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Simulation of the random violation of the quasi-phase-matching condition in optical parametric process

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***Abstract*. The parametric interaction of light waves in a nonlinear photonic crystal (NPC) with the random violation of the quasi-phase-matching condition is considered. A model with correlated fluctuations of the width of adjacent domains is used which corresponds to NPCs fabricated, for example, by the repolarisation method. Both a numerical calculation algorithm and analytic stochastic approach taking into account the influence of fluctuations of the domain width are presented. The behaviour of the average intensity and dispersion of intensity fluctuations of parametrically interacting waves, including the strong energy exchange regime, is studied by the example of a $LiNbO_3$ NPC. The obtained results can be used to estimate the required accuracy of manufacturing the nonlinear structure of the crystal.**

Keywords: *parametric amplification, quasi-phase matched interactions, nonlinear photonic crystal, domain width fluctuations.*

1. Introduction. Formulation of the problem

The first proposals [1 – 3] and realisations [4, 5] of parametric processes in optics in the early 1960s (see also review [6]) were based on phase matched wave interactions in homogeneous nonlinear-optical crystals. At the same time, a considerable extension of applications of parametric processes in a number of optical sources such as, for example, tunable coherent radiation sources, lasers emitting ultrashort pulses, sources of nonclassical light and entangled quantum states of light necessitated the introduction of crystals with the spatially modulated nonlinear susceptibility into the assortment of nonlinear optics. These are crystals with a regular domain structure [which are also called periodically poled nonlinear crystals or nonlinear photonic crystals (NPCs)] and crystals with a chirped nonlinear structure. In such inhomogeneous nonlinear-optical media, the efficient energy exchange between interacting waves occurs if the quasi-phase-matching conditions are fulfilled. In this case, the phase mismatch between them is compensated by the wave vector of the reciprocal nonlinear lattice [7 – 9].

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At present, KTP and $LiNbO_3$ NPCs have found wide applications. Let us emphasise the role of S.A. Akhmanov in the investigations of quasi-phase matched interactions in nonlinear optics. The first experiments on the laser radiation frequency doubling in a polydomain $LiNbO_3$ crystal were performed at the Laboratory of Nonlinear Optics at the Moscow State University in 1966 [10]. On the initiative of S.A. Akhmanov, one of the authors of the present paper, his postgraduate at that time, investigated theoretically this process [11]. S.A. Akhmanov supported the studies of quasi-phase matched optical interactions in the 1980s as well (see, for example, [12, 13]). Beginning from the mid-1990s, researchers at the Laboratory of Quantum and Nonlinear Optics at the MSU are engaged in theoretical and experimental studies of multiwave quasi-phase matched interactions in passive and active crystals with the quadratic optical nonlinearity (see reviews [14, 15]), which are aimed at the development of compact multifrequency lasers and creation of multimode entangled quantum states.

Quasi-phase matched wave interactions provide the universal method for solving the problem of realisation of three-frequency nonlinear-optical processes. For a particular nonlinear process, they in fact eliminate the necessity for searching nonlinear-optical crystals with certain dispersion properties or choosing the geometry of interacting waves. Moreover, coupled multiwave interactions can be performed in aperiodic NPCs, which allows one to realise parametric amplification upon low-frequency pumping [16] and generate higher optical harmonics in quadratically nonlinear crystals [17].

Periodic NPCs can be fabricated by several methods, among which the most popular are the growth method [18] and methods of chemical diffusion and repolarisation [19]. The latter two methods can be used to create the domain structure of a crystal to an arbitrary template, which is especially important for formation of chirped and aperiodic nonlinear structures. However, random deviations can appear during their production, the statistic of these deviations depending on the production method [20]. In the case of manufacturing NPCs by the repolarisation method, a mask used as a template can be fabricated with a high accuracy. But due to somewhat different conditions under which domains are grown (internal mechanical stresses, the crystal thickness, surface defects, etc.), their boundaries can be displaced with respect to the 'ideal' structure. This displacement depends on the properties of the crystal region containing a given domain and is independent of the displacements of other boundaries of adjacent domains. Such random deviations of domain walls in a NPC cause correlated fluctuations of the thickness of adjacent domains. It is clear that if the periodic structure of the NPC is calculated to compensate for the phase mismatch in the nonlinear process, fluctuations of the domain walls will cause a random violation of the phase-matching condition.

