

# Stochastic quasi-phase matching in nonlinear-optical crystals with an irregular domain structure

E.Yu. Morozov, A.S. Chirkin

**Abstract.** A theory of the interaction between light waves in polydomain crystals with a random variation in the domain thickness described by a random telegraph process is developed. The second harmonic generation and parametric amplification upon high-frequency pumping are considered. It is found that the maximum efficiency of nonlinear conversion is achieved when the phase mismatch between the interacting waves is equal to the doubled spatial frequency at which the nonlinearity sign is changed (the condition of stochastic quasi-phase matching).

**Keywords:** quasi-phase-matched interactions, second harmonic generation, parametric amplification, domain structure.

## 1. Introduction

In polydomain optical crystals with the so-called 180° domains and in periodically poled nonlinear-optical crystals, i.e., in crystals with inverted optical axes, the coefficients of the quadratic nonlinear susceptibility can change their sign from layer to layer (see reviews [1, 2]). This results in the appearance of a nonlinear-susceptibility grating. In this case, the reciprocal vector of the grating can compensate for the phase mismatch  $\Delta k$  between the interacting waves:

$$\Delta k = \frac{2\pi}{\Lambda_0} m. \quad (1)$$

This relation is called the quasi-phase matching condition. Here,  $\Lambda_0$  is the spatial period of the nonlinear-susceptibility modulation and  $m$  is the quasi-phase matching order. Quasi-phase-matched interactions allow the use of types of interactions or nonlinear crystals for which the conventional phase matching condition cannot be fulfilled. From the practical point of view, the most important is the fact that in the case of quasi-phase-matched interactions, the maximum nonlinearity of a crystal can be achieved by choosing appropriately the polarisation of the interacting

waves. For example, in a polydomain (or periodically poled) LiNbO<sub>3</sub> crystal, the ee–e interaction is used, which is related to the nonlinear coefficient  $d_{33}$  exceeding the other coefficients of this nonlinear crystal almost by an order of magnitude.

Quasi-phase-matched interactions are used at present in nonlinear optics to obtain both intense coherent radiation in the spectral range from IR to UV [1, 3] and nonclassical light [4, 5]. In addition, quasi-phase-matched interactions make it possible to realise nonlinear-optical interactions of a new type: consecutive interactions, when several coupled three-frequency processes with common waves proceed simultaneously in a nonlinear medium. A brief review of experimental and theoretical studies in this field is presented in Ref. [2].

The extension of the field of applications of quasi-phase-matched interactions has posed new problems related not only to the improvement of the technology of synthesis of periodically poled crystals but also to the development of the theory of nonlinear-optical interactions to describe adequately a new experimental situation. The latter problem arises not because the nonlinear coupling coefficient of the waves is a spatially periodic function but because this periodicity is violated.

The matter is that the modulation of the nonlinear coupling coefficient of the waves in polydomain crystals used in experiments differs from a strictly periodic modulation due to different technological reasons. These can be variations in the refractive index in a crystal volume caused by the inhomogeneous distribution of impurities and internal mechanical stresses produced during the crystal growth and its subsequent treatment. The period of a nonlinear structure also can differ from a perfect one due to an unstable operation of various units of a crystal growth apparatus, etc.

In this paper, we develop the theory of three-frequency nonlinear-optical interactions when the spatial modulation of the nonlinear coupling coefficient of the waves is simulated by a random telegraph process. Such a process describes a random variation in the domain thickness from one domain to another along with the simultaneous inversion of the optical axis. We have already used this model of the randomly modulated coupling coefficient of the waves to analyse frequency doubling in a disordered polydomain crystal [6], in which both the domain thickness and phase mismatch caused by the inhomogeneities of the refractive index and fluctuations in the optical-axis direction are random.

