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# High-power picoseconds 355 nm laser by third harmonic generation based on CsB<sub>3</sub>O<sub>5</sub> crystal

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ABSTRACT We report on the high average power third harmonic generation (THG) of a mode-locked picosecond laser in a CsB<sub>3</sub>O<sub>5</sub> (CBO) crystal. The picosecond laser beam at 1064 nm is produced by a home-made 30 W master oscillator power-amplifier (MOPA) Nd:YVO<sub>4</sub> laser system. The maximum THG output at 355 nm is up to 5.4 W. We also investigate the phase matching angle at different temperatures. During high power operation, the temperature of the CBO crystal is set at a high temperature of more than 100 °C. The THG system has shown a fine long-term stability for more than two months of operation.

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## 1 Introduction

Recently, high average power all-solid-state ultrafast ultraviolet (UV) radiation sources have shown potential for application in many fields. For example, all-solid-state, 355 nm-radiation sources are desirable for applications in micromachining of dielectrics and metals because of the advantages of high reliability and low maintenance cost. LiB<sub>3</sub>O<sub>5</sub> (LBO) crystals have been generally used to generate 355 nm UV-radiation through third harmonic generation (THG) of an all-solid-state Nd:YAG/Nd:YVO<sub>4</sub> (1064 nm) laser. Recently a new UV nonlinear crystal CsB<sub>3</sub>O<sub>5</sub> (CBO) has shown superiority characteristics for THG application over other crystals. It has been successfully used to generate 17 W UV-radiation at 355 nm and has shown a high tripling efficiency in our laboratory [1]. The pump source mentioned above is a Q-switched Nd:YAG laser with an output pulse width of 70 ns and a repetition rate of several kHz. Such a system could be used in many fields. However, for some applications, such as printed circuit board (PCB) direct imaging, semiconductor inspection, etc., high repetition rate picosecond UV-sources are more preferable.

We report here on the third harmonic generation (THG) at 355 nm of a picosecond laser using a CBO crystal. The fundamental beam is generated from an amplified semiconductor

saturated absorber mirror (SESAM) mode-locked Nd:YVO<sub>4</sub> laser at a wavelength of 1064 nm. The fundamental beam is first focused into a non-critical phase matched LBO crystal to generate second harmonics at 532 nm, and then the THG is performed by sum frequency-mixing of the fundamental beam with the second harmonics in a CBO crystal.

### 2 Characterization of CsB<sub>3</sub>O<sub>5</sub> crystal

The CBO crystal is a nonlinear optical (NLO) crystal that can be used for deep ultraviolet generation up to 185 nm. It is a relatively new crystal, being first reported in 1993 [2]. Since then some research have been done with it in the field of UV laser generation [1, 3, 4, 8]. This crystal has a wide transparency range from 167 nm to 3000 nm, a high bulk optical damage threshold of 26 GW/cm<sup>2</sup>, and a large effective NLO coefficient for UV light generation [2, 6].

Table 1 shows the related parameters of the CBO and LBO in THG generation [1–3, 5, 10–12]. The data in Table 1 shows that the effective NLO coefficient of the CBO is 1.5 times that of the LBO, though the other parameters of LBO, such as the spatial walk-off and the angular acceptance, are better than for CBO. Therefore, theoretically the efficiency of THG of CBO is expected be twice that of LBO under the conditions of small-signal approximation and neglecting group velocity mismatch (GVM). This has been proven experimentally in a *Q*-switched nanosecond system [1].

Because of hygroscopic deterioration, CBO can not be used safely at room temperature for a long time. This will limit the applications of CBO. To solve this problem, one of the best ways is to heat the crystal to a temperature higher than  $100\,^{\circ}\text{C}$ . However, the temperature coefficient of Sellmeier's equation of CBO has never been reported and, to our knowledge, no one has ever investigated the characterization of THG generation with CBO at different temperatures. We set up an experiment to determine the change of phase matching angle  $(\varphi)$  at different temperatures as is shown in Fig. 1. The CBO crystal is cut for type II phase matching with  $\varphi=42.4^{\circ}$  and  $\theta=90^{\circ}$ .  $\theta$  remained  $90^{\circ}$  during the experiment.

We employ an Nd:YAG laser with pulse width 10 ns, and a repetition rate of 10 Hz at 1064 nm as the pump source. The fundamental light incidents into a type I phase matching LBO crystal for second harmonic generation. Then THG was generated by sum frequency-mixing of the fundamental

	CBO, type II, eo— e, xy-plane	LBO, type II, oe— o, yz-plane
Phasematching angle $d_{ii}$	$\theta = 90^{\circ}, \varphi = 42.4^{\circ}$ $d_{14}(CBO) = 1.17d_{31}(LBO)$	$\theta = 42.7^{\circ}, \varphi = 90^{\circ}$
Nonlinearity $d_{\text{eff}}$ $d_{\text{eff}}(\text{CBO})/d_{\text{eff}}(\text{LBO})$ Spatial walk-off	$d_{14} \sin(2\varphi) \ 1.5 \ 0.93^{\circ}$	$d_{31}\cos{ heta}$ 1 0.53°
Angular acceptance $\Delta\Theta$ <i>L</i> Bandwidth acceptance $\Delta\lambda$ <i>L</i>	2.05 mrad cm 8.11 cm <sup>-1</sup> cm	3.53 mrad cm 11.83 cm <sup>-1</sup> cm

