PACS numbers: 42.65.An; 42.64.Hw; 42.70.Mp
DOI: 10.1070/QE2010v040n09ABEH014310

Angular phase-matching bandwidths in biaxial nonlinear crystals for frequency converters

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Abstract. **It is shown that the angular phase-matching bandwidths in biaxial nonlinear crystals in the general case must be calculated in the coordinate system in which the angular deviations of the crystal and the laser beam divergence are determined consistently. The angular phase-matching bandwidths in this coordinate system may considerably differ from the conventionally determined values. The optimum orientation of the coordinate system for determining the angular phase-matching bandwidths is found. It is established that, in the general case in biaxial crystals, as in uniaxial ones, phase matching is always angle-critical along one coordinate and noncritical along the other and that it is possible to realise angle-noncritical phase matching of the fourth order.**

Keywords: *biaxial crystals, frequency conversion, phase matching, angular phase-matching bandwidths, noncritical phase matching.*

1. Introduction

The nonlinear and phase-matching parameters are very important for the problems of nonlinear-optical frequency conversion in crystals [1]. While the nonlinear parameters (effective nonlinearity coefficient) determine the nonlinear response at each point of the medium length, the phase-matching parameters determine both the phase-matching (coherent accumulation) condition and the limitations of the conversion efficiency. In particular, the frequency and spatial dispersion, as well as the temperature dependence of refractive indices, determine the spectral, angular, and temperature widths of phase matching. The method of determining the angular phase-matching bandwidths, which was initially developed and used for uniaxial crystals, was completely transferred to biaxial crystals. Some specific features of biaxial crystals show that this method cannot correctly describe the properties of crystals for nonlinear optical frequency conversion. In the present work, we choose a coordinate system in which the angular phase-matching bandwidths, laser beam divergence, and the angular misalignment of the crystal are determined consistently. It is shown that the angular phase-matching bandwidths in this coordinate system differs from those determined conventionally. It is also demonstrated that, in the most general case, the phase matching in a biaxial crystal is always critical along one angular coordinate and noncritical (both of the second and higher orders) along the other.

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Received 11 March 2010; revision received 7 June 2010
Kvantovaya Elektronika **40** (9) 822–827 (2010)
Translated by M.N. Basieva

2. Coordinate systems for description of the crystal and radiation properties

The phase-matching direction in all types of crystals is determined in the polar coordinate system (crystal-optic) by two angles, φ and θ (Fig. 1). The angle φ determines the rotation of plane II with respect to the initial position – basic plane I (xz plane), while the angle θ determines the direction of wave vectors $\boldsymbol{k}_i$ of interacting waves in plane II which satisfy the phase-matching condition $\Delta \boldsymbol{k} = \boldsymbol{k}_3 - \boldsymbol{k}_2 - \boldsymbol{k}_1 = 0$. Hereinafter, we restrict ourselves to the consideration of scalar phase matching.

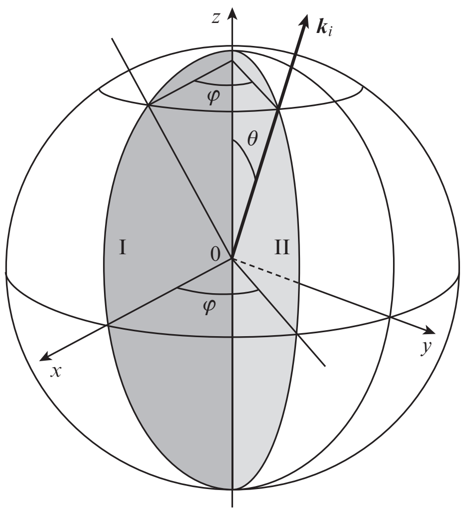

Figure 1. Crystal coordinate system.

For uniaxial crystals, the angle θ, which determines the phase-matching direction, is the cone angle of the cone of phase-matching directions, whose axis is the z axis. For biaxial crystals, the phase-matching directions form conical surfaces of the fourth order [2–5], whose axis is one of the crystal axes.