The considered model of a random deviation of the NPC structure from a periodic one was used to analyse the proceeding of two coupled nonlinear-optical processes [21]. By describing analytically the interaction of light waves in crystals with a random nonlinear structure, which is manifested in a random spatial modulation of the coupling coefficient of the waves, it is necessary to assume that fluctuations of the 'wave number' of the reciprocal nonlinear lattice are delta-correlated in order to obtain equations for the average intensities of the interacting waves [21]. The calculation of the dispersion of intensity fluctuations for the interacting waves is a rather complicated problem even in the undepleted-pump field approximation. In the case of a strong energy exchange between the waves, when the pump depletion takes place, to obtain closed equations for the average intensities of interacting waves, it is necessary to use the assumption about their statistics [22]. It should be also taken into account that the behaviour of average values in the analysis of stochastic nonlinear processes can be substantially different from the behaviour of an individual realisation. Thus, the nonlinear interaction of light waves in a NPC with random parameters of the nonlinear structure can be correctly analysed only by using the numerical simulation of the interaction process.

The aim of this paper is to develop a numerical algorithm taking into account fluctuations of domain boundaries, which can be used to analyse nonlinear-optical processes in periodic and aperiodic NPCs and chirped nonlinear crystals. As an example, we consider traditional three-frequency parametric process. The average intensities and dispersions of the intensity fluctuations of the interacting waves are calculated. The average wave intensities are also calculated by the analytic method developed in paper [21]. The analytic results are compared with numerical calculations. The results obtained in the present paper allow us to determine the requirements to the accuracy of manufacturing a periodic nonlinear crystal lattice for the efficient realisation of the optical parametric process.

2. Simulation of domain-boundary fluctuations

The modulation of the sign of the nonlinear coupling coefficient for waves in a periodic NPC can be written in the case under study in the form

$$g(z) = \operatorname{sign}\left(\sin\frac{2\pi z}{\Lambda}\right), \qquad (1)$$

where sign x is the signature [$g(x) = 1$ for $x > 0$, $g(x) = -1$ for $x < 0$, and $g(x) = 0$ for $x = 0$]; z is the coordinate along the wave propagation direction; and Λ is the modulation period of the nonlinear lattice.

Our approach is based on the numerical simulation of the three-frequency nonlinear-optical process [see equation (5)] and involves the following operations:

(i) The search for coordinates of the zeroes of the specified function $g(x)$ characterising the dependence of the nonlinear susceptibility sign [for example, determined by expression (1)] and corresponding to the exact quasi-phase matching condition. The search algorithm can be applied both to periodic and aperiodic functions $g(z)$, making this method applicable for structures of any type.

(ii) The construction of a new function $g^{(\mathrm{r})}(z)$, which differs from the 'exact' function $g(z)$. In this case, the coordinate $z_j^{(\mathrm{r})}$ of the jth zero of the function $g^{(\mathrm{r})}(z)$ is a random Gaussian quantity with the average value equal to the coordinate z_j of a zero of the function $g(z)$ (Fig. 1).

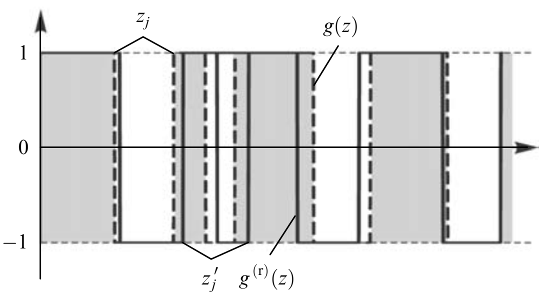

Figure 1. Periodic function $g(z)$ (dashed straight line) and the 'real' dependence $g^{(\mathrm{r})}(z)$ (solid straight line) of the sign of the nonlinear coefficient.

(iii) The numerical solution of the system of truncated equations [(5) in our case] describing the spatial dynamics of the amplitudes of interacting waves for the particular realisation $g^{(\mathrm{r})}(z)$.

(iv) The obtainment of a new realisation of the function $g^{(\mathrm{r})}(z)$, i.e. a random change of the coordinates of its zeroes.

Then, operations 2–4 are repeated, and after the accumulation of an ensemble of realisations, the average values and dispersions of the intensity fluctuations of the interacting waves are calculated for this ensemble.