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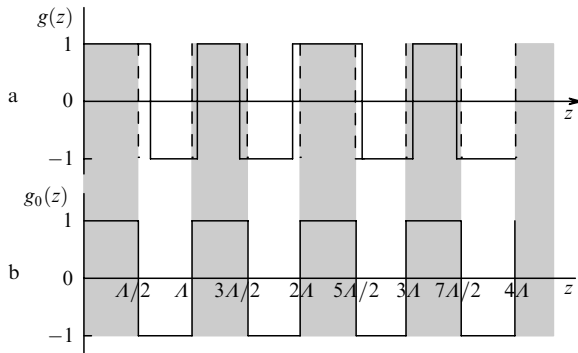
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## 2. Spatially modulated nonlinear coefficient as a random telegraph process

To analyse the statistic properties of the nonlinear coupling coefficient of waves, we will write the equations for the second harmonic generation in the plane wave approximation:

$$\begin{aligned} \frac{dA_1}{dz} &= -ig(z)\beta A_2 A_1^* \exp(-i\Delta kz), \\ \frac{dA_2}{dz} &= -ig(z)\beta A_1^2 \exp(i\Delta kz), \end{aligned} \quad (2)$$

where  $A_1 = A_1(z)$ ,  $A_2 = A_2(z)$  are the slowly varying complex amplitudes of the fundamental radiation at frequency  $\omega$  and the second harmonic, respectively;  $\Delta k$  is the mismatch of the wave vectors of the interacting waves;  $\beta = d_{\text{eff}} 2\pi\omega / [cn(\omega)]$  is the modulus of the nonlinear coupling coefficient of waves; the coefficient  $d_{\text{eff}}$  takes into account the interaction geometry and polarisation of the waves. The function  $g(z)$  describes the spatial modulation of the nonlinear coefficient caused by the inversion of the optical axis in passing from one domain to another (Fig. 1).



**Figure 1.** Random modulation of the nonlinear coupling coefficient of waves  $g(z)$  (random telegraph signal) (a) and a regular modulation of the nonlinear coefficient  $g_0(z)$  (b).

We assume that the function  $g(z)$  describes a random telegraph process, which takes the values  $+1$  and  $-1$  with equal probabilities  $+1$  and  $-1$  [ $g^2(z) \equiv 1$ ] [7, 8], i.e.,

$$g(z) = (-1)^{n(0,z)},$$

where  $n(z_1, z_2)$  is a random sequence of integers describing the number of the sign changes in the nonlinear coefficient over the length  $(z_1, z_2)$ . The number  $n(0, z)$  of changes of the sign at the interval  $(0, z)$  obeys the Poisson statistics

$$P(n) = \frac{(vz)^n}{n!} \exp(-vz), \quad (3)$$

where  $v$  is the average number of sign changes per unit length (the average spatial frequency). Therefore, the average number  $\bar{n}$  of sign changes over the length  $z$  and its dispersion  $\sigma_n^2$  are

$$\bar{n} = vz, \quad \sigma_n^2 = \overline{n^2} - \bar{n}^2 = \bar{n}, \quad (4)$$

respectively. The function  $g(z)$  has the following statistic properties [7, 8] (we will denote the statistic average both by angle brackets and the bar over the averaged quantity):

$$\langle g(z) \rangle = \exp(-2vz), \quad \langle g(z')g(z'') \rangle = \exp(-2v|z' - z''|). \quad (5)$$

If  $\mathcal{F}(z, g(z))$  is a functional, then the expression [8]

$$\left( \frac{d}{dz} + 2v \right) \langle g(z) \mathcal{F}[z, g(z)] \rangle = \left\langle g(z) \frac{d\mathcal{F}[z, g(z)]}{dz} \right\rangle \quad (6)$$

for differentiation is valid, which we will use in our calculations.

## 3. Second harmonic generation

Let us analyse frequency doubling in nonlinear crystals with a random aperiodic structure. We rewrite system (2) for the intensity  $I_j(z) = |A_j(z)|^2$  ( $j = 1, 2$ ). By introducing the notation  $U_i(z) = \text{Im}[A_2(z)A_1^{*2} \exp(-i\Delta kz)]$  and  $U_r(z) = \text{Re}[A_2(z)A_1^{*2} \exp(-i\Delta kz)]$ , we obtain

$$\frac{dI_1}{dz} = 2g(z)\beta U_i,$$

$$\frac{dI_2}{dz} = -2g(z)\beta U_i,$$

$$\frac{dU_i}{dz} = -g(z)\beta I_1^2 + 2g(z)\beta I_1 I_2 - \Delta k U_r, \quad (7)$$