The angular phase-matching bandwidths $2\Delta\theta$ and $2\Delta\varphi$ are determined on a unit length (traditionally, L = 10 mm) of the crystal as an allowable deviation from the phase-matching direction at which the frequency conversion efficiency decreases by a given factor. In the existing method of describing the crystal properties, one of the angles determining the angular phase-matching bandwidths is the angle θ corresponding to the allowable deviation in plane II, in which the radiation beam axis lies. The angle φ (at a fixed angle θ) determines the allowable deviation on the surface of the cone whose rotation axis is the z axis (Fig. 1).

The laser beam divergence in the general case is determined in two mutually orthogonal planes which intersect along a line coinciding with the direction of axes of the interacting beams. Let us call this coordinate system the laser beam coordinate system. In this case, only the deviation $\Delta\theta$ from the angle θ in plane II (angular phase-matching bandwidth $2\Delta\theta$) (Fig. 2) can be correlated with the beam divergence. The deviation $\Delta\varphi$ from the angle φ does not correspond to the divergence along the other coordinate. This was not important for uniaxial crystals (except for the case when a beam was focused into a crystal by a cylindrical lens), in which the phase matching is noncritical with respect to the angle φ and has an angular width much larger than the angular width $2\Delta\theta$ of critical phase matching. The situation in biaxial crystals is different and, as will be shown below, there is a considerable difference in the angular phase-matching bandwidths, which can lead to considerable errors in subsequent calculations.

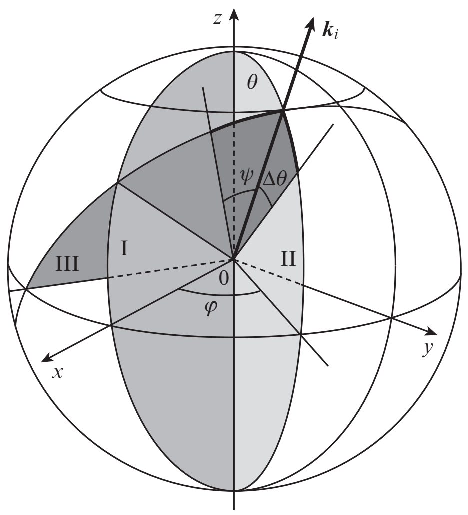

Figure 2. Laser-beam coordinate system.

To consistently describe the variations in the crystal properties and laser beam divergence, it is necessary to consider detuning in the plane orthogonal to plane II (plane III in Fig. 2). The line of intersection of planes II and III coincides with the laser beam axis. We will denote the angular deviation from the phase-matching direction in plane II as $\Delta\psi$, i.e., pass to the Euler coordinate system. While the angle φ does not change with changing the angle θ, a change in the angle ψ leads to simultaneous change in θ and φ.

There is no point in passing to the description of the crystal properties and phase-matching directions in the laser beam coordinate system. It is sufficient to determine the relation between ψ and the angles θ, φ and continue to describe the crystal properties in the crystal optic coordinate system. The expressions describing this relation have the form

$$\cos\theta = \cos\theta_0 \cos\psi, \tag{1}$$

$$\cos\varphi = \frac{\cos\varphi_0 \sin\theta_0 \cos\psi - \sin\varphi_0 \sin\psi}{\sqrt{1-\cos^2\theta_0 \cos^2\psi}}. \tag{2}$$

Here, φ_0 and θ_0 are the initial values of the angles φ and θ characterising the phase matching direction.

At $\theta_0 = 90°$, from (2) one can find the obvious relation $\varphi = \varphi_0 + \psi$, while at $\theta_0 = 0$ in the general case ($\psi \neq 0$) we have $\varphi = \varphi_0 \pm 90°$.

The difference in the angular phase-matching bandwidths $2\Delta\varphi$ and $2\Delta\psi$ is caused by the nonlinear dependence of φ on ψ. The dependence of angles θ and φ on ψ is nonlinear and determined by the angle θ_0. Figure 3 shows the dependences of angles φ and θ on ψ at different angles θ_0. In the region of small ψ (corresponding to typical angular phase-matching bandwidths of crystals), the value $d\varphi/d\psi$ changes from 1.0 (at $\theta_0 = 90°$) to infinity (at $\theta_0 = 0$). In a particular case of $\theta_0 = 90°$, the relation between φ and ψ is linear, which yields $2\Delta\varphi = 2\Delta\psi$. It can be assumed that this relation is also valid at $\theta_0 \geq 70°$. The angular phase-matching bandwidth $2\Delta\psi$ is determined by the character of the angular distribution of the cone of phase-matching directions, which is described for biaxial crystals by a fourth-order equation, and by the birefringence.

