

# Algorithm for estimation of permissible mixing ratio variations of solid solutions of nonlinear crystals

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Algorithm for estimation of acceptable mixing ratio variations of solid solutions is represented. Numerical estimates have been obtained for the cases of using LiGaSe<sub>1-x</sub>S<sub>x</sub> solid solutions in the second harmonic generators and optical parametric oscillators, and for the general case of three-frequency interactions in the same crystal. The estimates obtained are presented in a graphical form.

## Introduction

Outlook for applications of mixed nonlinear crystals in applied optical systems, including mobile systems for remote sensing of composition of the atmosphere<sup>1</sup> is quite clear now. On the other hand, all grown mixed crystals has variations of the mixing ratio  $x$  of the two initial crystals along and across the growth direction, which can essentially decrease the efficiency of the frequency conversion. Variations of  $x$  along the cross direction are less essential, especially if taking into account that working elements are usually cut of central significantly homogeneous pieces. Minimum variation of the mixing ratio of 1.5% along the growth direction was reached at the length of 40.7 mm in the central part of an 80-mm ingot of a AgGa<sub>1-x</sub>In<sub>x</sub>Se<sub>2</sub> crystal.<sup>2</sup> At the same time, variations of  $x$  up to 15% were observed along the entire length of an ingot of a Hg<sub>1-x</sub>Cd<sub>x</sub>Ga<sub>2</sub>S<sub>4</sub> crystal grown by the Bridgeman–Stockbarger method with the deviation of  $x$  of the obtained composition from  $x$  in compositions of the initial substances reaching 25%.<sup>3</sup> However, to date no algorithm has been developed for calculation of the permissible variations of  $x$ , at which the efficiency of any frequency conversion would be kept at a certain prescribed level. In this paper we describe the technique for such estimates, algorithm and results obtained using it.

## Permissible variations of the mixing ratio for generators of second harmonics and optical parametric oscillators

It is proposed to obtain permissible variations of the mixing ratio for the second harmonic generators (SHG) and optical parametric oscillators from the conditions of phase matching. In the general case, these conditions for three-frequency interactions are written in the following form<sup>4,5</sup>:

$$\Delta k = 2\pi n_1/\lambda_1 + 2\pi n_2/\lambda_2 - 2\pi n_3/\lambda_3 = 0, \quad (1)$$

$$\lambda_1 \geq \lambda_2 > \lambda_3.$$

For mixed crystals one should take into account the validity of the following formula<sup>6</sup>:

$$n_i(x) = [(n_i^A)^2 x + (1-x)n_i^B]^2, \quad i = 1, 2, 3,$$

where the superscripts A and B mean two initial crystals. At the presence of small variations  $\Delta x$  in the mixing ratio from the value  $x_0$  providing for meeting the phase-matching conditions, the phase mismatch appears  $\Delta k \neq 0$ . Let us determine it by differentiating Eq. (1):

$$\Delta k = 2\pi \sum \Delta x \Delta_i / \lambda_i, \quad (2)$$

then

$$\Delta x = \Delta k / (2\pi) \left( \sum \Delta_i / \lambda_i \right)^{-1}.$$

Here  $\Delta_i = \pm \partial n_i(x) / \partial x$ , the sign “+” is applied for  $i = 1, 2$ , and “-” is for  $i = 3$ . Taking into account that  $\Delta x \ll x$  and assuming that accompanying variations of  $n_i$  are small, let us determine the permissible limits of variations of  $x$ . Let us consider linear approximation of the terms  $\sum_{i=1}^3 \Delta_i / \lambda_i$  and  $\left( \sum_{i=1}^3 \Delta_i / \lambda_i \right)^{-1}$  as functions of  $x$ , then expanding them into a series we obtain

$$\begin{aligned} & \left( \sum_{i=1}^3 \frac{\Delta_i}{\lambda_i} \right)^{-1} = \\ & = \frac{1}{\sum_{i=1}^3 \Delta_i / \lambda_i \Big|_{x=x_0}} \left( 1 - \left( \sum_{i=1}^3 \frac{\Delta'_i}{\lambda_i} / \sum_{i=1}^3 \frac{\Delta_i}{\lambda_i} \right) \Big|_{x=x_0} (x-x_0) \right) + \dots \approx \\ & \approx \alpha [1 + \varepsilon(x-x_0)], \\ & \Delta x = \Delta k \alpha / (2\pi) [1 + \varepsilon(x-x_0)]. \quad (3) \end{aligned}$$