$$\frac{dU_r}{dz} = \Delta k U_i.$$

Let us average system (7) over an ensemble of random functions  $g(z)$ . By multiplying the two last equations of system (7) by  $g(z)$  and using (6), we obtain

$$\frac{d\bar{I}_1}{dz} = 2\beta\psi_1,$$

$$\frac{d\bar{I}_2}{dz} = -2\beta\psi_1,$$

$$\frac{d\psi_1}{dz} = -2v\psi_1 - \Delta k\psi_2 - \beta\langle I_1^2 \rangle + 2\beta\langle I_1 I_2 \rangle, \quad (8)$$

$$\frac{d\psi_2}{dz} = \Delta k\psi_1 - 2v\psi_2,$$

where  $\psi_1 = \langle g(z)U_i \rangle$ ;  $\psi_2 = \langle g(z)U_r \rangle$ .

System of equations (8) is unclosed because it contains, except the average intensities  $\langle I_j \rangle$  ( $j = 1, 2$ ), the second-order moments  $\langle I_j I_q \rangle$  ( $j, q = 1, 2$ ). Generally speaking, we can write equations for these moments as well; however, they will contain higher-order intensity moments. This situation is typical for nonlinear equations.

At the same time, it is reasonable to assume that the statistics of the second-harmonic field in the ‘developed’ regime of its excitation is Gaussian. We can easily obtain

from (7) the relation  $I_1(z) + I_2(z) = I_{10}$ , where  $I_{10}$  is the initial intensity of the fundamental radiation. Then, the second order intensity moments can be expressed in terms of the average intensity  $\bar{I}_2(z)$  of the second harmonic [9]

$$\langle I_1 I_2 \rangle = I_{10} \bar{I}_2 - 2\bar{I}_2^2, \quad (9)$$

$$\langle I_1^2 \rangle = I_{10}^2 - 2I_{10} \bar{I}_2 + 2\bar{I}_2^2.$$

Let us substitute (9) into (8) and pass to the dimensionless quantities

$$x = \frac{\bar{I}_2}{I_{10}}, \quad y_i = \frac{\psi_i}{I_{10}^{3/2}}, \quad \zeta = \frac{z}{L_{\text{nl}}}, \quad \alpha = 2vL_{\text{nl}}, \quad \Delta = \Delta k L_{\text{nl}}, \quad (10)$$

where  $L_{\text{nl}} = 1/(\beta I_{10}^{1/2})$  is the characteristic nonlinear length and  $\alpha$  and  $\Delta$  are the reduced spatial frequency and phase mismatch, respectively. As a result, the system of equations (8) will take the form

$$\begin{aligned} \frac{dx}{d\zeta} &= -2y_1, \\ \frac{dy_1}{d\zeta} &= -\alpha y_1 - \Delta y_2 - (1 - 4x + 6x^2), \\ \frac{dy_2}{d\zeta} &= \Delta y_1 - \alpha y_2, \end{aligned} \quad (11)$$

which is convenient for a further analysis. Note that the parameters  $\alpha$  and  $\Delta$  characterise the disorder of the crystal structure and the phase mismatch over the nonlinear length  $L_{\text{nl}}$ .

The system of equations (11) can be solved only numerically. However, some analytic results, which are important for understanding the features of second harmonic generation in crystals with a random domain structure, can be obtained in the fixed field approximation. In this approximation, the terms containing  $x$  and  $x^2$  in the right-hand side of the second equation in (11) can be neglected compared to unity. By solving the system of equations obtained this way with the initial condition  $[x(0) = 0]$ , we obtain the normalised intensity of the second harmonic

$$\begin{aligned} x(\zeta) &= \frac{2[-\alpha^2 + \Delta^2 + \alpha(\alpha^2 + \Delta^2)\zeta]}{(\alpha^2 + \Delta^2)^2} \\ &+ \frac{2e^{-\alpha\zeta}[(\alpha^2 - \Delta^2)\cos\Delta\zeta - 4\alpha\Delta\sin\Delta\zeta]}{(\alpha^2 + \Delta^2)^2}. \end{aligned} \quad (12)$$