All the said in this section is true for biaxial crystals of all point symmetry groups.

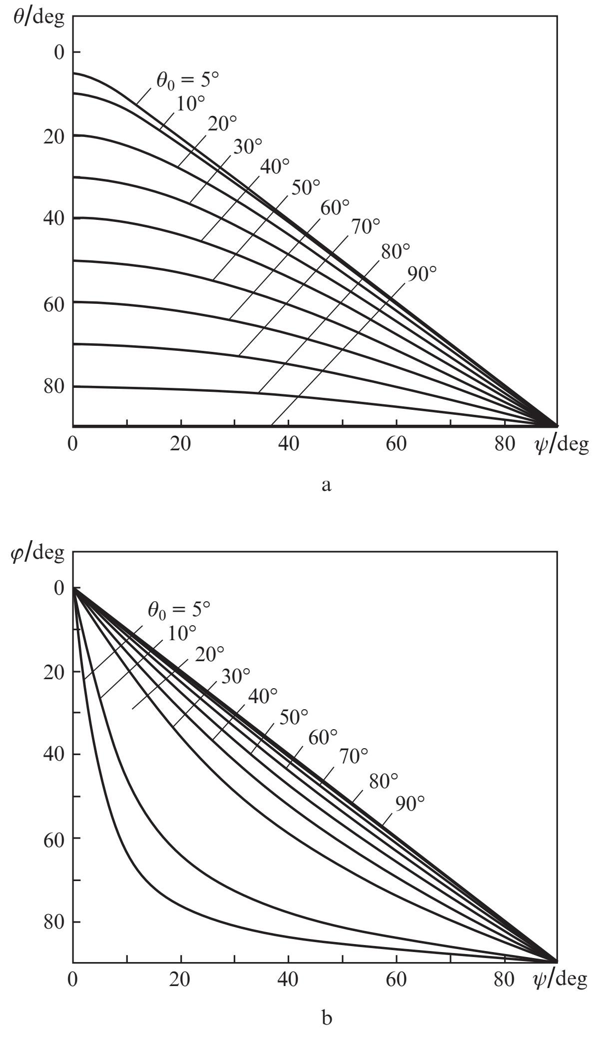

Figure 3. Dependences of the θ (a) and φ (b) angles on ψ at different θ_0.

3. Phase-matching properties of LBO crystal

Let us consider the said above on the example of the third harmonic generation (THG) in an LBO crystals in the case of incident radiation with the wavelength λ = 1.064 μm (the process $\omega + 2\omega$), which allows us to demonstrate all the main regularities and specific features of determining the angular phase-matching bandwidths in different coordinate systems. The diagram of phase-matching directions for this case is shown in Fig. 4 (curves AB and CD). In this frequency conversion regime, two interaction types are realised, ssf (slow-slow-fast) and fsf (fast-slow-fast). Points A, B, C, and D denote the phase-matching directions in the main crystal planes.

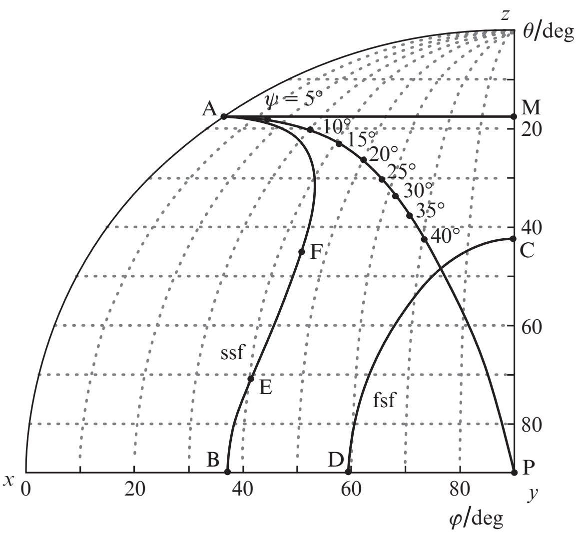

Figure 4. Phase-matching directions and coordinate system for measuring the angular deviations.

