All-Solid-State 228.5 nm DUV Light Source by quadrupling of an Acousto-Optically Q-Switched Nd:YVO₄ Laser

ZHIBIN ZHAO¹,², HAO CHEN², GUOJUN LIU³, CHENG CHENG², QUAN LI², ZHONGLIANG QIAO², YI QU², QIONGTAO XIE², JIANG ZHU², QUAN ZHENG³, and BAOXUE BO*¹

¹ State Key Laboratory of High Power Semiconductor Laser, Changchun University of Science and Technology, Changchun, Jilin, 130022, China
² College of Physics and Electronic Engineering, Key Laboratory of Laser Technology and Optoelectronic Functional Materials of Hainan Province, Hainan Normal University, Haikou, Hainan, 571158, China
³ Changchun New Industries Optoelectronics Technology Co., Ltd., Changchun, Jilin, 130103, China

Corresponding author: Guojun Liu (gjlu626@126.com), Yi Qu (2686566673@qq.com), and Baoxue Bo (1523001971@qq.com).

This work was supported in part by Major Science and Technology Program of Hainan Province of China under Grant ZDKJ2019005; in part by the Hainan Provincial Natural Science Foundation of China under Grant 618QN241; in part by the Young Talents Science and Technology Innovation Project of Hainan Association for Science and Technology under Grant GQXM2018010; in part by Special Research Project of Hainan Academician Innovation Platform under Grant SPTZX202034; in part by the National Natural Science Foundation of China under Grant 61774024, Grant 61864002, Grant 11764012, and Grant 61964007; in part by Key R&D Projects in Hainan Province under Grant ZDYF2020020, Grant ZDYF2020036 and Grant ZDYF2020217; in part by Hainan Natural Science Foundation Innovation Research Team under Grant 2018CXTD336.

ABSTRACT All-solid-state 228.5 nm Deep Ultraviolet (DUV) laser has been studied with two-step second harmonisation generation (SHG) of Nd:YVO₄ 914 nm fundamental laser, by optimizing its resonator design and SHG configurations. Adopting three-mirror folded V cavity and acoustooptical Q-switching, high peak power 457 nm laser output has been achieved by intracavity frequency-doubling of LD pumped Nd:YVO₄ 914 nm fundamental laser. At pump power of 41 W, the average output power for 457 nm laser has reached 600 mW at repetition frequency 10 kHz, 50 ns pulse width. With Type-I phase matching BBO crystal, externally frequency-doubling of 457 nm blue output was realized and optimized. Under LD pump power of 41 W, DUV laser at 228.5 nm with average output power of 35 mW has been achieved, at repetition frequency 10 kHz and pulse width of 46 ns. Under these conditions, the frequency doubling conversion efficiency is 5.8%, and the DUV laser output power instability is less than 2% in 2-hour test.

INDEX TERMS All-solid-state laser, Acousto-optical Q-switching, Deep Ultraviolet laser, 228.5 nm output.

I. INTRODUCTION

DUV lasers, owing to their special wavelength range, have found many applications in material processing and spectral analysis [1]-[6]. At present, these applications mainly adopt excimer lasers as the laser source. In comparison, DUV all-solid-state laser based on nonlinear frequency conversion has gained extensive attention due to its high efficiency, compactness and low cost of system maintenance [7], [8]. The most common way of realizing all-solid-state DUV lasers is quadruple-frequency conversion of Nd:YAG 1064 nm laser, achieving DUV 266 nm output [9]-[12]. However, for some applications, such as 5G electronic material, grating and waveguide processing, wave-lengths shorter than 250 nm can be more appealing owing to higher efficiencies [13]. Still, some special spectral applications need specific DUV wavelengths. Lasers at about 228 nm wave-lengths have proved to be especially suitable for testing genome DNA Methylation [14], protein structure [15], and Raman spectral analysis of explosives [16]. One way of achieving ~228 nm lasers is the quadruple-frequency conversion of either the 914 nm output of Nd:YVO₄ crystal, or 912 nm output of Nd:GdVO₄ crystal. Sergei V. Bykov et al. [17] reported acousto-optically Q-Switched, frequency quadrupled 912 nm Nd:GdVO₄ laser with the average output power of 30 mW. Nd:YVO₄ and Nd:GdVO₄ have some advantages in common, including high absorption cross section, broad absorption bandwidth and polarization output. Although there exist some thermal lens effect inside Nd:YVO₄ crystal as the laser gain medium, this thermal lens effect can be taken into consideration in the cavity design, thermal stable cavity can be achieved using the Nd:YVO₄ crystal pumping facet as one mirror of the laser cavity. This not only simplifies the cavity
system, the overall system cost is also lower. What is more, Nd:GdVO₄ crystal is ~3 times more expensive than Nd:YVO₄ crystal, that is one of the reasons we conduct DUV laser study by quadrupling of Nd:YVO₄ laser. So far, the 914 nm spectral line of Nd:YVO₄ crystal has been mainly reported to generate the second harmonic to obtain the CW 457 nm laser [18], [19]. The 914 nm laser output of Nd:YVO₄ crystal corresponds to a Quasi-three-level system, with much smaller stimulated emission cross section than four-level system, and thus lower efficiency. In order to enhance the quadruple frequency conversion efficiency and thus the DUV 228.5 nm output, it is necessary to optimize the whole laser system. In this work, three-mirror folded V cavity and acousto-optical Q-switching were used to achieve 914 nm fundamental laser pulse operation, LBO crystal was adopted to perform the intracavity SHG and achieve 457 nm output. Finally, BBO crystal was used to achieve the external-cavity frequency-doubled 228.5 nm pulsed laser output.