If one accepts  $\Delta k = \pi/l$  ( $l$  is the crystal length), then the 59% decrease of the efficiency of conversion takes place due to the phase mismatch  $\text{sinc}^2(\Delta kl/2) = 4/\pi^2 \approx 41\%$ .<sup>3,4</sup> Then the simple algorithm is obtained from Eq. (3) for determining the permissible  $x$  variations in the considered case:

$$\Delta x = \alpha[1 + \varepsilon(x - x_0)]/2l.$$

It is seen that they are proportional to  $\alpha$  (through  $\alpha$ ) and inversely proportional to the crystal length. If, at different  $x$ , the phase-matching curves have the cross point at some wavelength, the permissible range of variations can reach 1.0. Let us note that it is not an interesting case for practical applications, because the mixed crystal takes the properties identical to the properties of the initial crystals at any mixing ratio. For determining the positive  $\Delta x^+$  and negative  $\Delta x^-$  variations of  $x$  one can use the following equations

$$\begin{aligned} \Delta k(x + \Delta x^+, \lambda_1, \lambda_2, \lambda_3) &= \pi/l \\ \Delta k(x + \Delta x^-, \lambda_1, \lambda_2, \lambda_3) &= -\pi/l. \end{aligned} \quad (4)$$

To determine the optimal (that, which corresponds to the position of the first local maximum in the efficiency of the frequency conversion) and permissible crystal lengths, enabling one to keep efficiency of the frequency conversion within the aforementioned limit in the presence of variations in the mixing ratio, let us consider two typical cases. The first one assumes the presence of smooth deviation of  $x$  from  $x_0$ , and the second one presents the situation with statistically distributed local variations of the mixing ratio. To analyze the first case, let us introduce the gradient  $\beta$  of  $x$  along the direction of radiation propagation, then

$$x = x_0 + \beta(z - l/2) \text{ and } \Delta x = x - x_0 = \beta(z - l/2),$$

where  $z$  is the longitudinal coordinate inside the crystal with  $z = 0$  at the input surface and  $z = l$  at the output one. Then in approximation of a preset field we have for the SH amplitude<sup>6</sup>:

$$A_{\text{sh}} = [i\omega_p/(n_{\text{sh}}c)]d_{\text{eff}}A_p^2 \int_0^l \exp(i\Delta kz)dz \propto \int_0^l \exp(i\Delta kz)dz, \quad (5)$$

where  $A_p$  and  $A_{\text{sh}}$  are the amplitudes of the fundamental wave and of the SH wave,  $d_{\text{eff}}$  is the nonlinear susceptibility coefficient of the second order,  $n_{\text{sh}}$  is the refractive index for the fundamental (at the wavelength  $\lambda_p$ ) and the second harmonics ( $\lambda_{\text{sh}}$ ) waves in the direction of phase matching,  $\omega_p$  is the fundamental frequency, and  $c$  is the speed of light in vacuum. Both  $A_p$  and  $A_{\text{sh}}$  are related with the complex amplitudes of the field and the intensities:

$$E_p(r, t) = (1/2)A_p \exp[i(\mathbf{k}_p \cdot \mathbf{r} - \omega t)] + \text{c. c.};$$

$$E_{\text{sh}}(r, t) = (1/2)A_{\text{sh}} \exp[i(\mathbf{k}_{\text{sh}} \cdot \mathbf{r} - 2\omega t)] + \text{c. c.};$$

$$I_p(r, t) = (1/2)n_p c \varepsilon_0 |A_p|^2, \quad I_{\text{sh}}(r, t) = (1/2)n_{\text{sh}} c \varepsilon_0 |A_{\text{sh}}|^2, \quad (6)$$

where  $\varepsilon_0$  is the dielectric constant of vacuum. Restricting oneself to the first-order approximation of the expansion of the general formula for  $\Delta x$  and solving it relative to  $\Delta k$ , we obtain

$$\Delta k \approx 2\pi\Delta x/\alpha = 4\pi K\Delta x = 4\pi K\beta(z - l/2), \quad (7)$$

where  $K = 1/(2\alpha)$ .