Figure 4 also presents the projections of the coordinate systems on the phase-matching directions in the xz plane (point A), in which the angles φ (at the AM arc in the plane parallel to the xy plane) and ψ (at the AP arc in the plane perpendicular to the xz plane at φ = 0) are determined.

At the point $\varphi_0 = 0$, $\theta_0 = 17°37'12''$ (point A), the plane in the laser beam coordinate system in which the ψ angle is measured (plane III in Fig. 2) is a tangent to the cone of phase-matching directions, which allows us to expect a larger angular phase-matching bandwidth in this plane. However, due to the large value $d\varphi/d\psi = 3.3$, a small deviation in the angle ψ from the phase-matching direction leads to significant changes in the angle φ and, as a result, to a large deviation from the phase-matching direction. Some values of angles ψ measured from the initial direction $\varphi_0 = 0$, $\theta_0 = 17°37'12''$ in the laser beam coordinate system are given in Fig. 4 on the AP arc.

Figure 5 shows the dependences of the angular phase-matching bandwidths $2\Delta\varphi$ (determined conventionally and given in the reference literature) and $2\Delta\psi$ along the phase-matching direction on the angle φ for ssf and fsf interactions in an LBO crystal upon THG of radiation with λ = 1.0642 μm. Hereinafter, all the angular phase-matching bandwidths are determined inside a 10-mm-long crystal at the half-intensity level. At $\varphi_0 = 0$, $2\Delta\varphi$ and $2\Delta\psi$ for the ssf interactions differ

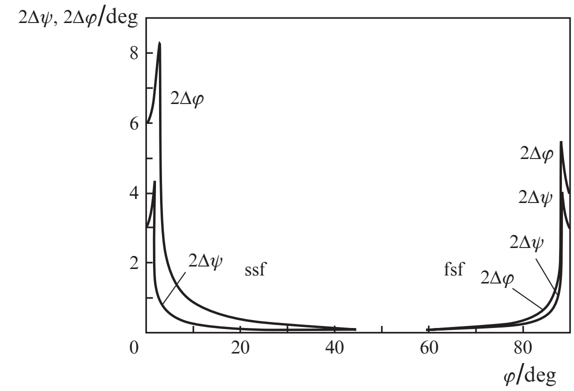

Figure 5. Dependences of the angular phase-matching bandwidths for the ssf and fsf interactions.

by a factor of two ($2\Delta\varphi = 5°56'24''$ and $2\Delta\psi = 2°57'7''$), and these difference monotonically decreases with increasing the angle θ. Similar dependences for the ssf interaction in the region of $\theta_0 = 90°$ are shown in Fig. 6. The loop-like behaviour of the dependences corresponds to the two branches of the phase-matching curve (AF and FB arcs in Fig. 4). Here, one also observes a considerable difference between the widths $2\Delta\varphi$ and $2\Delta\psi$. At $\theta_0 = 90°$ (in the xy plane), we have $2\Delta\varphi = 2\Delta\psi$. A similar relation is also valid for the fsf interaction (Fig. 5). However, the large θ_0 in the yz plane ($\theta_0 = 42°11'19''$) compared to the value in the xz plane results in a smaller difference between the $2\Delta\varphi$ and $2\Delta\psi$ widths. In the yz plane, the angular widths are $2\Delta\varphi = 3°53'47''$ and $2\Delta\psi = 2°55'12''$.

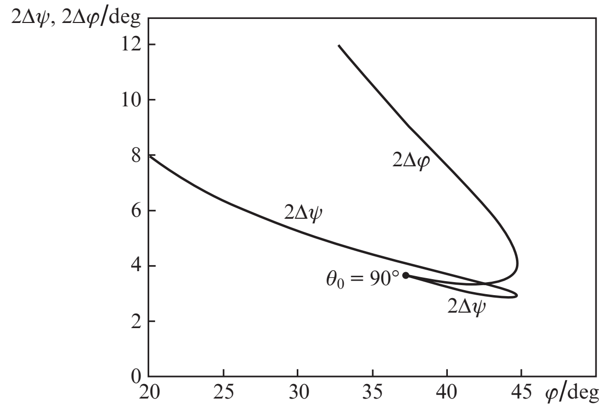

Figure 6. Dependences of the angular phase-matching bandwidths for ssf interaction in the region of $\theta_0 = 90°$.

