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β-BaB_2O_4 with special cut-angle applied to single crystal cascaded third-harmonic generation*

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High-efficiency single crystal cascaded third-harmonic generation (THG) was realized in β-BaB_2O_4 (BBO) material with special cut-angle. By analyzing effective nonlinear optical coefficient (d_{eff}) of the cascaded THG process, which was composed by type-II frequency doubling and type-I sum-frequency, the optimum phase matching (PM) direction in BBO crystal was determined to be ($\theta = 32.1°$, $\phi = 11°$). With an optimized 9-mm long sample which was processed along this direction, the highest cascaded THG conversion efficiency reached 42.3%, which is much superior to the similar components reported previously, including ADP, KDP, and $Gd_xY_{1-x}COB$ crystals.

Keywords: polarization, frequency conversion, β-$B_aB_2O_4$ crystal, nonlinear optical crystals, optical frequency converters

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1. Introduction

Now, the bulk nonlinear crystals are still the primary mediums for optical frequency up-conversions. The popular nonlinear optical (NLO) crystals include KDP, KTP, β-BaB_2O_4 (BBO), LBO, and so on. For the third-harmonic generation (THG) of near infrared laser, traditionally two NLO crystals are necessary: the first is for second-harmonic generation (SHG) and the second is for sum-frequency generation (SFG).[1–6] Recently, we developed a cascaded THG apparatus that only uses one NLO crystal. It possesses some advantages such as wide available waveband, high efficiency, and cost saving. At the same time, it has been successfully applied to several NLO materials, including KDP, ADP, and $Gd_xY_{1-x}COB$.[7–9] In present paper, for this design the BBO crystal was used as the NLO medium which brought much larger nonlinearity, and at the same time a special coated quarter-wave plate (QWP) was employedto permit completely utilizing the fundamental wave. As a result, with the optimized BBO sample, we achieved a highest conversion efficiency of 42.3%, which is much better than those previously obtained from other NLO materials.

2. Theoretical analysis of BBO crystal

2.1. Phase-matching angle

According to the crystallography tradition, the polar coordinates (θ, ϕ) are used to denote the crystal spatial direction, where θ is the included angle with the Z-axis, and ϕ is the azimuthal angle in the plane perpendicular to the Z-axis. As a trigonal symmetric, optical uniaxial crystal, the Z-axis of BBO crystal is coincident with the optical axis, and the phase matching (PM) condition is only determined by the parameter θ. Based on the Sellmeier equations of BBO crystal,[10] the PM angles θ are calculated as a function of fundament wavelengths for the SHG ($\omega + \omega \to 2\omega$) and SFG ($\omega + 2\omega \to 3\omega$) processes respectively, as shown in Fig. 1. Among all of the five PM styles, type-II SHG and type-I THG present the closest PM directions, as indicated by the red and blue lines in Fig. 1. The discrepancy of their PM angles θ is $-3.0°$ at 750 nm and 2.9° at 1200 nm, respectively. If $\Delta\theta < 3°$ is used as an application standard of one crystal cascaded THG

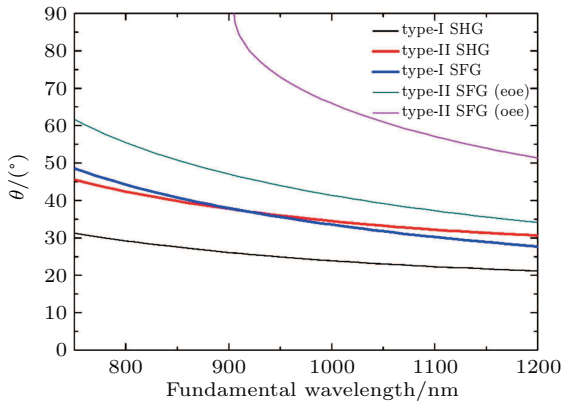


Fig. 1. (color online) Variation of phase-matching angle with the fundamental wavelength of BBO crystal.

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technique, the work bandwidth of BBO crystal is larger than 450 nm, which covers 750–1200 nm. This value is much better than that of the KDP or ADP crystal, whose work bandwidth is about 100 nm, i.e., the available fundamental wavelength is from 1000 to 1100 nm.[7,8] At the wavelength of Nd:YAG laser (1064 nm), the θ value of BBO crystal is 32.9° for type-II SHG and 31.3° for type-I SFG. It means that the θ direction of the practical processed sample should be controlled between these two angles.