To perform operation 2, it is necessary to find all the zeroes of the rapidly oscillating function $g(z)$ within the crystal length L. Then, the function $g(z)$ can be represented in the interval $[0, L]$ by the polynomial

$$P(z) = (z - z_0) \times \ldots \times (z - z_m)(z - z_{m+1}) \times \ldots \times (z - z_n), \qquad (2)$$

so that

$$g(z) = \operatorname{sign}[P(z)], \qquad (3)$$

where z_j are the zeroes of $g(z)$ ($z_j \in [0, L]$, $j = \overline{1, n}$).

The search for the zeroes of $g(z)$ is performed numerically with the aim of extending the possibilities of the method and the class of nonlinear structures. We discarded the pairs of zeroes separated by a distance smaller than the minimal width $l_{\min}$ of a domain that can be obtained in experiments. In the case of the repolarisation method, the minimal domain width, for example, for a $LiNbO_3$ crystal is about 1.5 μm [19]. At the same time, the function $g(z)$ was constructed by using the number of points that was sufficient to provide their great amount falling within the minimal possible domain width, which excluded the 'omission' of the zeroes of $g(z)$. A function reconstructed from (3) in such a way in the interval $[0, L]$ coincides with the ideal function $g(z)$, except for domains that cannot be created. As a result, the factors $(z - z_m)(z - z_{m+1})$, for which $|z_m - z_{m+1}| < l_{\min}$, come out from polynomial (2).

The 'real' function $g^{(r)}(z)$ was simulated by introducing a random addition δz_j to z_j:

$$g^{(r)}(z) = \text{sign}\{[z - (z_0 + \delta z_0)] \times \ldots \times [z - (z_n + \delta z_n)]\}$$
$$= \text{sign}\left[\left(z - z_0^{(r)}\right) \times \ldots \times \left(z - z_n^{(r)}\right)\right]. \quad (4)$$

The displacement of the domain boundary with respect to the 'ideal' position can be caused by numerous random factors, and therefore it is reasonable to assume that the distribution of random values of δz_j is Gaussian and the dispersion $\langle \delta z_j \rangle^2$ is the same for all j, i.e. $\langle (\delta z_j)^2 \rangle = (\Delta z)^2$ and $\langle \delta z_j \rangle = 0$. The root-mean-square value of Δz depends on the quality of the nonlinear structure of the crystal.

Note that the algorithm described above nowhere uses the particular form of the function $g(z)$, pointing out only that this is a 'sign function'. All the steps and calculations described above can be performed for any function $g(z)$ and, correspondingly, for any multi-wave process. This universal nature is fundamentally important for studying complicated coupled processes, which is the main goal of the method being developed. However, the application of this method is illustrated in this paper by the example of the traditional quasi-phase matched parametric process, which makes it possible to compare the obtained results with the results of paper [21], where the nonlinear optical process was analysed analytically in the parametric approximation.

3. Three-frequency parametric interaction: a numerical experiment

Consider a three-frequency optical parametric process $\omega_p = \omega_1 + \omega_2$, where ω_p is the frequency of the intense pump wave and $\omega_{1,2}$ are the frequencies of the signal and idler amplified waves.

The quasi-phase-matching condition for this process in a NPC upon collinear interaction has the form $\Delta k = k_p - k_1 - k_2 = 2\pi/\Lambda$. Consider, for example, the e – ee interaction in a $LiNbO_3$ NPC for $\lambda_p = 1.064$ μm and $\lambda_1 = 2$ μm. Then, $\lambda_2 = 2.274$ μm, and the period of changing the sign of the nonlinear susceptibility is $\Lambda = 20.6$ μm when the pump wave propagates perpendicular to the optical axis (calculations are performed by using data [23]). This geometry provides the maximum possible effective nonlinear coupling coefficient for the process under study.

The truncated equations for the three-frequency process considered here (neglecting loses and effects related to higher-order nonlinearities) have the form

$$\frac{dA_p}{dz} = ig^{(r)}(z)\beta_p A_1 A_2 \exp(i\Delta kz),$$
$$\frac{dA_1}{dz} = ig^{(r)}(z)\beta_1 A_p A_2^* \exp(-i\Delta kz), \quad (5)$$
$$\frac{dA_2}{dz} = ig^{(r)}(z)\beta_2 A_p A_1^* \exp(-i\Delta kz),$$

where A_j are the complex amplitudes of the interacting waves and β_j are the nonlinear coupling coefficients [24].