II. THEORETICAL ANALYSIS

A. OPTIMIZATION OF RESONANT CAVITY PARAMETERS

The three-mirror folded V cavity adopted in this work is shown in Fig.1. The pumping facet of Nd:YVO₄ crystal was used as one of the reflecting mirrors of the laser cavity. The thermal focal length was taken into consideration in the selection of resonant cavity parameters. In this work, thermal lens focal length of the laser crystal \( f_{\text{thermal}} \) was calculated numerically according to [20], [21]:

\[
f_{\text{thermal}} = \frac{\pi k_e w_p^2}{p_{ph}(dn/dT)} \left( \frac{1}{\exp(-al)} \right) \tag{1}
\]

Where \( p_{ph} \) is the thermally induced power; \( dn/dT \) is the coefficient of change of refractive index with temperature; \( k_e \) is the thermal conductivity; \( w_p \) is pump beam radius; \( \alpha \) is absorption coefficient, \( l \) is the crystal length.

In this work, Nd:YVO₄ length \( l \) is 5 mm, Nd³⁺ atomic doping concentration is 0.1%. For this crystal, \( k_e = 0.0523 \) W/cm/K, \( \alpha = 4.1 \) cm⁻¹, \( dn/dT \) is \( 2.2 \times 10^6 \) K⁻¹, the power conversion percentage from pump power to heat is 15%. From these parameters and for different pump beam radius \( w_p = 200 \) µm, 300 µm and 400 µm, the heat lens focal lengths have been obtained under different pump powers, as shown in Fig.2. For pump power of 41 W, the heat lens focal lengths are 50 mm, 109 mm and 194 mm, respectively. Putting these focal lengths into V cavity parameters, using ABCD Matrix and cavity stability condition, by Matlab numerical calculations, the cavity beam waist radius for M1 and M2 as the cavity mirrors are 118 µm, 125 µm and 128 µm, respectively. So from the perspective of mode matching, the pump beam radius of 200 µm has been adopted in this design. The effects of L1 and L2 on optical beam radius inside the cavity have been obtained, as depicted in Fig.3 and Fig.4. From these figures, it is clear that L1 has little effect on the beam radius at different positions inside the cavity, while L2 has bigger impact on the beam radius. Therefore in this work, L2 needs to be carefully adjusted. Based on the above analysis and device parameters, L1 and L2 have been set at 83 mm and 31 mm, respectively.

![Experimental apparatus](https://example.com/figure1.png)

**FIGURE 1.** Experimental apparatus.

![Relationship between Nd:YVO₄ thermal focal length and pump power](https://example.com/figure2.png)

**FIGURE 2.** Relationship between Nd:YVO₄ thermal focal length and pump power.
Where $L_c$ is the round trip loss of the fundamental light in the cavity, $n_{2\omega}$ is the refractive index of LBO crystal, $v_{\omega}$ is the fundamental light frequency, $I_S = h\nu_\omega / \sigma_0 \tau l$ is the saturation light intensity of fundamental light, $\sigma_0$ is the absorption cross section of lasing medium, $\omega_{p1}$, $\omega_{p2}$ are the beam waists of fundamental light at laser medium and frequency doubling crystal, respectively. When $L_c = 0.02$, the optimum LBO crystal length of about 16.4 mm for 457 nm light generation can be obtained. Considering the V cavity effective space and the free adjustment range of various devices, the LBO crystal length of 15 mm has been set in the experiments. Using SNLO software, the nonlinear characteristics of LBO and BiBO crystals can be obtained for the generation of 457 nm wavelength, as shown in Table 1.