Dependence (12) can be also obtained differently. In the fixed field approximation, we obtain directly from the system of equations (2) the solution for  $A_2(z)$ :

$$A_2(z) = -i\beta A_{10}^2 \int_0^z g(z) \exp(i\Delta kz) dz, \quad (13)$$

where  $A_{10} = A_1(z=0)$ . From (13), taking into account expression (5) for the correlation function of a random telegraph process, we obtain the average intensity of the second harmonic

$$\begin{aligned} \langle I_2(z) \rangle &= \beta^2 I_{10}^2 \int_0^z \int_0^z \exp(-2v|z' - z''|) \\ &\times \exp[i\Delta k(z' - z'')] dz' dz''. \end{aligned} \quad (14)$$

Integration in (14) gives (12).

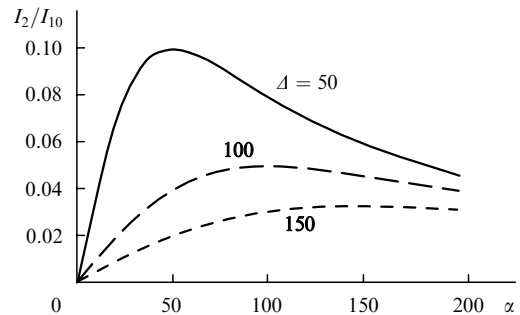
Analysis of expression (12) shows that there exists the optimal relation between the average spatial frequency  $v$  of the random process  $g(z)$  and the phase mismatch  $\Delta k$ . Assuming that  $v \sim \Delta k \gg 1$ , we obtain

$$\Delta k = 2v \left[ \frac{3}{2v\zeta} + \left( 1 - \frac{1}{v\zeta} + \frac{9}{4v^2\zeta^2} \right)^{1/2} \right]^{1/2}. \quad (15a)$$

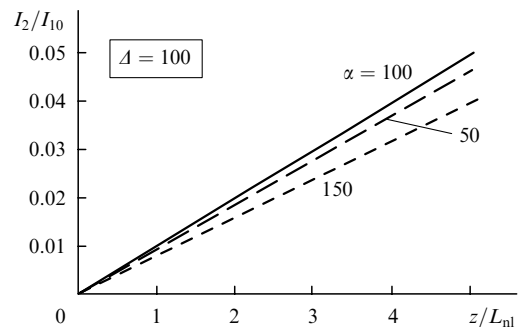
For a large number of domains on a length under study ( $2v\zeta \gg 1$ ), we have

$$\Delta k \approx 2v \left( 1 + \frac{1}{2v\zeta} + \frac{2.25}{(2v\zeta)^2} \right). \quad (15b)$$

Figures 2 and 3 present the dependences of the second-harmonic intensity calculated numerically from system (11). One can clearly see from Fig. 2 that there exists the optimal relation between the values of  $\alpha$  and  $\Delta$  when the second-harmonic intensity is maximum. Figure 3 demonstrates that the second-harmonic intensity increases most rapidly when  $\alpha \approx \Delta$ .



**Figure 2.** Dependences of the normalised intensity  $I_2(z)/I_{10}$  of the second harmonic on the reduced average spatial frequency  $\alpha$  of the change in the sign of the nonlinear coupling coefficient for  $\Delta = 50, 100$ , and  $150$  ( $z/L_{\text{nl}} = 5$ ).



**Figure 3.** Dependences of the normalised intensity  $I_2(z)/I_{10}$  of second harmonic on the reduced interaction length for different reduced average spatial frequencies  $\alpha$  ( $\Delta = 100$ ).

Relations (15) are the conditions for stochastic quasi-phase matching. They can be expressed in terms of the average period  $\bar{A} = 2/\nu$  of a layered structure. Note that the optimal relation in the case of a disordered medium depends on the interaction length  $\zeta$ . By comparing (1) and (15b), we obtain

$$\frac{\bar{A}}{A_0} \approx \frac{2}{\pi}.$$

Therefore, by considering the second harmonic generation, we have found the presence of stochastic quasi-phase matching, which provides the maximum energy exchange between the interacting waves in randomly aperiodic polydomain crystals. It should be expected that a similar situation also would take place in the case of nondegenerate three-frequency interactions.