2.2. Effective nonlinear optical coefficient

For type-II SHG and type-I SFG, the effective nonlinear optical coefficients ($d_{\rm eff}$) of BBO crystal can be calculated from[11]

$$d_{\rm eff,II} = d_{22}\cos^2\theta\cos 3\phi, \tag{1}$$

$$d_{\rm eff,I} = d_{31}\sin\theta - d_{22}\cos\theta\sin 3\phi, \tag{2}$$

where $d_{22} = 2.3$ pm/V and $d_{31} = -0.16$ pm/V.[12,13] When the θ angle is fixed, the largest $d_{\rm eff}$(II) is at $\phi = 0°$ direction, while the largest $d_{\rm eff}$(I) is at $\phi = 30°$ direction. Therefore, an optimized ϕ angle should be chosen to realize the largest THG output for the present one BBO crystal, cascaded frequency upconversions. Such a problem has never been met in previous investigations of KDP, ADP, and G_xY_{1-x}COB crystals, because for those crystals the largest $d_{\rm eff}$ values for SHG and SFG appear at the same spatial PM angle.[7–9] In a plane-wave fixed-field approximation, the SHG output E_2 and SFG output E_3 have the disciplines

$$E_2 \propto (d_{\rm eff,II})^2(E_1)^2, \tag{3}$$

$$E_3 \propto (d_{\rm eff,I})^2 E_1' E_1, \tag{4}$$

where E_1 is the original incident fundamental energy, and E_1' is the residual fundamental energy after the SHG process, which attends the followed SFG process. Under the ideal THG experimental conditions, $E_1' = 0.5E_2$.[14] Taking this relationship and Eq. (3) into Eq. (4), there is

$$E_3 \propto (d_{\rm eff,I})^2(d_{\rm eff,II})^4. \tag{5}$$

Utilizing Eqs. (1) and (2), the variation of $(d_{\rm eff,I})^2(d_{\rm eff,II})^4$ with ϕ angle (0–30°) can be obtained, and the result is shown in Fig. 2. In this calculation, the θ angle is fixed to be 32.1°. It can be seen that for the highest THG output the largest $(d_{\rm eff,I})^2(d_{\rm eff,II})^4$ value appears at $\phi = 11°$, not the middle angle $\phi = 15°$. At ($\theta = 32.1°$, $\phi = 11°$) direction, the $|d_{\rm eff,II}|$ and $|d_{\rm eff,I}|$ values are 1.38 and 0.79 pm/V respectively, which are much larger than those of KDP, ADP, and Gd_xY_{1-x}COB crystals (< 0.6 pm/V).

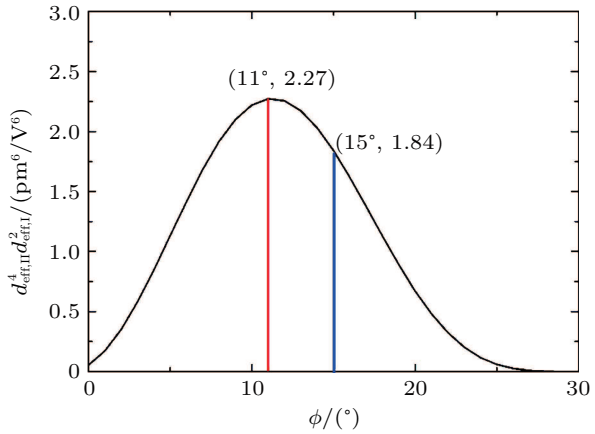


Fig. 2. (color online) Variation of $(d_{\rm eff,I})^2(d_{\rm eff,II})^4$ with the ϕ angle of BBO crystal.

2.3. Effective lengths[15]

The quasi-static interaction length $L_{\rm qs}$ can be calculated from

$$L_{\rm qs} = \tau/\nu, \tag{6}$$

where τ is the radiation pulse duration and ν is the inverse group-velocity mismatch. For BBO crystal, the parameter ν is in the order of 10^2–10^3 fs/mm, so under our experimental condition, i.e., 40 ps of FWHM (full width at half maximum) pulse duration, $L_{\rm qs}$ is 4–40 cm. The actual sample length used in our experiments is no more than 1.5 cm, so the present frequency conversion processes coincide with the quasi-static interaction condition, and the group-velocity mismatch can be ignored.