In uniaxial crystals, it is impossible to obtain phase-matching at $\theta_0 = 0$ (along the z axis), which can occur only in the case of zero dispersion of the medium. In biaxial crystals, an analogue of this situation is the impossibility of phase matching along the optical axes, while phase matching at $\theta_0 = 0$ occurs both in the general case of sum-frequency generation and in the case of second harmonic generation, which is clearly seen from generalised diagrams of phase-matching directions [3–5]. One can easily see that, when the phase-matching angle is $\theta_0 = 0$, the conventionally determined angular phase-matching bandwidth is $2\Delta\varphi = 360°$ and $2\Delta\theta$ depends on the angle φ. Figure 7 shows the relative conversion efficiency distribution

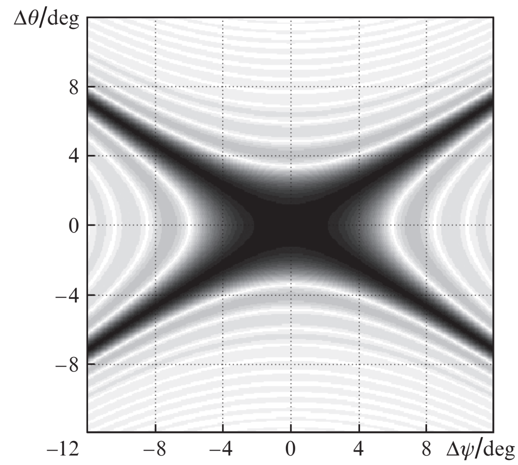

Figure 7. Relative conversion efficiency distribution for the ssf interaction in an LBO crystal for THG of radiation with $\lambda = 0.9731$ μm ($\varphi = 0$, $\theta = 0$). Here and in Figs 8, 9, the white and black colours correspond to the zero and maximum values, respectively.

(in the given field approximation) for the ssf interaction in the case of THG of radiation with $\lambda = 0.9731$ μm (the phase-matching condition direction coincides with the direction of the z axis). For this case, the angular phase-matching bandwidths in the main planes are $2\Delta\psi = 3°28'54'$ and $2\Delta\theta = 2°5'41''$. In these planes, one observes noncritical second-order phase matching ($\mathrm{d}\Delta k/\mathrm{d}\theta = 0$ and $\mathrm{d}^2\Delta k/\mathrm{d}\theta^2 \neq 0$).

4. Minimum and maximum angular phase-matching bandwidths

When choosing the crystal length, one compares the angular phase-matching bandwidths with the beam divergence. For axially symmetric beams, the minimum of the two angular phase-matching bandwidths should be chosen for this comparison. The widths $2\Delta\theta$ and $2\Delta\psi$ (or $2\Delta\varphi$) determined for biaxial crystals show that, in the general case (which is usually called the critical phase matching), the phase-matching cone in the considered direction (φ_0, θ_0) is oriented at an angle γ to

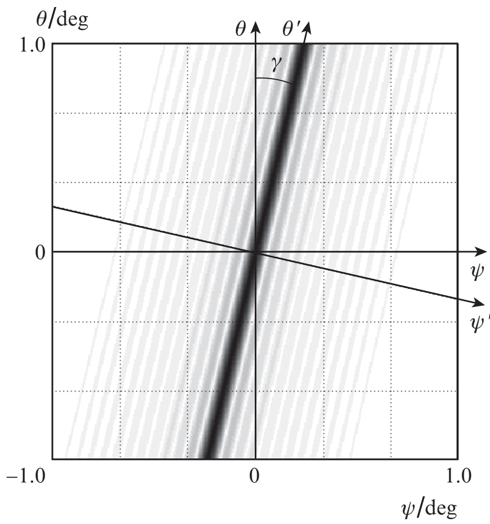

Figure 8. Relative conversion efficiency distribution for the ssf interaction at $\varphi_0 = 40.1°$, $\theta_0 = 71.52°$.