#### 4. Parametric frequency conversion

Consider now the three-frequency interaction between waves of the type  $\omega_3 = \omega_1 + \omega_2$ , which is described by the system of truncated equations

$$\begin{aligned} \frac{dA_1}{dz} &= -ig(z)\beta_1 A_3(z)A_2^*(z) \exp[i\Phi(z)], \\ \frac{dA_2}{dz} &= -ig(z)\beta_2 A_3(z)A_1^*(z) \exp[i\Phi(z)], \\ \frac{dA_3}{dz} &= -ig(z)\beta_3 A_1(z)A_2(z) \exp[-i\Phi(z)], \end{aligned} \quad (16)$$

where  $A_1(z)$ ,  $A_2(z)$ , and  $A_3(z)$  are the complex amplitudes of the waves with frequencies  $\omega_1$ ,  $\omega_2$ , and  $\omega_3$ , respectively;  $\beta_j$  are the absolute values of the nonlinear coupling coefficients of waves ( $j = 1, 2, 3$ ). The function  $g(z)$ , as above, describes a random modulation of the nonlinear coefficient. The function  $g(z)$  takes into account the phase shift due to the phase mismatch between the interacting waves over the interaction length  $z$ :

$$\Phi(z) = \int_0^z [k_3(z') - k_2(z') - k_1(z')] dz' = \Delta k z, \quad (17)$$

where  $k_j(z) = k_j$  is the wave number at the corresponding frequency and  $\Delta k$  is the regular phase mismatch.

Now we will proceed in the same way as in the previous section and convert the system of equations (17) to the form [cf. (7)]

$$\begin{aligned} \frac{dI_1}{dz} &= 2g(z)\beta_1 U_i, \\ \frac{dI_2}{dz} &= 2g(z)\beta_2 U_i, \\ \frac{dI_3}{dz} &= -2g(z)\beta_3 U_i, \\ \frac{dU_i}{dz} &= -g(z)\beta_3 I_1 I_2 + g(z)\beta_2 I_1 I_3 + g(z)\beta_1 I_2 I_3 - \Delta k U_r, \\ \frac{dU_r}{dz} &= \Delta k U_i, \end{aligned} \quad (18)$$

where  $U_i(z) = \text{Im}\{A_1^* A_2^* A_3 \exp[i\Phi(z)]\}$ ; and  $U_r(z) = \text{Re}\{A_1^* \times A_2^* A_3 \exp[i\Phi(z)]\}$ .

From (18), taking into account (6), we obtain for the averaged system:

$$\begin{aligned} \frac{d\bar{I}_1}{dz} &= 2\beta_1 \psi_1, \quad \frac{d\bar{I}_2}{dz} = 2\beta_2 \psi_1, \quad \frac{d\bar{I}_3}{dz} = -2\beta_3 \psi_1, \\ \frac{d\psi_1}{dz} &= -2\nu\psi_1 - \Delta k \psi_2 - \beta_3 \langle I_1 I_2 \rangle + \beta_2 \langle I_1 I_3 \rangle + \beta_1 \langle I_2 I_3 \rangle, \\ \frac{d\psi_2}{dz} &= \Delta k \psi_1 - 2\nu\psi_2, \end{aligned} \quad (19)$$

where  $\psi_1 = \langle g(z)U_i(z) \rangle$ ;  $\psi_2 = \langle g(z)U_r(z) \rangle$ .

Let us use the Manley–Rowe relations, which follows from the first three equations of system (18). Then, for low initial intensities of the fields being amplified compared to the pump intensity ( $I_{30} \gg I_{10}$ ) and the Gaussian statistics of the interacting waves, we obtain the relations

$$\begin{aligned} \frac{I_1}{\beta_1} = \frac{I_2}{\beta_2} = \frac{I_{30} - I_3}{\beta_3}, \\ \langle I_1 I_2 \rangle &= 2 \frac{\beta_2}{\beta_1} \bar{I}_1^2, \\ \beta_2 \langle I_1 I_3 \rangle &= \beta_1 \langle I_2 I_3 \rangle, \\ \langle I_1 I_3 \rangle &= I_{30} \bar{I}_1 - 2 \frac{\beta_3}{\beta_1} \bar{I}_1^2. \end{aligned} \quad (20)$$