The aperture length $L_{\rm a}$ can be obtained from

$$L_{\rm a} = d_0/\tan(\rho), \tag{7}$$

where d_0 is the beam diameter and ρ is the "walk-off" angle. For type-II SHG and subsequent type-I SFG of 1 μm laser, ρ is about 4° for BBO crystal. In our experiments, d_0 is 2 mm, so $L_{\rm a}$ is calculated to be 28.6 mm and the practical sample should be shorter than this length.

For the nonlinear interaction length $L_{\rm NL}$, there is

$$L_{\rm a} = (\sigma_3\alpha_0)^{-1}, \tag{8}$$

where σ_3 is the nonlinear coupling coefficient of the generated wave which is proportional to the effective nonlinear optical coefficients $d_{\rm eff}$, and inversely proportional to the refractive index n. For KDP crystal ($d_{\rm eff} \approx 0.35$ pm/V, $n \approx 1.5$), σ_3 is about 10^{-6} V^{-1}. Therefore, for the ($\theta = 32.1°$, $\phi = 11°$) oriented BBO crystal discussed above ($d_{\rm eff} \approx 0.8$–1.4 pm/V, $n \approx 1.6$), σ_3 will be 2×10^{-6}–4×10^{-6} V^{-1}. In Eq. (8), α_0 is the wave amplitude of fundamental laser at the input surface of the crystal, which can be calculated by

$$\alpha_0 = \left[\frac{752P_1}{\pi\omega_0^2 n}\right]^{1/2}, \tag{9}$$

where P_1 is the power of the fundamental radiation and ω is the beam radius. Taking Eq. (9) into Eq. (8), the variation of L_{NL} as a function of the input power density $P_1/\pi\omega_0^2$ can be plotted, as shown in Fig. 3. In this calculation about BBO crystal, we choose an average σ_3 value of 3×10^{-6} V^{-1} and the n value of 1.6. In our experiments, the highest crystal incident energy is 4 mJ at a full temporal duration of 100 ps and a beam radius of 1 mm, so the largest input power density is 1.3×10^9 W/cm^2. From Fig. 3, it can be seen that the corresponding L_{NL} is 4.3 mm, which means that in theory the optimum crystal length will be 8.6 mm or so ($2L_{NL}$).

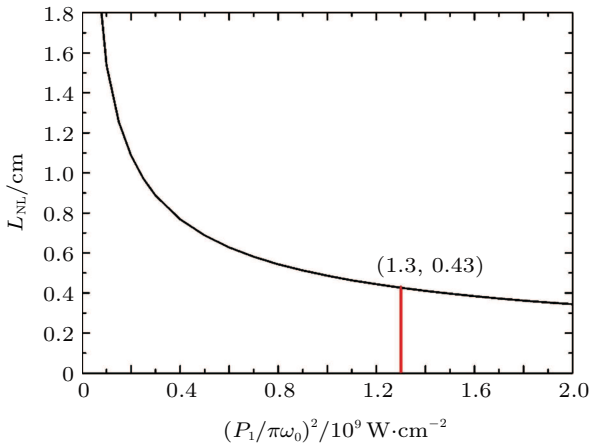


Fig. 3. (color online) Variation of the nonlinear interaction length L_{NL} with the fundamental power density of BBO crystal.

3. Experiment and discussion

The experimental equipment is similar with that used for KDP crystal. Besides different NLO crystal, an important change occurs on the 532 nm quarter wave plate (QWP). Previously, both the fundamental wave (1064 nm) and the 2ω wave (532 nm) passed through the QWP to and fro, which led to the fact that the polarization direction of fundamental wave cannot be optimized completely. Now its surface towards the NLO crystal is high-reflective (HR) coated at 1064 nm and anti-reflective (AR) coated at 532 nm, and the other surface towards the total reflection mirror M_2 is AR coated at 532 nm. The fundamental wave no longer passes though the 2ω QWP. In this way, the incident fundamental polarization can be adjusted freely, and the unfavorable fundamental polarization variation induced by the 2ω QWP is avoided, i.e., the optimal scheme (OS) is realized.[7] At the same time, the 2ω QWP is tightly attached to the mirror M_2 to decrease the optical path difference between the ω and 2ω waves.