The examples of the solution of this system of equations for the same initial conditions are presented in Fig. 2 for different root-mean-square deviations Δz of the position of domain walls. Note that the chosen value $\Delta z \simeq 3$ mm definitely exceeds the fabrication accuracy of NPCs by the repolarisation method. In other words, the realisation of the dynamics presented in Fig. 2 corresponds to the dynamics of a process in a poor-quality crystal. For the convenience of comparison, the enlarged parts of the dependences are also presented in insets in Fig. 2. One can see that the intensity dynamics for the 'real' structure has a more 'irregular' character, which is related to the violation of phase matching between the waves caused by the violation in the periodicity of the nonlinear structure. Note that, despite considerable deviations from the ideal nonlinear structure, the process proceeds efficiently but is prolonged in space, which is equivalent to the nonlinear interaction length. However, the maximum intensities of the waves with frequencies ω_1 and ω_2 for both realisations coincide. Therefore, the influence of errors in the manufacturing of a nonlinear structure is manifested in the energy exchange rate, but not in the possible efficiency. From the point of view of realisation of a nonlinear process, it should be taken into account that losses of the interacting waves in long crystals are greater and the fabrication of NPCs of length of a few centimetres is a complicated problem.

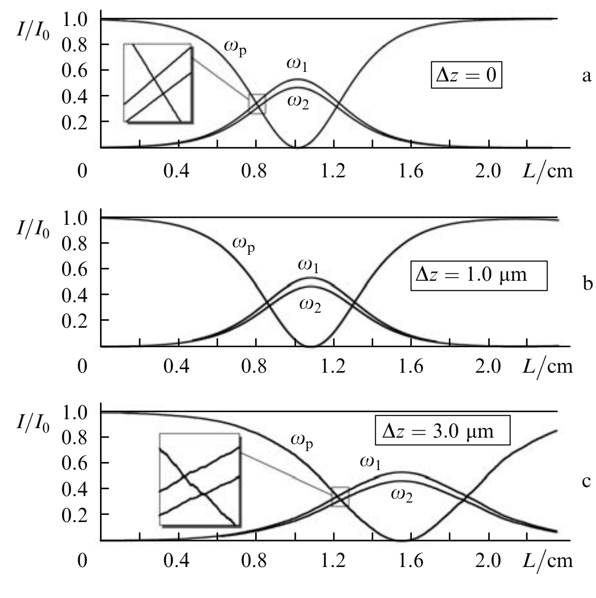

Figure 2. Spatial dynamics of the intensities of interacting waves with frequencies ω_p, ω_1, and ω_2 in the parametric process for different Δz. The curves are constructed for the nonlinear length $L_{nl} = 0.1$ cm; I_0 is the input pump intensity.

Therefore, by realising parametric amplification, it is important to take into account the possible errors in the structure parameters. Thus, to increase the conversion efficiency, it is necessary to select the length of a crystal (for the specified pump power) providing the maximum efficiency. To estimate this length, it is necessary to know the dispersion of fluctuations of the position of domain walls.

Figure 3 presents the results of numerical experiment on the influence of Δz on the energy exchange between the waves during parametric amplification in cross sections at different crystal lengths. Each point is the result of calculation of the system dynamics for 10 realisations of the 'real' function $g^{(r)}(z)$ (4) for fixed Δz; the average values and root-mean-square fluctuations of the intensities of the pump, signal, and idler waves at frequencies $\omega_{1,2}$ are shown. As pointed out above, the quality of the nonlinear structure affects the rate of energy exchange between the waves. Figure 3 shows that the larger the error dispersion in the fabricated structure, the lower the conversion efficiency over the given length of the crystal.

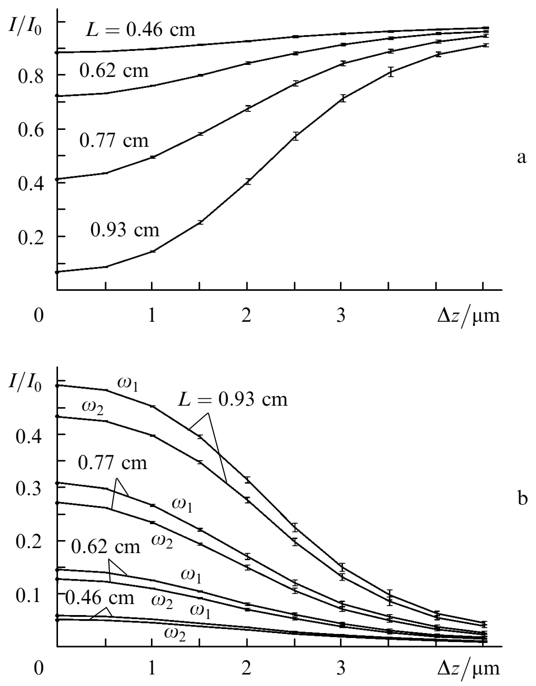

Figure 3. Dependences of the average intensities of the pump (a), signal and idler (b) waves on Δz for different crystal lengths L for $L_{nl} = 0.1$ cm; I_0 is the input pump intensity.