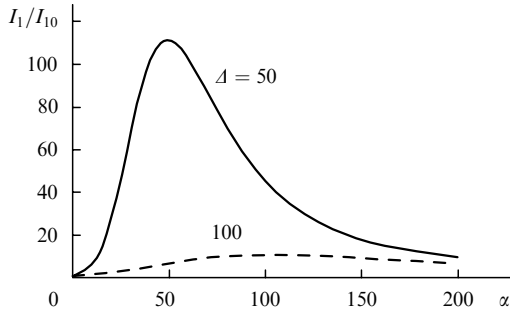
Taking into account (20), the system of equations (19) can be reduced to the form

$$\begin{aligned} \frac{dx_1}{d\zeta} &= 2\varepsilon_1 y_1, \\ \frac{dy_1}{d\zeta} &= -\alpha y_1 - \Delta y_2 + 2\varepsilon_2 x_1 - 6 \frac{\varepsilon_2}{\varepsilon_1} x_1^2, \\ \frac{dy_2}{d\zeta} &= \Delta y_1 - \alpha y_2. \end{aligned} \quad (21)$$

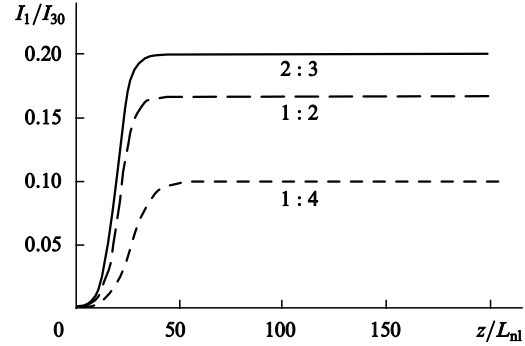
Here  $x_1 = \langle I_1/I_{30} \rangle$ ;  $y_{1,2} = \psi_{1,2}/I_{30}^{3/2}$ ;  $\varepsilon_{1,2} = \beta_{1,2}/\beta_3$ ;  $\zeta = z/L_{nl}$ ;  $\alpha = 2\nu L_{nl}$ ;  $\Delta = \Delta k L_{nl}$ ; and  $L_{nl} = 1/(\beta_3 I_{30}^{1/2})$ .

The numerical solution of the system of equations (21) is presented in Figs 4–7. Figure 4 demonstrates clearly the existence of the optimal relation between the parameter  $\alpha$  characterising the disorder of the domain structure and the phase mismatch  $\Delta$  for which the gain of the signal wave is maximal. In other words, in the case of parametric interaction, as for the second harmonic generation, stochastic quasi-phase matching takes place. The dependence of the signal wave intensity on the interaction length at the initial stage is linear (Fig. 5).

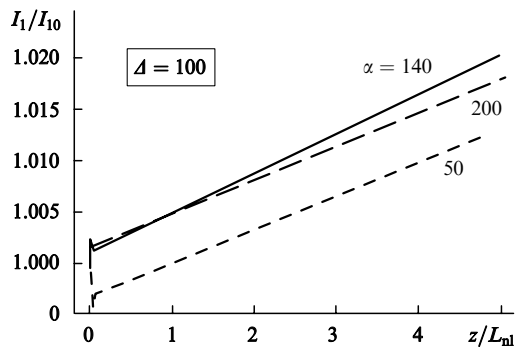
When the condition of stochastic quasi-phase matching at large interaction lengths is fulfilled ( $\Delta = \alpha$ ), the efficiency of the pump-energy conversion to the signal wave can be significant (Fig. 6). Figure 6 also demonstrates the established regime of energy exchange between the interacting waves. The maximum conversion efficiency equal to 0.2 corresponds to the chosen ratio 2 : 3 of the frequencies of the signal and idle waves, for which the dependences presented in Figs 4–6 were calculated.