the coordinate system $\theta\varphi$. As an example, Fig. 8 shows the relative frequency conversion efficiency for the ssf interaction in the laser beam coordinate system ($\psi\theta$) in the direction $\varphi_0 = 40.1°$, $\theta_0 = 71.52°$ (point E in Fig. 4) for THG of radiation with $\lambda = 1.0642$ μm. It is seen that the angular phase-matching bandwidths must be determined in the laser beam coordinate system rotated by the angle γ [$\tan\gamma = (\mathrm{d}\Delta k/\mathrm{d}\psi)/(\mathrm{d}\Delta k/\mathrm{d}\theta)$] around the axis coinciding with the phase-matching direction. Then, the angular detuning ψ will be determined in the plain tangent to the phase-matching cone. The initial laser beam coordinate system ($\psi\theta$) is transformed to the $\psi'\theta'$ coordinate system using the standard procedure of coordinate system rotation. In this case, we can determine the minimum and maximum angular phase-matching bandwidths. It is the minimum found angular phase-matching bandwidth that must be compared with the beam divergence. The dependences of the optimal rotation angle γ along the phase-matching curve on φ for the ssf and fsf interactions are shown in Fig. 9. The zero angle γ corresponds to the orientation parallel to the z axis. For the positive direction, we take the clockwise rotation (for an observer in the coordinate system centre). The points depicted in Fig. 9 correspond to phase matching in the xy plane ($\theta_0 = 90°$). The angle γ also changes correspondingly to the behaviour of the phase-matching curve. In the considered case, this angle takes both positive and negative values for the ssf interaction and only negative values for the fsf interaction. Figure 9 also shows the dependences of the orientation angle of intrinsic polarisations δ along the phase-matching curve on φ (these dependences are given for the second-harmonic wavelength). It is seen that the angles γ and δ do not coincide and vary within different ranges. In particular, for the fsf interaction, the angle δ does not exceed 10° and the angle γ changes from 0 to –90°.

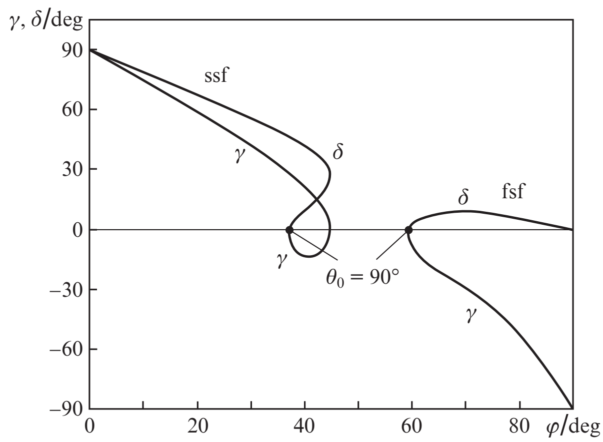

Figure 9. Orientation angles of intrinsic polarisations δ and optimal rotation angles γ along the phase-matching curve.

Figure 10 shows the dependences of the angular phase-matching bandwidths on φ for both interaction types – the minimum and maximum phase-matching bandwidths ($2\Delta\theta'$ and $2\Delta\psi'$) for each phase-matching direction. The loop shape of dependences for the ssf interaction corresponds to the two branches of the phase-matching curve (arcs AF and FB in Fig. 4). Along the phase-matching curve, the angular width of angle-critical phase matching $2\Delta\psi'$ lies within the range of $2.9'$–$4.2'$ for the ssf interaction and within the range of

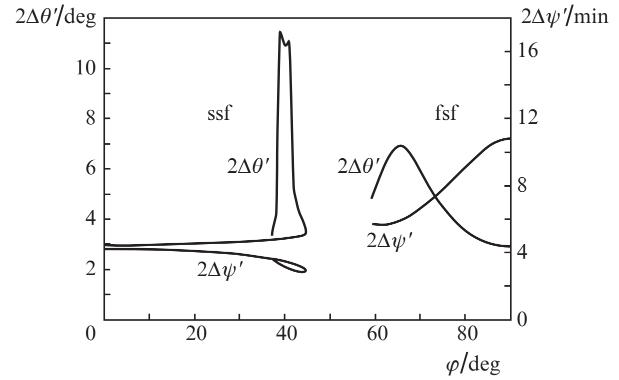

Figure 10. Minimum and maximum angular phase-matching bandwidths.

5.7′–10.8′ for the fsf interaction. The angular width of angle-noncritical phase matching $2\Delta\theta'$ lies within the range 3°–11.5° for the ssf interaction and within the range of 3°–7° for the fsf interaction. For both interaction types, the maximum angular phase-matching bandwidths $2\Delta\theta'$ are reached out of the main planes of the crystal.