As the refractive indices change due to the variations of  $x$ , the process of frequency conversion differs from the conversion process in usual nonlinear crystals. In this case one can obtain the correct result from new connected equations, which take into account the presence of weak gradients in the refractive indices, namely: it is necessary to replace the formula (7) for the phase mismatch  $\Delta k$  with  $\Delta k^* = 2\pi K\beta(z - l)$ . Then Eq. (5) is transformed to the form:

$$\begin{aligned} \int_0^l \exp(i\Delta k^* z) dz &= \int_0^l \exp[i2\pi K\beta(z - l)z] dz = \\ &= \int_0^l \exp\{i2\pi K\beta[(z - l/2)^2 - l^2/4]\} dz = \\ &= e^{-i\pi K l^2 \beta/2} \int_0^l e^{i2\pi K\beta(z - l/2)^2} dz = e^{-i\pi K l^2 \beta/2} \times \\ &\times \int_0^l \{\cos[2\pi K\beta(z - l/2)^2] + i \sin[2\pi K\beta(z - l/2)^2]\} dz = \\ &= e^{-i\pi K l^2 \beta/2} \sqrt{1/|K\beta|} \left[ C(l\sqrt{|K\beta|}) \pm i S(l\sqrt{|K\beta|}) \right], \end{aligned} \quad (8)$$

where

$$S(\tau) = \int_0^\tau \sin(\pi t^2/2) dt, \quad C(\tau) = \int_0^\tau \cos(\pi t^2/2) dt$$

are the integral sine and cosine,  $\tau = L|K\beta|^{1/2}$ . The sign “+” in Eq. (8) is used when  $K\beta$  is positive, and “-“ if it is negative.

Taking into account Eq. (8), let us find the intensity of SH wave from Eq. (6):

$$\begin{aligned} I_{\text{sh}} &= I_p \left[ C^2(l\sqrt{|K\beta|}) + S^2(l\sqrt{|K\beta|}) \right] / [K\beta L_{\text{nl}}^2] = \eta I_p; \\ L_{\text{nl}} &= \left( 8\pi^2 d_{\text{eff}}^2 I_p / [n_p^2 n_{\text{sh}} \varepsilon_0 c \lambda_p^2] \right)^{-1/2}, \end{aligned} \quad (9)$$

where  $\eta$  is the efficiency of conversion,  $L_{\text{nl}}$  is the nonlinear crystal length. Let us determine the optimal length  $l_m$  by differentiating Eq. (9) and reducing it to equation of the form:

$$C(\tau) \cos(\tau^2 \pi/2) + S(\tau) \sin(\tau^2 \pi/2) = 0.$$

The value determined by numerical method is  $\tau \approx 1.21$ . Then the optimal crystal length is determined as  $l_m = \tau\sqrt{1/|K\beta|}$ , and the absolute value of the mean variation of  $x$   $|\Delta x_d|$  is determined from the formula

$$\Delta x = x - x_0 = \beta(z - l/2)$$

as  $|\Delta x_d| = l|\beta|/4$ , then  $|\beta| = 4|\Delta x_d|/l$ . Substituting  $|\beta|$  and the second equality of Eq. (7) into the formula for  $l_m$  and equalizing  $l$  to  $l_m$ , we obtain the formula for the optimal length  $l_m = \tau^2/2|\alpha/\Delta x_d|$ . This relation represents the balance between the crystal length and the variation of  $x$ , which can be used for constructing the prototypes of the crystals.

The optimal crystal length decreases at significant variations of  $x$ , then the walk-off effect, absorption, and the efficiency of conversion also decrease, so the final formula for the optimal crystal length provides for quite a good accuracy of estimates.

