 4.8 W at 660 nm, a maximum average output power of 330 nm laser was obtained to be 1.28 W, corresponding to a frequency conversion efficiency of about 26.7%.

© 2015 Elsevier B.V. All rights reserved.

The UV 330 nm laser centered at 30, 272.51 cm^{-1} $(\lambda = 330.333 \text{ nm})$ with a linewidth of $\Delta v = 3.5 \text{ GHz}$ is suitable to excite the ${}^{3}S_{1/2} \rightarrow {}^{4}P_{3/2}$ sodium transition, which can be applied in producing polychromatic laser guide star (PLGS) to increase the sky coverage using adaptive optics in large telescopes [13–15]. Fig. 1 shows the sodium energy diagram and relaxation pathways of one photon excitation at 330 nm and two-photon excitation at 589 and 569 nm. It can be seen from Fig. 1, the PLGS can be generated by a one-photon direct excitation of the ${}^{3}S_{1/2} \rightarrow {}^{4}P_{3/2}$ sodium transition at 330 nm or two-photon excitation using the 589 and 569 nm lasers. The one-photon direct excitation has several advantages over the two-photon scheme, such as no time synchronization and spatial overlap, several robust solid state laser solutions are available for selection. Especially, the one-photon scheme is more efficient in producing a returned fluorescence flux at 330 nm. For example, 1 W laser at 330 nm is enough to get the same returned flux as the one which is obtained with two lasers at 589 and 569 nm of 15 W each [16]. On the other hand, Nd:YLF crystal is a natural birefringence material and has a longer upperlaser-level lifetime compared with the Nd:YAG and Nd:YVO4 crystals, which is suitable for generating high power output with high beam quality, particularly for pulse mode [17,18]. Moreover, a vacuum UV (VUV) laser with wavelength at 165 nm can be obtained by frequency-doubling of the 330 nm laser, which is







¹ Postal address: 29 Zhongguancun East Road, Haidian District, Beijing, 100190, PR China.

almost the shortest VUV laser wavelength through direct secondharmonic generation (SHG) with KBe₂BO₃F₂ (KBBF) crystal [19]. An angle-resolved photoemission spectroscopy (ARPES) with thus higher photon energy (7.52 eV) VUV 165 nm laser may be able to reach larger momentum space and maintain bulk sensitivity [20].

In this paper, we firstly report on an all-solid-state UV laser at 330 nm by SHG of an intra-cavity frequency-doubled Nd:YLF red laser in a CBO crystal. A maximum average output power of 1.28 W at 330 nm was obtained with pulse width of \sim 200 ns. The SHG conversion efficiency was 26.7% from red to ultraviolet. The beam quality factor M^{-2} was measured to be 2.5 at the maximum output power.

NLO crystal CBO belongs to the orthorhombic system with 222 point-group symmetry. According to the Sellmeier equations [21], the phase-matching (PM) angle of CBO for Type-I SHG from 660 nm to 330 nm can be calculated to be θ =41.7°. The effective NLO coefficient (d_{eff}) of the CBO crystal was expressed as follows [10]:

$$d_{eff} = d_{14} \sin 2\theta$$
 (for Type I, in the yz plane) (1)

where θ is PM angle. The value of d_{14} coefficient was calculated to be 1.08 pm/V [22]. The PM angle of θ =41.7° yields a moderate d_{eff} =1.07 pm/V according to Eq. (1) for the SHG at 330 nm with the walk-off angle of 21.9 mrad and acceptance angle of 1.93 mrad × cm. Considering these parameters, the CBO crystal shows a great potential for efficient 330 nm generation with Type-I PM SHG.

The experimental configuration is illustrated in Fig. 2. At first, we developed a red laser at 660 nm from intra-cavity frequencydoubled 1321 nm Nd:YLF laser. The Q-switched red laser delivers an average output power of 4.8 W with pulse duration of 280 ns at a repetition rate of 2.5 kHz and the beam quality factor M^2 was measured to be 1.9. The center wavelength and linewidth at 660 nm were measured to be 660.551 nm and 4.4 GHz with a wavelength meter (WS-7 High finesse GmbH, 350–1120 nm), respectively. Then, we employed lens F₁ and lens F₂ to optimize the power intensity for high conversion efficiency. All the lenses in the experiment were antireflection (AR) coated at 660 and 330 nm. The red laser beam was collimated by lens F₁, and then focused by lens F₂ into a Type I PM CBO ($4 \times 4 \times 8$ mm³, $\theta = 41.7^{\circ}$)



Fig. 1. Sodium energy diagram and relaxation pathways of one photon excitation at 330 nm and two-photon excitation at 589 nm+569 nm.

crystal with the beam diameter of $\sim\!120\,\mu\text{m}$ to generate the 330 nm laser. The CBO crystal was cut at PM angle and polished in both end faces without coatings. Finally, the UV laser was separated from the red laser beam by a quartz Brewster prism after collimated by lens F_3.