### TABLE 1. Nonlinear Optical Characteristics for 457 nm Generation in Borate Crystals.

| Name of Borate Crystals | LBO | BiBO |
|-------------------------|-----|------|
| Nonlinear coefficient deff (pm/V) | 0.803 | 3.44 |
| Acceptance angle (mrad/cm) | 4.56 | 1.13 |
| Walk off angle (mrad) | 12.48 | 44.99 |
| Phase-matching angle (deg) | $\theta=90$ | $\theta=159.6$ |
| Hygroscopicity | Slight | Moderate |

Barium borate $\text{BaB}_2\text{O}_4$ (BBO) is one of the most common nonlinear crystals for quadrupling application to generate UV laser, which has larger nonlinear coefficient, moderate birefringence, higher damage threshold, wider phase matching range, and better temperature stability. In the case of BBO crystal for the generation of 228.5 nm from 457 nm through SHG, which has relatively large nonlinear coefficient of 1.38 pm/V, a phase matching angle of 61.4° can be obtained using SNLO software and numerical calculation. In addition to BBO, $\text{RbB}_2\text{BO}_3\text{F}_2$ (RBBF) and $\text{KB}_2\text{BO}_3\text{F}_2$ (KBBF) can also be used to generate UV light through fourth harmonic generation, but with poor performance [23], [24].

### III. EXPERIMENTAL SETUP

Our experimental setup is shown in Fig.1. The pump source is a 808 nm fiber-coupled LD with a core diameter of 400 μm and a numerical aperture of 0.22, with maximum CW power of 110 W. The wavelength shift coefficient is 0.25 nm°C. The pumping wavelength can be finely tuned to match the central absorption wavelength of Nd:YVO$_4$ through temperature adjustment. The pumping light was focused and collimated into 200 μm in radius through collimation and focusing system, and injected into the Nd:YVO$_4$ crystal. The coupling system was composed of two flat convex mirrors with focal lengths of 10 mm, and 45° polarizer. The parameters of Nd:YVO$_4$ crystal are: Nd$^{3+}$ atomic doping concentration is 0.1%; 4 mm × 4 mm × 5 mm in size; the left facet was antireflection coated at 808 nm and 1064 nm and high reflection coated at 914 nm; the right facet was antireflection coated at 914 nm, 1064 nm and 1342 nm.
wavelengths. The laser crystal was wrapped in a layer of indium foil on the side and secured on a copper heat sink, which is capable of controlling the temperature through circulating water cooling. The output mirror M was a flat concave mirror with radius of curvature of 50 mm, in which the concave surface was antireflection coated at 457 nm, 1064 nm and 1342 nm and high reflection coated at 914 nm; the flat surface was antireflection coated at 457 nm, 914 nm, 1064 nm and 1342 nm. The high reflection mirror M2 was a flat concave mirror with 200 mm in radius of curvature which was high reflection coated at 457 nm and 914 nm. The V shape cavity was formed by the left facet of Nd:YVO₄ crystal M1 and M and M2, where the angle between the two arms is 10°. The long arm was inserted the acousto-optical Q-switch, while the short arm was inserted LBO crystal for SHG, which was put about 1 mm in front of the reflection mirror. The size of LBO crystal is 4 mm × 4 mm × 15 mm, both facets of which were antireflection coated at 457 nm, 914 nm and 1064 nm. The above cavity has an oscillation wavelength of 914 nm, which was converted by SHG in LBO into 457 nm wavelength. M3 was a focusing mirror for 457 nm, which was antireflection coated at 457 nm, at the focus of which BBO crystal was put for quadrupling wave generation. Both facets of BBO crystal were antireflection coated at 457 nm and 228.5 nm, and out from the BBO crystal was the 228.5 nm wavelength. The function of mirror M4 is to isolate 457 nm and 228.5 nm wavelengths.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. 457 NM PULSED LASER OUTPUT

In order to obtain the highest peak power 457 nm pulsed output, the repetition frequencies were set at 5 kHz, 10 kHz, 15 kHz and 20 kHz, respectively, and average output powers and pulse widths were measured at these frequencies. The results show that 457 nm laser has good performance at 10 kHz and 15 kHz, as shown in Fig.5 and Fig.6, from which we note the average power increases with increase of injection power, while the pulse width decreases with the increase of the injection power. At 10 kHz and 41 W injection power, maximum average power of 661 mW was obtained for the 457 nm laser when the pulse width was 50 ns, corresponding to a peak power of 1.2 kW.

