Phase mismatching analysis of third-harmonic generation in BBO crystal

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Phase mismatching for third-harmonic generation (THG) in barium metaborate (BBO) crystal is investigated in detail. Upon using BBO crystal in the Type I (oo→e) scheme, in the two independent planes (principal section of the crystal and the plane normal to principal section), when the input second-harmonic beam deviates from the expected direction, phase mismatching occurs and the angle deviation produces different effects on the conversion efficiency. We numerically simulate these two cases of phase mismatching and identify the relation between the conversion efficiency and the deviation angle in the air. The computational results indicate that the phase matching tolerance in the principal section is 0.2° (in the air), while the phase matching tolerance in the plane normal to principal section is 4.5° (in the air). After conducting a lab experiment, the results agree perfectly with the computational results, indicating that the deviation angle in the principal section has a greater effect on the conversion efficiency.

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Barium metaborate (BBO) crystal is a negative uniaxial crystal, whose large nonlinear efficiency and damage threshold make it a widely used material in many fields, including ultrashort pulse measuring, optical parametric process, photon metrology, etc.[1−5] Improving the frequency conversion efficiency to promote the authenticity of the signal has been the general focus of most research; of these, the key issue is the phase matching angle optimum design[3,6]. However, other aspects should also be considered so as to ensure high frequency conversion efficiency in practice. Likewise, there is a need to ensure that the beams enter the cutting crystal in the expected direction. If the input second-harmonic beam deviates from the expected direction in two independent planes, namely, the principal section of the crystal and the plane normal to principal section, phase mismatching is likely occur and the influence of the degree of deviation angle could have different results in the two cases. Based on momentum conservation and the refractive index ellipsoid, phase mismatching for type I (oo→e) third-harmonic generation (THG) in BBO crystal is investigated to illustrate the issue. In this letter, we aim to identify how the angle deviation affects the conversion efficiency and determine the adjusting accuracy in the two abovementioned cases.

We analyze the phase mismatching of type I THG[7] in BBO crystal with a size of $4 \times 4 \times 2$ (mm). Firstly, the wave vectors of the three frequencies involved must have the same direction, and the phase matching condition must be completely satisfied as

$$\Delta \boldsymbol{k} = \boldsymbol{k}_{3\omega} - \boldsymbol{k}_{2\omega} - \boldsymbol{k}_{1\omega} = 0, \qquad (1)$$

where $\boldsymbol{k}_{i\omega}(i = 1,2,3)$ are the wave vectors of the fundamental, second-harmonic, and third-harmonic frequencies, respectively, as indicated by

$$\boldsymbol{k}_{1\omega} = 2\pi n_{\mathrm{o}}(1\omega)/\lambda_{1\omega},$$
$$\boldsymbol{k}_{2\omega} = 2\pi n_{\mathrm{o}}(2\omega)/\lambda_{2\omega} = 4\pi n_{\mathrm{o}}(2\omega)/\lambda_{1\omega},$$
$$\boldsymbol{k}_{3\omega} = 2\pi n_{\mathrm{e}}(3\omega, \theta)/\lambda_{3\omega} = 6\pi n_{\mathrm{e}}(3\omega, \theta)/\lambda_{1\omega}.$$

Since the three frequencies are collinear, the phase matching condition can be written as

$$3n_{\mathrm{e}}(3\omega, \theta) - 2n_{\mathrm{o}}(2\omega) - n_{\mathrm{o}}(1\omega) = 0, \qquad (2)$$

where θ is the phase matching angle, the subscripts o and e refer to ordinary and extraordinary beams, and $n_{\mathrm{e}}(3\omega, \theta) = 1/\sqrt{\cos^2(\theta)/n_{\mathrm{o}}^2(3\omega) + \sin^2(\theta)/n_{\mathrm{e}}^2(3\omega)}$. BBO crystal is negative uniaxial ($n_{\mathrm{e}} < n_{\mathrm{o}}$).

Phase mismatching is induced when the fundamental frequency is incident on the BBO crystal surface vertically and the incident angle of the second-harmonic frequency changes. We calculate the phase mismatching when the direction of the second-harmonic incident frequency departs from fundamental beam in two vertical planes: one is the principal section of the crystal, and the other is vertical to the principal section.

Firstly, we consider the departure angle ϕ in the principal section of the crystal. When $\boldsymbol{k}_{2\omega}$ departs from the original direction, the magnitude remains the same. Both the direction and the magnitude of sum vector ($\boldsymbol{k}_{\mathrm{sum}} = \boldsymbol{k}_{2\omega} + \boldsymbol{k}_{1\omega}$) of 1ω and 2ω are changed. As such, the magnitude and direction of sum vector can be given by

$$k_{\mathrm{sum}} = \sqrt{k_{1\omega}^2 + k_{2\omega}^2 + 2k_{1\omega}k_{2\omega}\cos\phi}, \qquad (3)$$