Fig. 3 shows the output power of 330 nm UV laser versus the input power at 660 nm. As seen from Fig. 3, the UV laser output power grows monotonically with the increasing input power and shows no sign of saturation, which suggests that there is a potential to obtain higher UV power by means of increasing the power of the input laser. At an input power of 4.8 W at 660 nm with power density of $\sim 60.7 \text{ MW/cm}^2$, the maximum output power of 1.28 W at 330 nm was obtained, corresponding to a frequency conversion efficiency of about 26.7%. For comparison, we also used a Type I PM LBO crystal (4 mm \times 4 mm \times 30 mm, θ =90°, $\omega = 49.7^{\circ}$) to replace the CBO crystal and the beam diameter in LBO was same as used in CBO mentioned above for SHG at 330 nm. Both the entrance and the exit surfaces of the LBO crystal were AR coated at 660 and 330 nm. The maximum output power of the 330 nm laser was measured to be 0.74 W with a corresponding SHG conversion efficiency of about 15.2%. Although the length of CBO is shorter than that of LBO, the SHG efficiency and output power with CBO are 1.76 and 1.73 times than that of LBO, which indicates that the CBO crystal seems to be a better choice for higher UV output power.

The spectrum of generated UV laser monitored by an optical spectrum analyzer (AvaSpec-2048FT-SPU) at the maximum output power was exhibited in Fig. 4. The center of the SHG wavelength was deduced to be 330.276 nm based on the input laser wavelength at 660.551 nm. The linewidth of radiation at 330 nm can be estimated to be 6.2 GHz based on that at 660 nm with the following formulas [23]:

$$\Delta t \times \Delta v = C, \tag{2}$$

$$\Delta t_{\rm S} = \sqrt{\lambda_{\rm S}/\lambda_{\rm I}} \Delta t_{\rm I},\tag{3}$$



Fig. 3. Output power of 330 nm laser as a function of the input power at 660 nm.



Fig. 2. Experimental setup of the UV 330 nm Nd:YLF laser.



Fig. 4. Measured Spectrum of the UV 330 nm laser.



Fig. 5. A typical temporal profile of a single pulse at 330 nm.



Fig. 6. M^2 measurements of UV 330 nm laser under the highest output power. Inset: far-field 2D beam profile.

where, Δt is the pulse width, Δv is the linewidth, and *C* is a constant, Δt_s and Δt_l are the pulse width of the second-harmonic (SH) and input lights, λ_s and λ_l are the wavelength of the SH and input pulses, respectively. As the Nd:YLF gain spectra around 1321 nm is broad enough to include wavelength of interest

[16,24,25], the center wavelength of 330 nm laser can be tuned from 330.276 nm to 330.333 nm and the linewidth of the laser can be narrowed by inserting a dispersion element inside the cavity, such as an etalon [24,26]. The further work is on the way.

The pulse temporal profile was recorded by an oscilloscope (Tektronix DPO4104, 1.5 GHz bandwidth) and a fast silicon photodiode detector with the rise time of 2.3 ns. A typical oscilloscope trace of a single pulse is presented in Fig. 5 with a pulse width of about 200 ns at the maximum output power. The pulse energy and peak power were 0.51 mJ and 2.6 kW, respectively.

The beam quality at 330 nm was measured by a laser beam analyzer (Spiricon- M^2 200 s) at the output power of 1.28 W. As presented in Fig. 6, the measured beam quality factors are 2.7 in horizontal axis and 2.3 in vertical axis, which corresponds to an average value of M^2 =2.5. The inset in Fig. 6 exhibits a far-field two-dimensional (2D) beam intensity profile, which shows that the laser operates in a near Gaussian mode. The beam spot became slight ellipse, which may be caused by the walk-off effect occurred in the CBO crystal.

In summary, we have reported the first demonstration, to the best of our knowledge, on an UV 330 nm laser from frequencydoubled of an all-solid-state Nd:YLF red laser in a CBO crystal. A maximum average output power of 330 nm laser was obtained to be 1.28 W. The conversion efficiency from 660 nm to 330 nm is up to about 26.7%. This 330 nm laser is promising for PLGS. Mean-while, this high power UV laser can be frequency-doubled to 165 nm light for VUV laser-based ARPES. In addition, the further scaling of the 330 nm output power and higher conversion efficiency can be expected by means of increasing the red laser power or using a longer CBO crystal with AR coatings.

Acknowledgments

This research was supported by the National Natural Science Foundation China (Grant no. 61138004), the National Instrument of China (Grant no. 2012YQ120048), and the National Development Project for Major Scientific Research Facility (No. ZDYZ2012-2).