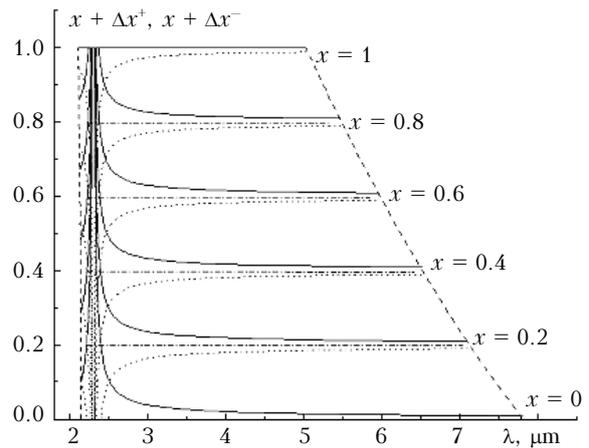
Only mean value of the phase-matching conditions or the decrease of efficiency of conversion can be estimated in the presence of local variations of  $x$  inside the crystal. If one introduces the mean absolute value of the deviations of  $x$   $|\Delta x_d|$ , one can obtain the optimal crystal length  $l_a$  providing for the accepted decrease of the efficiency of conversion from Eq. (3) ignoring  $\varepsilon$ , in the form  $l_a = 0.5|\alpha/\Delta x_d|$ . This formula provides for the qualitative estimate, because it does not reflect the true pattern of  $x$  variations.

Let us note that the formulas for the optimal and permissible crystal lengths differ only by the value  $\tau^2$ , in the second case it is approximately equal to 1.46. Let us also note that the accepted assumption about low efficiency of the frequency conversion we used excludes application of the proposed algorithm to analysis of the frequency conversion in crystals with large optimal or permissible lengths. However, in this case the characteristic crystal length  $L_{nl}$ , at which the efficiency of frequency doubling can reach 58% can be taken as a criterion.<sup>7</sup> Besides, applying the proposed method to estimation requires that the following conditions hold

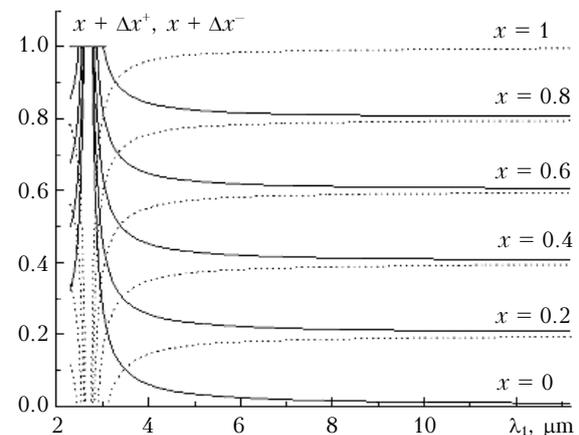
$$l_m < L_{nl}, \quad l_a < L_{nl},$$

while these inequalities assume the existence of the minimum variation of  $x$ , i.e.,  $|\Delta x_d| > 2\tau^2|\alpha|/L_{nl}$  in determining the optimal crystal length and  $|\Delta x_d| > 2|\alpha|/L_{nl}$  in determining the permissible length, for which the estimates are only valid.

One can perform analogous procedure for estimation of the efficiency of the optical parametric oscillation (OPO). The results of numerical estimation are shown in Figs. 1 and 2 for the second harmonics and the optical parametric oscillator assuming the relationship between the optical and crystallographic axes to be as follows  $XYZ \rightarrow bac$ .



**Fig. 1.** Permissible variations of the mixing ratio for solid solutions  $\text{LiGaSe}_{1-x}\text{S}_x$  (length = 1 cm) if using them as second harmonic generators under the phase matching conditions of type II in the main plane “ba” (XY):  $x + \Delta x^+$  (solid curves);  $x + \Delta x^-$  (dotted curves); border (dashed curves).



**Fig. 2.** Permissible variations of the mixing ratio for solid solutions  $\text{LiGaSe}_{1-x}\text{S}_x$  (length = 1 cm) if using them as optical parametric oscillators (Nd:YAG-laser) under the phase matching conditions of  $sf$ -type in the main plane “bc” (XZ):  $x + \Delta x^+$  (solid curves);  $x + \Delta x^-$  (dotted curves).