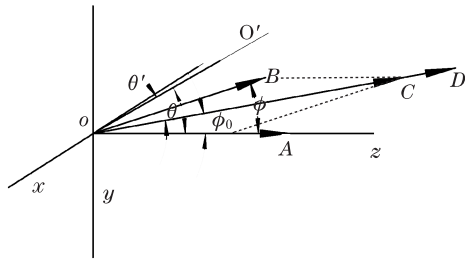$$\phi_0 = \arcsin(k_{2\omega}\sin\phi/k_{\mathrm{sum}}), \qquad (4)$$

Fig. 1. Wave vectors in the principal section of crystal.

where ϕ is the departure angle (see Fig. 1), which can either be positive or negative. It is clear that the magnitude of the sum vector is smaller than $\boldsymbol{k}_2$ departing from $\boldsymbol{k}_1$; thus the vector of sum frequency should be smaller to meet the phase matching condition. The extraordinary refraction of 3ω light at ϕ_0 can be written as

$$n_{\mathrm{e}}(3\omega, \theta') = 1/\sqrt{\cos^2(\theta')/n_{\mathrm{o}}^2(3\omega) + \sin^2(\theta')/n_{\mathrm{e}}^2(3\omega)}, \quad (5)$$

where $\theta' = \theta - \phi_0$ (Fig. 1). We can then rewrite the phase mismatching as

$$\Delta\boldsymbol{k} = \boldsymbol{k}_{3\omega} - \boldsymbol{k}_{2\omega} - \boldsymbol{k}_{1\omega} = \boldsymbol{k}_{3\omega} - \boldsymbol{k}_{\mathrm{sum}}, \quad (6)$$

where its magnitude is obtained through

$$\Delta k = k_{3\omega} - k_{\mathrm{sum}}$$
$$= \frac{2\pi}{\lambda_{1\omega}} \left[3/\sqrt{\cos^2(\theta - \phi_0)/n_{\mathrm{o}}^2(3\omega) + \sin^2(\theta - \phi_0)/n_{\mathrm{e}}^2(3\omega)} \right.$$
$$\left. -\sqrt{n_{\mathrm{o}}^2(1\omega) + 4n_{\mathrm{o}}^2(2\omega) + 4n_{\mathrm{o}}(1\omega)n_{\mathrm{o}}(2\omega)\cos\phi} \right]. \quad (7)$$

When $\phi = 0$, Eq. (7) is reduced to Eq. (2).

In THG, the intensity of sum frequency $I(3\omega)$ and Δk has the relationship[8]:

$$I(3\omega) \propto \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2} L^2, \quad (8)$$

where L is the generation length, which has a relationship with BBO crystal thickness d as indicated by

$$L = d/\cos(\phi_0). \quad (9)$$

We can identify the relation between $I(3\omega)$ and ϕ from Eqs. (7) – (9). Considering refraction in BBO,

Fig. 2. Theoretical relation between THG efficiency and the deviation angle (in the air) of BBO and the experimental results (ϕ in principal section of crystal). The FWHM of the curve is 0.2°. FWHM: fnu-width at half-maximum.
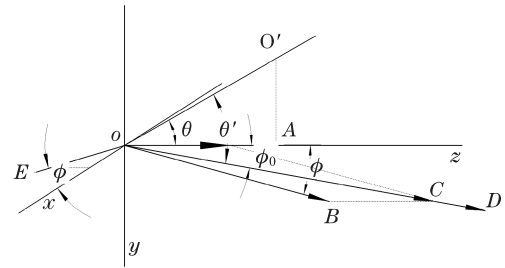
Fig. 3. Wave vectors in the plane normal to the principal section of crystal.

the departure angle in the crystal is obtained by $\phi = \arcsin[\sin(\psi)/n_{\mathrm{o}}(2\omega)]$, where ψ is the incident angle in the air; the refractive index is listed in Table 1.[9] The simulation result is shown in Fig. 2, and the full-width at half-maximum (FWHM) of the curve is 0.2°.

Secondly, we consider the departure angle ϕ in the plane normal to the principal section of crystal. When $\boldsymbol{k}_{2\omega}$ departs from the original direction, Eqs. (3) and (4) are quoted to describe the sum vector $\boldsymbol{k}_{\mathrm{sum}} = \boldsymbol{k}_{2\omega} + \boldsymbol{k}_{1\omega}$. However, the angle between the two extraordinary axes $\theta' = \angle COO'$ is different from the case above; this is formulated as

$$\theta' = \arccos[\cos\theta\cos(\phi_0)], \quad (10)$$

and

$$\Delta k = k_{3\omega} - k_{\mathrm{sum}} = \frac{2\pi}{\lambda_{1\omega}} \left\{ 3/\sqrt{\cos^2\theta\cos^2(\phi_0)/n_{\mathrm{o}}^2(3\omega) + [1 - \cos^2\theta\cos^2(\phi_0)]\sin^2(\theta - \phi_0)/n_{\mathrm{e}}^2(3\omega)} \right.$$
$$\left. -\sqrt{n_{\mathrm{o}}^2(1\omega) + 4n_{\mathrm{o}}^2(2\omega) + 4n_{\mathrm{o}}(1\omega)n_{\mathrm{o}}(2\omega)\cos\phi} \right\}. \quad (11)$$