Figure 4 presents the results of numerical experiments on determining the dependence of intensity fluctuations ΔI_j of the interacting waves on Δz. One can see that the value of ΔI_j is smaller for smaller interaction lengths L. At the same time, the value of ΔI_j decreases as the value of Δz is further increased (above 4 μm in our case), which is explained, in our opinion, by a decrease in the energy exchange at the corresponding crystal length for such Δz.

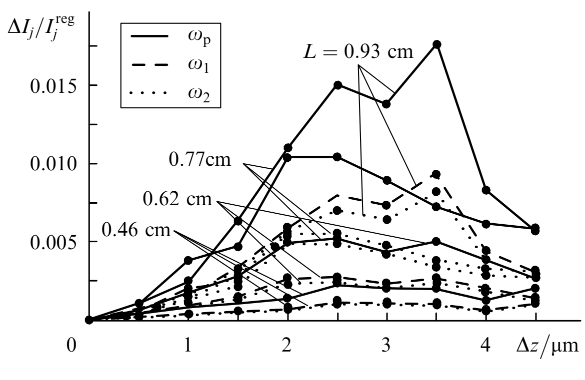

Figure 4. Relative levels of the intensity fluctuations of interacting waves as functions of Δz for different crystal lengths L; $j = \text{p}, 1, 2$; I_j^{reg} is the intensity of the jth wave for the ideal NPC.

4. Undepleted-pump field approximation: the stochastic approach

Analytic results in the study of parametric interaction of waves in a NPC with randomly varying domain width can be obtained by using the undepleted-pump field approximation [equation for the amplitude A_p is omitted in system (5) and we assume that $A_p = \text{const}$ in all other equations]. The system of equations (5) was studied in this parametric approximation for the nonlinear coupling coefficient of the waves simulated by a random telegraph signal [25]. The model of the nonlinear coefficient considered here, corresponding to the correlated fluctuations of the thicknesses of adjacent domains, was used in [21] for studying parametric amplification upon low-frequency pumping, which involves traditional three-frequency parametric amplification processes upon high-frequency pumping and frequency up-conversion. The results obtained in the absence of the latter process are related to the process studied here. However, this particular case was not discussed in [21]. Therefore, we will present below these analytic results, by presenting preliminarily some intermediate calculations.

Let us write the closed system of equations for the signal and idler wave intensities in the undepleted-pump field. According to (5), we have

$$\frac{\mathrm{d}I_1}{\mathrm{d}z} = \mathrm{i}g^{(\mathrm{r})}(z)\gamma_1 B \exp(-\mathrm{i}\Delta kz) + \text{c.c},$$

$$\frac{\mathrm{d}I_2}{\mathrm{d}z} = \mathrm{i}g^{(\mathrm{r})}(z)\gamma_2 B \exp(-\mathrm{i}\Delta kz) + \text{c.c}, \tag{6}$$

$$\frac{\mathrm{d}B}{\mathrm{d}z} = -\mathrm{i}g^{(\mathrm{r})}(z)(\gamma_1 I_2 + \gamma_2 I_1)\exp(\mathrm{i}\Delta kz),$$

where $B = A_1^* A_2^*$ and $\gamma_j = \beta_j A_p$.

Let us apply the method of secondary simplification [13] to the system of equations (6) by integrating equations (6) over the modulation period of $g^{(\mathrm{r})}(z)$ assuming that the intensities of the interacting waves change insignificantly at this spatial scale. Thus, we replace the function $g^{(\mathrm{r})}(z)$, containing the coordinate of the jth wall of a domain, by the quantity $\xi(z_j)$:

$$\xi(z_j) = \frac{1}{\Lambda}\int_{z_j-\Lambda/2}^{z_j+\Lambda/2} g^{(\mathrm{r})}(x)\exp(-\mathrm{i}\Delta kx)\mathrm{d}x, \tag{7}$$

where z_j is determined by the condition $g(z_j) = 0$.