**TABLE 1** Characteristic data of CBO and LBO crystals for third-harmonic generation by sum-frequency mixing of the fundamental 1064 nm radiation with the 532 nm second harmonic at 532 nm

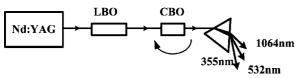
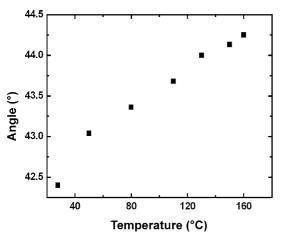


FIGURE 1 Schematics of the experiment setup for determination of the phase matching angle  $(\varphi)$  at different temperature



**FIGURE 2** CBO phase matching angle  $(\varphi)$  of THG at 355 nm as a function of temperature

beam with the residual second harmonics in a CBO crystal. The CBO crystal is set in an oven, whose temperature is controlled with an accuracy of 0.1 °C. Furthermore, we put the oven on a rotation stage to adjust the incidence angle of the two light beams. Finally, a Brewster prism was used to separate 355 nm light from the residual 1064 nm and 532 nm light. We measured the phase matching angles ( $\varphi$ ) from 24 °C to 160 °C. The experimental result is plotted in Fig. 2.

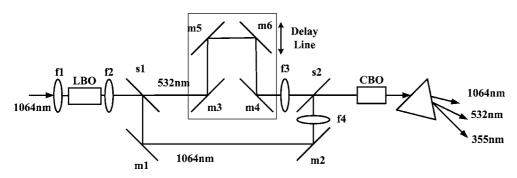
From Fig. 2, we can see that the phase matching angle  $(\varphi)$  only change from 42.4° at 27.8 °C to 44.3° at 160 °C. We now investigate the THG generation of a high average power, picosecond laser system.

#### 3 Experiment setup

Our pump source in the experiment is a home-made master oscillator power-amplifier (MOPA) system. The oscillator is a passively mode-locked Nd:YVO<sub>4</sub> with a semiconductor saturated absorber mirror (SESAM). It is similar to the system reported previously [9], except that the SESAM used were from different sources. The output power of the master oscillator is 3 W with a pulse width of 20 ps, and a repetition rate of 73 MHz at a wavelength of 1064 nm. The power amplifier is a two-stage system. After the first stage, the power is increased to 17 W. The output power is then increased to 30 W in the second stage. The beam quality is near the diffraction limit with  $M_x^2 = 1.06$  and  $M_y^2 = 1.11$  (measured by a M2-200, Spiricon). The details of our MOPA system will be described elsewhere.

The third harmonic (THG) is generated by the sumfrequency mixing (SFM) of the residual fundamental beam at 1064 nm and the second harmonic at 532 nm in the CBO crystal. The CBO crystal used in the THG experiment has dimensions of 4 mm  $\times$  4 mm  $\times$  16 mm and is cut for type II phasematching. The angles are  $\varphi = 43.9^{\circ}$  and  $\theta = 90^{\circ}$  for high temperature (> 100 °C) operation. The surfaces of the crystal are optically polished and uncoated. Finally, the CBO crystal is heated in an oven to prevent hygroscopic deterioration.

Figure 3 shows the setup of the experiment. F1 is the focal lens for second harmonic generation (SHG). F2 is a lens used to collimate the fundamental and SHG beams. S1, S2 are dichroic mirrors with incidence angle  $45^{\circ}$ , R = 99.5% at 1064 nm and R < 1% at 532 nm. The two beams from SHG are separated with S1 and recombined with S2. M1 and M2 are two flat mirrors with incidence angle  $45^{\circ}$ , R > 99.5% at 1064 nm. M3, M4, M5, M6 are all flat mirrors with incidence angle  $45^{\circ}$ , R > 99.5% at 532 nm. These four mirrors build up a delay line. The function of the delay line will be described below. F3 and F4 are focal lenses for THG. The fundamental beam is focused with F4 and the SHG beam is focused



**FIGURE 3** Scheme of the experiment setup of SHG in LBO and THG in CBO

with F3. Finally, a Brewster prism is employed to separate the 355 nm light beam from the 1064 nm and 532 nm light beams.

For SHG, the 1064 nm laser is focused into a 3 mm ×  $3 \text{ mm} \times 20 \text{ mm}$  LBO crystal, cut for non-critical ( $\theta = 90^{\circ}$ ,  $\varphi = 0^{\circ}$ ) type I (0 + 0 = e) phase matching [7]. The noncritical type I phase matching has some advantages over the critical type II phase matching (PM), especially for SFM. For type II phase-matching, the two fundamental beams need to have different polarization, one is ordinary (o-)polarized, while the other is extraordinary (e-)polarized. As a result, in a type II phase matching the phase and group velocities of both beams are different. Due to the walk-off, the o- and e-beams will be partially separated both temporally and spatially after propagating through the crystal. Therefore, the spatial overlap of the two fundamental beams becomes elliptical. On the contrary, such problems do not occur in type I phase-matching for SHG since the two fundamental beams have the same polarization and group velocity. The noncritical phase-matched SHG requires a crystal temperature of about 148.5 °C. In the experiment the crystal is heated in an oven, whose temperature is controlled with an accuracy of 0.1 °C. Under optimized experimental conditions, with a pumping power of 30 W at 1064 nm, a 16.3 W of SHG in the green can be obtained with a beam quality factor of  $M^2 \sim 1.3$ .