These results show that phase matching in biaxial crystals in the most general case is always critical along one angular coordinate and noncritical along the other. This is necessary to emphasise because phase matching in numerous works is traditionally determined as noncritical only in the main planes.

5. Angle-critical and noncritical phase matching of different orders

In uniaxial crystals, the normal surfaces for refractive indices of interacting waves are described by a second-order equation, and phase matching in the general case is critical with respect to the angle θ ($\mathrm{d}\Delta k/\mathrm{d}\theta \neq 0$) and noncritical at $\theta_0 = 90°$ ($\mathrm{d}\Delta k/\mathrm{d}\theta = 0$ and $\mathrm{d}^2\Delta k/\mathrm{d}\theta^2 \neq 0$). The width of angle-noncritical phase matching depends on the crystal length as $2\Delta\theta \propto 1/\sqrt{L}$. This width is larger than for critical phase matching.

In biaxial crystals, the normal surfaces for the refractive indices for interacting waves n_s and n_f in the general case are described by fourth-order equations [1]. Therefore, as will be shown below, it is in principle possible to obtain angle-noncritical phase matching up to the fourth order inclusive, i. e., when $\mathrm{d}^n\Delta k/\mathrm{d}\alpha^n = 0$ ($n = 1-3$), $\mathrm{d}^4\Delta k/\mathrm{d}\alpha^4 \neq 0$, where $\alpha = \theta'$ or $\alpha = \psi'$. In the main planes of a biaxial crystal, the n_s and n_f components are described by second-order equations. Hence, at small angular deviations from the main planes, the angle-noncritical phase matching will have the second order ($\mathrm{d}\Delta k/\mathrm{d}\alpha = \mathrm{d}^3\Delta k/\mathrm{d}\alpha^3 = \mathrm{d}^4\Delta k/\mathrm{d}\alpha^4 = 0$, $\mathrm{d}^2\Delta k/\mathrm{d}\alpha^2 \neq 0$).

According to the standard determination, in the case of angle-noncritical phase matching, the first-order derivative is equal to zero and the main contribution is made by the second-order derivative. In biaxial crystals, in the general case, the second-order and higher derivatives can made comparable contributions.

In particular, the phase matching along the main axes of the crystal (for example, along the z axis, Fig. 7) is an angle-noncritical process of the second order in the main crystal planes and of the fourth order along the phase-matching curves. For the example given in Fig. 7, the directions of phase-matching distributions make angles of ±60° with the xz plane.

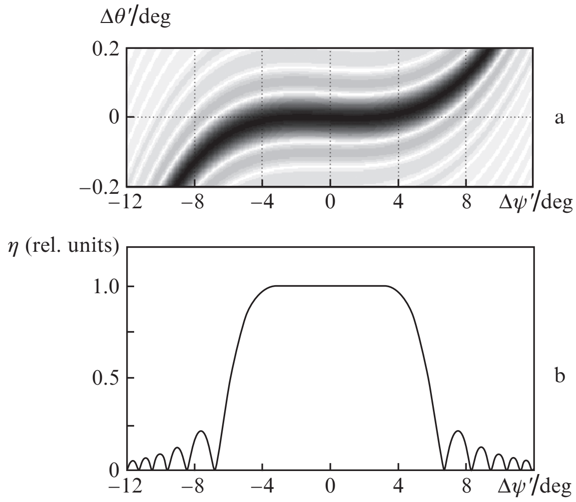

Figure 11. Projection of the three-dimensional distribution of relative conversion efficiency η for the ssf interaction (a) and cross section of this distribution at $\Delta\theta' = 0$ (b).