References

- H. Kitano, T. Matsui, K. Sato, N. Ushiyama, M. Yoshimura, Y. Mori, T. Sasaki, Opt. Lett. 28 (2003) 263.
- [2] Z.C. Wang, F. Yang, G.C. Zhang, Y. Bo, S.S. Liu, S.Y. Xie, Y.T. Xu, N. Zong, F.Q. Li, B. L. Liu, J.L. Xu, Q.J. Peng, J.Y. Zhang, D.F. Cui, Y.C. Wu, Z.Y. Xu, Opt. Lett. 37 (2012) 2403.
- [3] P. Zhu, D.J. Li, Q.Y. Liu, J. Chen, S.J. Fu, P. Shi, K. Du, P. Loosen, Opt. Lett. 38 (2013) 4716.
- [4] B.T. Zhang, H.T. Huang, J.F. Yang, J.L. He, C.H. Zuo, J.L. Xu, X.Q. Yang, S. Zhao, Opt. Commun. 283 (2010) 2369.
- [5] F.Q. Jia, Q. Zheng, Q.H. Xue, Y.K. Bu, L.S. Qin, Appl. Opt. 46 (2007) 2975.
- [6] X. Yan, Q. Liu, H. Chen, X. Fu, M. Gong, D. Wang, Laser Phys. Lett. 7 (2010) 563.
- [7] C. Jung, W. Shin, B.A. Yu, Y.L. Lee, Y.C. Noh, Opt. Express 20 (2012) 941–948.
- [8] Y.J. Huang, Y.P. Huang, P.Y. Chiang, H.C. Liang, K.W. Su, Y.F. Chen, Appl. Phys. B 106 (2012) 893–898.
- [9] X.P. Yan, Q. Liu, C. Pei, D.S. Wang, M.L. Gong, J. Opt. 16 (2014) 045201.
- [10] Y.C. Wu, P.Z. Fu, J.X. Wang, Z.Y. Xu, L. Zhang, Y.F. Kong, C.T. Chen, Opt. Lett. 22 (1997) 1840.
- [11] L. Guo, G.L. Wang, H.B. Zhang, D.F. Cui, Y.C. Wu, L. Lu, J.Y. Zhang, J.Y. Huang, Z. Y. Xu, Appl. Phys. B 88 (2007) 197.
- [12] F. Yang, B.L. Liu, Z.C. Wang, Y. Bo, G.C. Zhang, Q.J. Peng, J.Y. Zhang, D.F. Cui, Y. C. Wu, Z.Y. Xu, IEEE J. Quantum Electron. 50 (2014) 423.
- [13] T. Stacewicz, P. Kozlowski, Appl. Phys. B 65 (1997) 69.
- [14] J.P. Pique, I.C. Moldovan, V. Fesquet, J. Opt. Soc. Am. A 23 (2006) 2817.
- [15] T. Stacewicz, N.A. Gorbunov, P. Kozlowski, Appl. Phys. B 66 (1998) 461.
- [16] J.P. Pique, I.C. Moldovan, V. Fesquet, H.G. Chatellus, F. Marc, Proc. SPIE 6272 (2011) 62723D.
- [17] R.C. Botha, H.J. Strauss, C. Bollig, W. Koen, O. Collett, N.V. Kuleshov, M.J. D. Esser, W.L. Combrinck, H.M. Bergmann, Opt. Lett. 38 (2013) 980.
- [18] A.M. Deana, M.A.P.A. Lopez, N.U. Wetter, Opt. Lett. 38 (2013) 4088.
- [19] C.T. Chen, G.L. Wang, X.Y. Wang, Y. Zhu, Z.Y. Xu, T. Kanai, S. Watanabe, IEEE J. Quantum Electron. 44 (2008) 617–621.

- [20] G.D. Liu, G.L. Wang, Y. Zhu, H.B. Zhang, G.C. Zhang, X.Y. Wang, Y. Zhou, W. T. Zhang, H.Y. Liu, L. Zhao, J.Q. Meng, X.L. Dong, C.T. Chen, Z.Y. Xu, X.J. Zhou, Rev. Sci. Instrum. 79 (2008) 023105.
- [21] K. Kato, IEEE J. Quantum Electron. 31 (1995) 169.
- [22] C.T. Chen, Y.C. Wu, A.D. Jiang, B.C. Wu, G.M. You, R.K. Li, S.J. Lin, J. Opt. Soc. Am. B 6 (1989) 616.
- [23] Z. Xu, W. Tu, F. Yang, N. Zong, B.S. Wang, F.F. Zhang, S.J. Zhang, Z.M. Wang, Q. J. Peng, D.F. Cui, J.Y. Zhang, X.Y. Wang, C.T. Chen, Z.Y. Xu, IEEE Photon. Technol. Lett. 26 (2014) 980.
- [24] Y. Louyer, F. Balembois, M.D. Plimmer, T. Badr, P. Georges, P. Juncar, M. E. Himbert, Opt. Commun. 217 (2003) 357.
- [25] L. Fornasiero, T. Kellner, S. Kück, J.P. Meyn, P.E.-A. Möbert, G. Huber, Appl. Phys. B 68 (1999) 67.
- [26] P.Y. Wang, S.Y. Xie, Y. Bo, B.S. Wang, J.W. Zuo, Z.C. Wang, Y. Shen, F.F. Zhang, K. Wei, K. Jin, Y.T. Xu, J.L. Xu, Q.J. Peng, J.Y. Zhang, W.Q. Lei, D.F. Cui, Y.D. Zhang, Z.Y. Xu, Chin. Phys. B 23 (2014) 094208.