As the vector $\boldsymbol{k}_{2\omega}$ departs from the original direction by an angle ϕ, the electric field amplitude of 2ω light along the ordinary axis E_{o} is reduced to $E_{\mathrm{o}} = E_{2\omega}\cos(\phi)$, and the intensity of 2ω light beam for THG is reduced to $I_{\mathrm{o}} = I_{2\omega}\cos^2(\phi)$. Thus, Eq. (8) should be written as

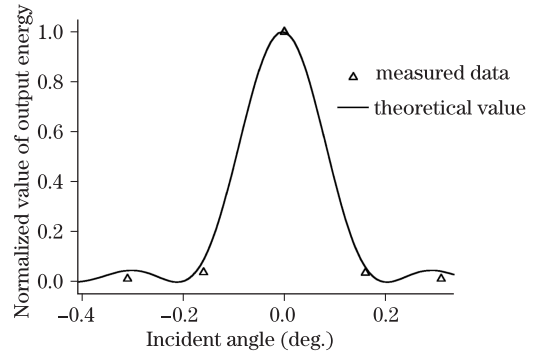$$I(3\omega) \propto \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2} \cos^2(\phi)L^2. \quad (12)$$

We can identify the relation between $I(3\omega)$ and ϕ from Eqs. (11) and (12). Considering refraction in BBO, the departure angle in the crystal is

$$\phi = \arcsin[\sin(\psi)/n_{\mathrm{o}}(2\omega)],$$

where ψ is the incident angle in the air. The simulation result in Fig. 4 shows that the FWHM of the curve is 4.5°.
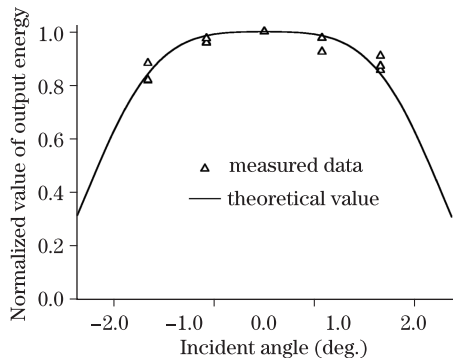
Fig. 4. Theoretical relation between THG efficiency and the deviation angle (in the air) of BBO and the experimental results (ϕ in the plane normal to principal section of crystal).
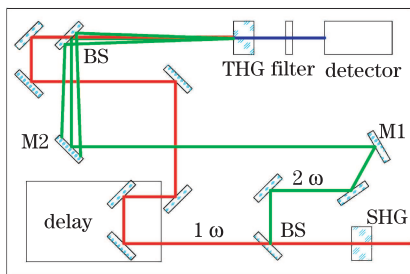

Fig. 5. Schematic diagram of the experimental setup. SHG: second-harmonic generation; BS: beam splitter; M1, M2: mirrors.

We carried out the experiment (see Fig. 5) on the laser SGR-10 of Beamtech Co., Ltd., whose pulse width and wavelength were 10 ns and 1053 nm, respectively. The fundamental frequency (1053 nm) was incident on BBO crystal normally, and mirrors were rotated to change the incident angle of the second-harmonic ψ mentioned above. The calorimeter measured the third-harmonic energy E at picojoules level.

Figures 2 and 4 show the comparison between experimental results (Tables 2 and 3) and computational results when the data are normalized by accounting the energy at angle $\psi = 0°$ as 1. All the measured data fit the computational results well, indicating that the presented theoretical models are applicable.

In conclusion, the numerical simulations indicate that the phase matching tolerance in the principal section is $0.2°$ (in the air), while the phase matching tolerance in the plane normal to the principal section is $4.5°$ (in the air). Owing to the greater effect of the deviation angle in the principal section on the conversion efficiency, it is necessary to design a fine tuning structure to control the angle deviation in the principal section of the BBO

Table 1. Refractive Index of BBO

Wavelength (nm)	n_{o}	n_{e}
1053	1.655	1.539
526	1.675	1.555
351	1.707	1.577

Table 2. Departure Angle ϕ in the Principal Section of Crystal

Angle (deg.)	−0.77	−0.31	0	0.31	0.77
Energy (nJ)	0	0.015	1.12	0.015	0

Table 3. Departure Angle ϕ Normal to the Principal Section of Crystal

Angle (deg.)	Energy (nJ)		
	Input 1	Input 2	Input 3
−1.55	0.893	0.788	0.912
−0.77	0.986	0.926	1.066
0	1.012	0.965	1.113
0.77	0.935	0.893	1.086
1.55	0.882	0.826	1.012

crystal precisely. As the phase matching tolerance in the plane normal to the principal section is relatively large, the angle deviation in the principal section of the BBO crystal does not need to be controlled precisely. On the other hand, this can be used to substitute the noncollinear phase matching under other conditions[2,5].

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