## Permissible variations of the mixing ratio for three-frequency interactions

In contrast to the particular cases of SHG (two interacting frequencies are degenerate) and OPO (splitting of the pump photon into a pair of photons determined by the energy conservation law) for the general case of three-frequency interactions, the number of variants of interactions is infinite (any two frequencies can interact yielding generation of a third frequency determined by the energy conservation law). Sum frequency generation (SFG) and difference frequency generation (DFG) are among such interactions. To determine the permissible variations of  $x$  in this case, let us use the initial general formulas for determining the values of the refractive indices from Eqs. (7) and (8). In an anisotropic crystal they are set along the direction of propagation

$\mathbf{u}(\theta, \phi)$  by the Fresnel equation, which has two solutions:

$$n^+(\omega_i, \theta, \phi) = \left(2/\left[-B_i - (B_i^2 - 4C_i)^{1/2}\right]\right)^{1/2},$$

$$n^-(\omega_i, \theta, \phi) = \left(2/\left[-B_i + (B_i^2 - 4C_i)^{1/2}\right]\right)^{1/2}, \quad (10)$$

where  $n^+(\omega_i) > n^-(\omega_i)$ , and

$$B_i = -u_x^2(b_i + c_i) - u_y^2(a_i + c_i) - u_z^2(a_i + b_i),$$

$$C_i = u_x^2 b_i c_i + u_y^2 a_i c_i + u_z^2 a_i b_i. \quad (11)$$

Here  $a_i = n_x^{-2}$ ;  $b_i = n_y^{-2}$ ;  $c_i = n_z^{-2}$ ;  $u_x = \sin\theta \cos\phi$ ,  $u_y = \sin\theta \sin\phi$ , and  $u_z = \cos\theta$  are the Cartesian coordinates of a unit vector  $\mathbf{u}(\theta, \phi)$ ;  $n_x(\omega_i)$ ,  $n_y(\omega_i)$ ,  $n_z(\omega_i)$  are the main refractive indices of the refractive index ellipsoid. Energy conversion in SFG and DFG processes also means fulfillment of the energy conservation law:

$$\omega_1 + \omega_2 = \omega_3, \quad \omega_1 \leq \omega_2 < \omega_3,$$

and the law of momentum conservation:

$$\mathbf{k}(\omega_1, \theta, \phi) + \mathbf{k}(\omega_2, \theta, \phi) = \mathbf{k}(\omega_3, \theta, \phi),$$

where  $\mathbf{k}$  is the wave vector of the normal unit vector  $\mathbf{u}(\theta, \phi)$ . The phase-matching conditions for I, II, and III types of interaction have the following form:

$$\omega_1 n^+(\omega_1, \theta, \phi) + \omega_2 n^+(\omega_2, \theta, \phi) = \omega_3 n^-(\omega_3, \theta, \phi),$$

$$\omega_1 n^-(\omega_1, \theta, \phi) + \omega_2 n^+(\omega_2, \theta, \phi) = \omega_3 n^-(\omega_3, \theta, \phi),$$

$$\omega_1 n^+(\omega_1, \theta, \phi) + \omega_2 n^-(\omega_2, \theta, \phi) = \omega_3 n^-(\omega_3, \theta, \phi).$$

For mixed crystals:

$$\omega_1 n^+(\omega_1, \theta, \phi, x) + \omega_2 n^+(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^-(\omega_1, \theta, \phi, x) + \omega_2 n^+(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x) + \omega_2 n^-(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x).$$

To determine the permissible variations of composition  $\Delta x^+$  and  $\Delta x^-$  for I, II, and III types of interaction, one can use the following formulas:

$$\omega_1 n^+(\omega_1, \theta, \phi, x) + \omega_2 n^+(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x + \Delta x^+) + \omega_2 n^+(\omega_2, \theta, \phi, x + \Delta x^+) -$$

$$-\omega_3 n^-(\omega_3, \theta, \phi, x + \Delta x^+) = \pi c/l;$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x) + \omega_2 n^+(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x + \Delta x^-) + \omega_2 n^+(\omega_2, \theta, \phi, x + \Delta x^-) -$$

$$-\omega_3 n^-(\omega_3, \theta, \phi, x + \Delta x^-) = -\pi c/l;$$

$$\omega_1 n^-(\omega_1, \theta, \phi, x) + \omega_2 n^+(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^-(\omega_1, \theta, \phi, x + \Delta x^+) + \omega_2 n^+(\omega_2, \theta, \phi, x + \Delta x^+) -$$