In the experiment the fundamental and the second harmonic beams are focused separately (Fig. 3). There are two reasons for this: first, the pulse widths of the two beams are all about 20 ps. The time delay due to group velocity mismatch (GVM) in CBO is 2.5 ps, compared with 0.6 ps in LBO. Such a GVM would reduce the efficiency of THG. To compensate the effect of GVM, one of the beams is intentionally delayed by a delay line when they enter the CBO crystal. The separated focusing also allows us to adjust the focus points of the two beams and control the spot size freely.

## 4 Results and discussion

We measured the output power of THG as a function of the input power of the fundamental beam at 1064 nm. Figure 4 shows the output power of the UV 355 nm output at various pumping powers of the fundamental laser. The high-

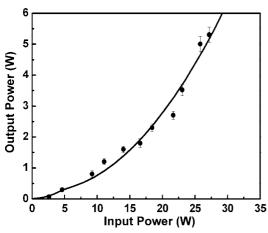


FIGURE 4 THG output power at 355 nm as a function of the total power of the fundamental and the SHG power

est output of 5.4 W is obtained at a pump power of  $\sim$  27 W. It is found that the output of THG is not sensitive to the operation temperature. When the temperature of CBO changes by  $\pm 0.5$  °C, the THG efficiency only changes by  $\sim 10\%$ .

The efficiency of the THG also depends on the temporal and spatial overlap of the light pulses, the pulse powers of the fundamental and the green SHG radiation, and the power ratio of the two beams. The temporal overlap is adjusted by changing the optical path in the delay line, while the spatial overlap mainly depends on the confocal parameters and the position of the beam waists  $w_1$  and  $w_2$  of the fundamental and SHG beams, which can be adjusted by the focal lengths and the positions of the lenses. Theoretically, the optimum situation is achieved if the input photons at 532 nm and 1064 nm are 1:1 matched. It requires a power ratio of 2:1. However, due to dispersion, it is not possible for the two beams to spatially match each other completely. At the same time, GVM causes a temporal mismatching and a pre-delay is introduced. Therefore, the optimum parameters are usually determined by experiment. It is found that the best efficiency is achieved for  $w_1 = w_2 \approx 90 \,\mu\text{m}$ , and the power ratio is  $\sim 1:1$  under the conditions of our experiment. The highest THG efficiency is  $\sim 20\%$  from the input fundamental beam. Further reducing the waists of the beams does not raise the efficiency. Figure 5

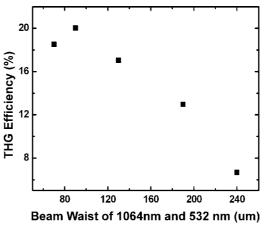


FIGURE 5 THG efficiency as a function of the beam waist of 532 nm and 1064 nm. The sizes of the two beams are the same

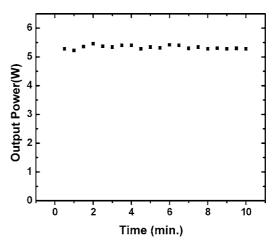


FIGURE 6 Short-term stability of THG output power 355 nm within 10 min

presents the THG efficiency with different beam waists of 1064 nm and 532 nm with a power ratio of 1:1. The sizes of the two beams remains the same. As we have mentioned above, the surfaces of the CBO remain uncoated and the total reflectivity of two the surfaces is measured to be about 7% at 1064 nm and 532 nm. So the actual THG efficiency is  $\sim$  21%. On using an anti-reflection coating for the CBO at 1064 nm, 532 nm and 355 nm, a higher output power can be expected.

To test the stabilization of the laser, we have operated it for 4.5 h continuously. The long term stability of the THG has been very good. The fluctuation of the THG within the 4.5 h test is less than 4%. The short-term stability of the THG output is also excellent. Figure 6 shows the stability within 10 min after 1 h of operation. During the operation, the power meter showed that the 355 nm power was 5.2 W–5.4 W with a fluctuation of less than 4%. The long-term stability of the system is also excellent such that the highest output power is still 5.4 W without any adjustment after two months operation.

#### 5 Summary

We have demonstrated a high average power 5.4 W picosecond laser operation at 355 nm by THG in a new non-linear crystal CBO pumped with an amplified Nd:YVO<sub>4</sub> SESAM mode-locked laser at wavelength 1064 nm. The THG output shows a high efficiency of  $\sim 20\%$  and a power fluctuation less than 4%. We have also investigated the phase

matching angle at different temperatures. Our data show that CBO is a prospective nonlinear crystal for high average power UV laser generation.

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