We prepared six BBO samples for this experiment, which were cut along two directions, (32.1°, 11°) and (32.1°, 15°). For each direction, three crystal lengths were selected, 6 mm, 9 mm, and 12 mm. The cascaded THG results are shown in Fig. 4. All of the six samples exhibit efficient cascaded THG conversion with conversion efficiencies larger than 10%, which proves that the selection of θ angle from the Fig. 1 is reliable. Generally, at the same crystal length, the (32.1°, 11°) sample always presents better performance than the (32.1°, 15°) sample. For example, the largest THG output and the highest conversion efficiency of the 6 mm, (32.1°, 11°) BBO are 0.95 mJ and 38.1%, respectively, and the corresponding data of the 6 mm, (32.1°, 15°) BBO are 0.62 mJ and 20.0%. Such property proves the theoretical analysis result of Fig. 2, i.e., the (32.1°, 11°) direction possesses larger combined NLO coefficient than the (32.1°, 15°) direction for the cascaded THG process. For BBO samples at the same direction, the output energy and conversion efficiency of the 9 mm crystal are obviously superior to the results of 6 mm and 12 mm crystals. It experimentally manifests that the optimized crystal length is 9 mm or so, which is in good agreement with Fig. 3. To sum up, the best experiment data come from the (32.1°, 11°), 9 mm sample, i.e., the largest THG output of 1.17 mJ at a fundamental energy of 3.51 mJ, and the highest conversion efficiency of 42.3% at a fundamental energy of 1.02 mJ. The most inferior are from the (32.1°, 15°), 12 mm sample.

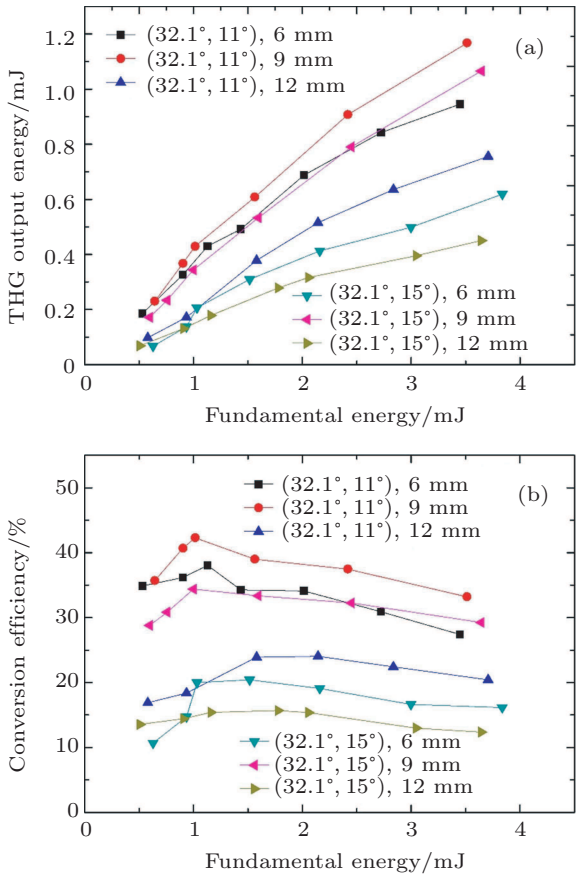


Fig. 4. (color online) Variation of (a) cascaded THG output energy and (b) conversion efficiency with the fundamental energy of different BBO samples.

4. Conclusion

BBO crystal is one of the most popular ultra-violet NLO crystals. In this paper, we realized its single block, cascaded

THG for the first time. By optimizing the crystal PM direction and the crystal length, as well as utilizing the polarization OS, the large nonlinearity of BBO crystal is fully exploited and a high THG conversion efficiency of 42.3% is obtained. Comparing with the traditional cascaded THG converter composed by two NLO crystals, the present design exhibits some advantages such as compact, cost saving, and super wide applicable waveband (>450 nm). Its work wavelength extends to 800 nm around, so efficient single crystal, THG converter maybe developed for ultrafast Ti:sapphire or diode lasers in the future. Besides the practical value, this study will also lend good design reference for other cascaded frequency upconversion processes and NLO materials.

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