$$-\omega_3 n^-(\omega_3, \theta, \phi, x + \Delta x^+) = \pi c/l;$$

$$\omega_1 n^-(\omega_1, \theta, \phi, x) + \omega_2 n^+(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^-(\omega_1, \theta, \phi, x + \Delta x^-) + \omega_2 n^+(\omega_2, \theta, \phi, x + \Delta x^-) -$$

$$-\omega_3 n^-(\omega_3, \theta, \phi, x + \Delta x^-) = -\pi c/l;$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x) + \omega_2 n^-(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x + \Delta x^+) + \omega_2 n^-(\omega_2, \theta, \phi, x + \Delta x^+) -$$

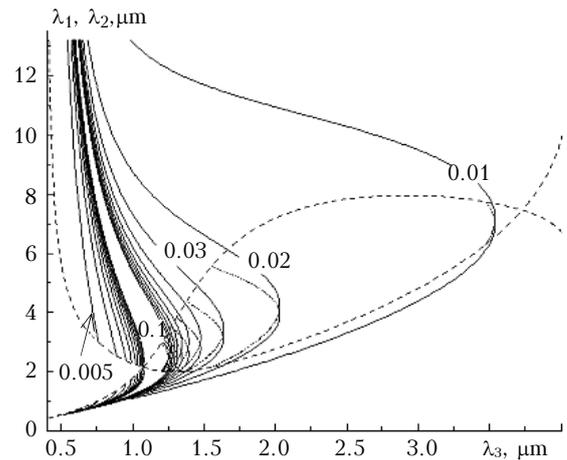
$$-\omega_3 n^-(\omega_3, \theta, \phi, x + \Delta x^+) = \pi c/l;$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x) + \omega_2 n^-(\omega_2, \theta, \phi, x) = \omega_3 n^-(\omega_3, \theta, \phi, x),$$

$$\omega_1 n^+(\omega_1, \theta, \phi, x + \Delta x^-) + \omega_2 n^-(\omega_2, \theta, \phi, x + \Delta x^-) -$$

$$-\omega_3 n^-(\omega_3, \theta, \phi, x + \Delta x^-) = -\pi c/l.$$

At the fixed values of the parameters  $\omega_1(\lambda_1)$ ,  $\omega_2(\lambda_2)$ ,  $\omega_3(\lambda_3)$ ,  $l$ , and  $x$ , there are three variables  $\theta$ ,  $\phi$ , and  $\Delta x^\pm$ , which one needs to determine, but two equations (in the case of uniaxial crystal the solution does not depend on the angle  $\phi$  (or  $\theta$ ) and can be obtained numerically). So, in practice it is necessary to determine the azimuth  $\phi$  or polar angles  $\theta$ , and the complicated dependence between them  $\theta = \theta(\phi)$  in case of an arbitrary direction of the trajectory. Their choice is necessary not only for producing the prototypes of the crystals, but also for mutual reference of crystallographic and optical coordinate systems. The results of estimation of the permissible values of variations of the mixing ratio for three-frequency interactions (SFG/DFG) are shown in Fig. 3.



**Fig. 3.** Permissible variations of the mixing ratio for solid solutions  $\text{LiGaSe}_{1-x}\text{S}_x$  ( $x = 0,1$ ; length = 1 cm) if using them as sum/difference frequency generators under the phase matching conditions in the main plane “ba” (XY): *fsf*-type (solid curves); *sff*-type (dotted curves); border (dashed curves).

It follows from Figs. 1 to 3 that the requirements to the permissible variations of the mixing ratio for

solid solutions  $\text{LiGaSe}_{1-x}\text{S}_x$  are acceptable in a wide wavelength range.

## Conclusions

The algorithm developed has made it possible, for the first time, to estimate the permissible variations of the mixing ratio of solid solutions of nonlinear crystals. Its application to different types of conversion in the known solid solutions has shown that even with the centimeter size the permissible variations of the mixing ratio for wide wavelength ranges lie within reasonable limits from 1 to 10%. The results of estimation can be used for determination of technological tasks and ordering the working elements of solid solutions of nonlinear crystals.

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