At 41 W injection power, maximum average power of 661 mW was obtained for the 457 nm pulse laser output at 15 kHz when the pulse width was 62 ns, corresponding to a peak power of 710 W. These results show that highest peak power can be achieved at 10 kHz repetition frequency. Fig.7 shows the 457 nm laser spot and beam quality at 10 kHz and maximum average power of 600 mW, where TEM₀₀ mode can be clearly seen but with somewhat ellipse spot, which we believe, is the results of the definite angle exists in the V shape folded cavity resulting in the astigmatism. By performing Binomial fitting of beam radius at different positions, M² factors for X and Y directions can be obtained as 1.32 and 1.15, respectively.
The reason for the narrower 228.5 nm DUV laser pulse width than the 457 nm blue laser pulse width is that, at the beginning and end of 457 nm laser pulse, the power is too low for BBO crystal to generate DUV laser output. Shown in Fig.10 is the 228.5 nm laser spot with somewhat elliptic shape, which is the result of the non-negligible walk off angle, about 75.68 mrad at 228.5 nm laser. The laser output stability has been measured for the 228.5 nm DUV laser at maximum average output power of 35 mW, as shown in Fig.11. It is seen that the output power stability is within 35 mW ±1% within 2 hours.

V. CONCLUSION

This paper reports the all-solid-state pulsed 228.5 nm laser design and characterization, in which the 914 nm laser from the Nd:YVO₄ laser gain medium was used as the fundamental frequency. Pulsed blue laser at 457 nm was obtained when V shape folded cavity and acousto-optical Q-switching technology were adopted, and intra cavity SHG was performed with LBO crystal under type-I phase matching. Further, 228.5 nm DUV pulsed laser, with typical output power of 35 mW, was realized using extra cavity frequency conversion with BBO crystal, with stable output power and good output beam quality. To the best of our knowledge, it is the highest average output power of acousto-optically Q-
switched 228.5 nm laser. The 228.5 nm DUV laser source we developed can be favorably used in spectroscopic measurement of explosives and for biomolecules study.

REFERENCES

[1] F. Shao, W. Wang, W. Yang, Z. Yang, Y. Zhang, J. Lan, A. D. Schlieter, and R. Zenobi, "In-situ nanospectroscopic imaging of plasmon-induced two-dimensional [4+4]-cycloaddition polymer-ization on Au (111),” *Nature communications*, vol. 12, p. 4557, Jul. 2021.

[2] A. Steube, T. Schenk, A. Treytasov, and H. P. Saluz, "High-intensity UV laser ChIP-seq for the study of protein-DNA interactions in living cells,” *Nature Communications*, vol. 8, p. 1303, Nov. 2017.

[3] R. Li, D. Dhanikap, J. Chen, A. Krishnamoorthy, T. C. Cesario, and P. M. Rentzepis, “Identification of Live and Dead Bacteria: A Raman Spectroscopic Study,” *IEEE Access*, vol. 7, pp. 23549-23559, Feb. 2019.

[4] R. Qi, X. L. Yu, Z. B. Li, and W. M. Liu, “Non-Abelian Josephson effect between two F=2 spinor Bose-Einstein condensates in double optical traps,” *Physical review letters*, vol. 102, no. 18, p. 18S301, May. 2009.

[5] A. C. Ji, Q. Sun, X. C. Xie, and W. M. Liu, “Josephson effect for photons in two weakly linked microcavities,” *Physical review letters*, vol. 102, no. 2, p. 023602, Jan. 2009.

[6] A. C. Ji, X. C. Xie, and W. M. Liu, “Quantum magnetic dynamics of polarized light in arrays of microcavities,” *Physical review letters*, vol. 99, no. 18, p. 183602, Nov. 2007.