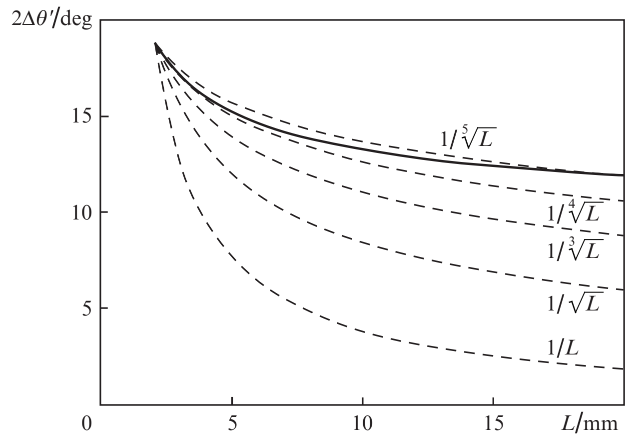

Figure 12. Dependences of the angular phase-matching bandwidth $2\Delta\theta'$ on the crystal length.

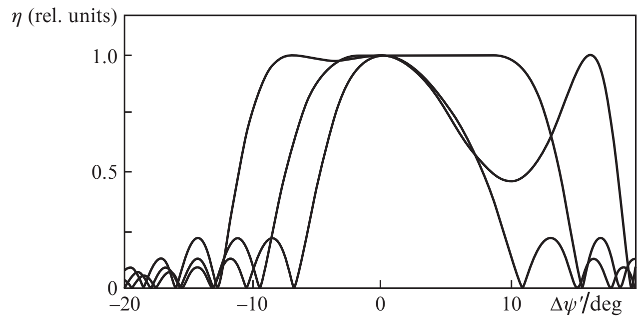

Figure 13. Distributions of the relative conversion efficiency η at different wave detunings.

Along these directions, the angular phase-matching bandwidths are $2\Delta\psi' = 25°\,18'26''$.

Angle-noncritical fourth-order phase matching also occurs for most general crystal cuts. For example, at $\varphi_0 = 40.1°$, $\theta_0 = 71.48°$, and $\gamma = 76.7°$ for THG of radiation with $\lambda = 1.0642$ μm (point E in Fig. 4), the relative conversion efficiency distribution can be seen in Fig. 11. The solid curve in Fig. 12 shows the dependence of the angular phase-matching bandwidth on the crystal length for this case. For comparison, the dashed curves in Fig. 12 show the dependences $2\Delta\theta' \propto 1/\sqrt[n]{L}$ ($n = 1-5$) corresponding to critical and noncritical phase matchings of different orders. This behaviour of the angular width corresponds to comparable contributions of all derivatives with the dominant contribution from the fourth-order derivative. Figure 13 shows the relative conversion efficiency distributions at different initial wave detunings, which demonstrate variations in contributions from derivatives of different orders.

6. Conclusions

Thus, the angular phase-matching bandwidths for biaxial crystals must be calculated in a coordinate system that determines the laser beam divergence and the angular deviations of the crystal. The angular phase-matching bandwidths determined in the traditionally used crystal coordinate system, in the general case, are overestimated for angle-noncritical phase matching and do not allow one to find the angular phase-matching bandwidths along the z axis of the crystal at small phase-matching angles θ_0. In biaxial crystals, phase matching is always noncritical along one angular coordinate and critical along the other. In contrast to uniaxial crystals, in biaxial crystals it is possible to obtain angle-noncritical phase matching of a higher order (up to the fourth order), whose angular width (for example, for THG of radiation with $\lambda = 1.0642$ μm in an LBO crystal) exceeds 25° inside a 10-mm-long crystal.

All the above said refers not only to the determination of angular phase-matching bandwidths. The problem of frequency conversion of laser radiation in nonlinear crystals should be solved in the coordinate system of intrinsic polarisations of radiation in order to minimise the number of equations and to ensure the solution stability. In the general case of phase-matching directions, the conventional method of determination of phase-matching bandwidths in the crystal and laser beam coordinate systems will yield phase matching critical in both angles. Taking into account only the linear angular dependence of refractive indices, one will lose data on the finite phase-matching bandwidth in the directions lying out of the main planes. This is especially important for the problems of frequency conversion when the beam is focused into a crystals by a cylindrical lens. It is necessary to determine the angular phase-matching bandwidth and the optimal orientation of the focusing plane, which does not coincide with the main planes of the crystal. All this is also important for the problems of formation of femtosecond laser beams (whose high intensity frequently allows one to dismiss the question of the finite angular phase-matching bandwidth) and for problems of parametric generation (from the viewpoint of the spectral width of the formed radiation).

All the calculations were performed using the LID-FC program set (Laser Investigator & Designer – Frequency conversion) [6].

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