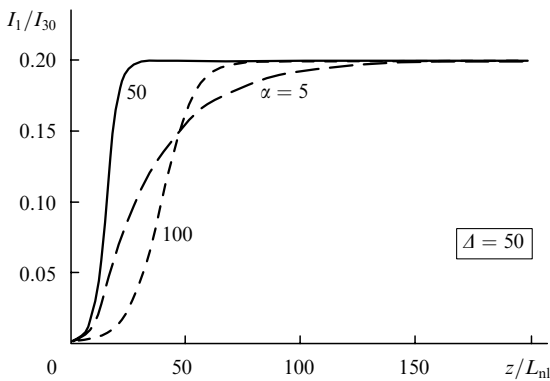
**Figure 4.** Dependences of the signal-wave gain  $I_1(z)/I_{10}$  on the reduced average spatial frequency  $\alpha$  of the sign change in the nonlinear coupling coefficient for different phase mismatches for  $I_1(0) = 10^{-3}$  and  $z/L_{nl} = 50$ .



**Figure 7.** Dependences of the normalised intensity  $I_1(z)/I_{30}$  of the signal wave on the reduced interaction length for different ratios of frequencies ( $\alpha = \Delta = 50$ ).



**Figure 5.** Dependences of the signal-wave gain  $I_1(z)/I_{10}$  on the reduced interaction length for different values of the parameter  $\alpha$  and  $I_1(0) = I_{10} = 10^{-3}$  and  $\Delta = 100$ .



**Figure 6.** Dependences of the normalised intensity  $I_1(z)/I_{30}$  of the signal wave on the reduced interaction length for different values of  $\alpha$  and  $\Delta = 50$ .

Therefore, there is a peculiar dynamic equilibrium in the established regime of nonlinear interaction, when each of the excited waves contains the same number of photons. This is illustrated by curves in Fig. 7, which were plotted for different ratios of the frequencies of the signal and idle waves.

## 5. Conclusions

We have developed the theory of three-frequency nonlinear-optical processes in polydomain crystals with a random

variation in the domain thickness, which is manifested in a random modulation of the nonlinear coupling coefficient of the waves. We simulated a random variation in the nonlinear coefficient by a random telegraph process. The second harmonic generation and the parametric interaction between light waves in these crystals were studied. We found the existence of the condition for stochastic quasi-phase matching, which is satisfied when the phase mismatch between the interacting waves is equal to the doubled average spatial modulation frequency of the nonlinear coefficient ( $\Delta = 2\nu$ ). This condition is similar to the condition of usual quasi-phase matching and corresponds to the most efficient energy exchange between the interacting waves.

It is clear that the aperiodicity of a layered structure reduces the efficiency of nonlinear-optical conversions. However, when the length of a nonlinear crystal is equal to a few tens of nonlinear lengths (which can be realised in the field of ultrashort pulses), the stochastic quasi-phase matching enhances the interaction efficiency compared to interactions in a homogeneous medium in the absence of phase matching.

Let us estimate a scatter in the thickness of domains in a LiNbO<sub>3</sub> crystal for which our theory can be applied. For the crystal length  $z = 1$  cm and the average modulation period of the nonlinear coefficient  $\bar{\lambda} = 20 \mu\text{m}$  ( $\nu = 10^3 \text{ cm}^{-1}$ ), according to (4), the average number of domains is  $\bar{n} = 10^3$ . A variation in the number of domains from crystal to crystal is described by relation (3). The most probable value lies in the interval  $\bar{n} - \sigma_n \leq n \leq \bar{n} + \sigma_n$ . Therefore, the length  $l_d$  of domains lies predominantly in the interval

$$\frac{\bar{\lambda}/2}{1 + 1/\sqrt{\bar{n}}} \leq l_d \leq \frac{\bar{\lambda}/2}{1 - 1/\sqrt{\bar{n}}}.$$

In our case, for  $\bar{n} = 10^3$ , fluctuations of the domain thickness lie within 3%.

The approach we developed for studying the interaction between light waves with a random coupling coefficient can be also used to investigate consecutive nonlinear-optical interactions [2] and nonlinear frequency conversion in the X-ray region [10].

Note that the dependence of nonlinear-optical processes on fluctuations of the phase mismatch caused by weak fluctuations of the optical axis of a crystal (at fixed

nonlinear coefficient) was extensively studied in 1970–1980s [11, 12]. Optical frequency doubling was recently studied taking into account fluctuations of the phase mismatch and the nonlinear coupling coefficient in Ref. [6].

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