[7] R. Hsiao, Y. Chen, M. Huang, C. Chen, Y. Lin, and C. Wu, “Innovative finding of 266-nm laser regulating CD90 levels in SDSCs,” *Scientific Reports*, vol. 11, p. 13932, Jul. 2021.

[8] B. Zhang, J. Liu, C. Wang, K. Yang, C. Lee, H. Zhang, and J. He, “Recent Progress in 2D Material-Based Saturable Absorbers for All Solid-State Pulsed Bulk Lasers,” *laser and photonics review*, p.900240, Dec. 2019.

[9] Z. Hou, L. Liu, Z. Fang, L. Yang, D. Yan, X. Wang, D. Xu, and C. Chen, “High-power 266 nm laser generation with a NaSrBe2B4O12 crystal,” *Optics Letters*, vol. 43, no.22, pp. 5599-5602, 2018.

[10] Z. Fang, Z. Hou, F. Yang, L. Liu, X. Wang, Z. Xu, and C. Chen, “High-efficiency UV generation at 266 nm in a new nonlinear optical crystal NaSrBe2B4O12,” *Optics Express*, vol. 25, no. 22, pp. 26500-26507, Oct. 2017.

[11] Q. Liu, X. Yan, X. Fu, M. Gong, and D. Wang, “High power all-solid-state fourth harmonic generation of 266 nm at the pulse repetition rate of 100 kHz,” *Laser Phys*, vol. 6, no. 3, pp. 203-206, Nov. 2008.

[12] G. Wang, A. Geng, Y. Bo, H. Li, Z. Sun, Y. Bi, D. Cui, Z. Xu, X. Yuan, X. Wang, G. Shen, and D. Shen, “28.4 W 266 nm ultraviolet-beam generation by fourth-harmonic generation of an all-solid-state laser,” *Opt. Communications*, vol. 259, no. 2, pp. 820-822, Mar. 2006.

[13] L. Deyra, I. Martial, J. Didierjean, F. Balebois, and P. Georges, “Deep-UV 236.5 nm laser by fourth-harmonic generation of a single-crystal fiber Nd:YAG oscillator,” *Optics Letters*, vol. 39, no. 8, Apr. 2014.

[14] F. D’Amico, P. Zucchiatti, K. Latella, M. Pachetti, A. Gessini, C. Masciovecchio, L. Vaccari, and L. Pascolo, “Investigation of genomic DNA methylation by ultraviolet resonant Raman spectroscopy,” *Journal of Biophotonics*, vol. 13, no. 12, p. e20200150, Jul. 2020.

[15] D. K. Asamoto, and J. E. Kim, “UV Resonance Raman Spectroscopy as a Tool to Probe Membrane Protein Structure and Dynamics,” in *Lipid-Protein Interactions*, Humana, New York, NY, 2019, pp. 327-349.

[16] B. S. Leigh, K. L. Monson, and J. E. Kim, “Visible and UV resonance Raman spectroscopy of the peroxide-based explosive HMTD and its photoproducts,” *Forensic Chemistry*, vol. 2, pp. 22-28, Nov. 2016.

[17] S. V. Bykov, R. D. Roppell, M. Michael, A. A. Sanford, “228-nm quadrupled quasi-three-level Nd:GdVO₄ laser for ultraviolet resonance Raman spectroscopy of explosives and biological molecules,” *Journal of Raman Spectroscopy*, vol. 51, no. 12, pp. 2478-2488, Sep. 2020.

[18] Q. H. Xue, Z. Zheng, Y. K. Bu, F. Q. Jia, and L. S. Qian, “High-power efficient diode-pumped Nd:YVO₄/LiB₃O₃ 457 nm blue laser with 4.6 W of output power,” *Optics letters*, vol. 31, no. 8, pp. 1070-1072, Apr. 2006.

[19] H. Yin, S. Zhu, Z. Chen, Q. He, A. Li, Z. Li, Y. Liu, K. Su, G. Zhang, and Y. Chen, “Research on all-solid-state intracavity frequency doubling 457 nm laser with LBO and BIBO crystal,” *Optik*, vol. 127, no. 8, pp. 3862-3866, Apr. 2016.

[20] M. E. Innocenzi, H. T. Yura, C. L. Fincher, and R. A. Fields, "Thermal modeling of continuous-wave end-pumped solid-state lasers," *Applied Physics Letters*, vol. 56, no. 19, pp. 1831-1833, Feb. 1990.