For the periodic nonlinear structure in the first quasi-phase matching order, $\xi(z_j) = 2/\pi$. In the presence of fluctuations in the position of the domain walls, the value of $\xi(z_j)$ changes randomly from domain to domain and has the average value [21]

$$\langle\xi(z_j)\rangle = \mathrm{i}\varepsilon, \quad \varepsilon = \frac{2C(q_m)}{(1+2m)\pi}, \tag{8}$$

where $(1 + 2m)$ is the quasi-phase matching order; $C(q_m)$ is the characteristic function of random quantities δz_j; $C(q_m) = \langle\exp(-\mathrm{i}q_m\delta z_j)\rangle$; and $q_m = 2(1 + 2m)\pi/\Lambda$

Let us introduce the deviation from the average value $\mu(z) = \xi(z) - \langle\xi(z)\rangle$. Then, equations (6) will take the form

$$\frac{dI_1}{dz} = -2\varepsilon\gamma_1 B + 2\gamma_1 \mathrm{Im}[\mu(z)B(z)],$$

$$\frac{dI_2}{dz} = -2\varepsilon\gamma_2 B + 2\gamma_2 \mathrm{Im}[\mu(z)B(z)], \quad (9)$$

$$\frac{dB}{dz} = -[\varepsilon + i\mu^*(z)](\gamma_1 I_2 + \gamma_2 I_1).$$

The averaging of equations (9) reduces the problem to determining the correlators $\langle\mu(z)B(z)\rangle$, $\langle\mu(z)I_1(z)\rangle$, and $\langle\mu(z)I_2(z)\rangle$. Our calculations showed that the correlation length of a random process $\mu(z)$ is equal to the average period of the nonlinear lattice. Because a NPC contains many domains, while the fluctuations of domain widths in a good quality crystal are small, the correlation function of the process $\mu(z)$ can be replaced by the delta function:

$$\overline{\mu(z_1)\mu^*(z_2)} = R_\mu(z_1, z_2) = K\delta(z_2 - z_1). \quad (10)$$

According to results [21], the coefficient $K = 8(\Delta z)^2/\Lambda$.

The delta correlation of the process $\mu(z)$ allows us to use stochastic methods and to apply in this case the Furutzu–Novikov formula [26]:

$$\overline{F\mu(z)} = \int \left\langle \frac{\delta F}{\delta\mu^*} \right\rangle R_\mu(z)dz = K\left\langle \frac{\delta F}{\delta\mu^*} \right\rangle, \quad (11)$$

where $F = F(\mu(z), \mu^*(z))$ is the functional and $\delta F/\delta\mu^*$ is the variational derivative.

As a result, we obtain the system of equations

$$\frac{d\bar{I}_1}{\gamma_1 dz} = \frac{d\bar{I}_2}{\gamma_2 dz} = -2\varepsilon\bar{B} + 2K(\gamma_1\bar{I}_2 + \gamma_2\bar{I}_1),$$
$$(12)$$
$$\frac{d\bar{B}}{dz} = 2\gamma_1\gamma_2 K\bar{B} - \varepsilon(\gamma_1\bar{I}_2 + \gamma_2\bar{I}_1)$$

for the average quantities. By solving equations (12) with the boundary conditions $I_1(z=0) = I_{10}$, $I_2(z=0) = 0$, we obtain the average intensities

$$\bar{I}_1 = [1 + G(z)]I_{10}, \quad \bar{I}_2 = (\gamma_2/\gamma_1)G(z)I_{10}, \quad (13)$$

where

$$G(z) = \frac{1}{2}\exp(3\alpha z)\left[\cosh\Gamma z + \frac{\alpha}{\gamma}\sinh\Gamma z\right] - \frac{1}{2}. \quad (14)$$

Here, $\alpha = K\gamma_1\gamma_2 = 8\gamma_1\gamma_2(\Delta z)^2/\Lambda$; $\Gamma = (4\varepsilon^2\gamma_1\gamma_2 + \alpha^2)^{1/2}$; and the parameter ε is defined in (8).

Consider the proceeding of the process in the first quasi-phase matching order $q_0 = 2\pi/\Lambda$ ($m = 0$). In the absence of fluctuations in the NPC, $\alpha = 0$ and $C(q_0) = 1$, $\varepsilon = 2/\pi$, and we obtain from (13) the well-known result

$$G^{\mathrm{reg}}(z) = \frac{1}{2}\left[\cosh\left(\frac{4}{\pi}\sqrt{\gamma_1\gamma_2}z\right) - 1\right].$$

For Gaussian fluctuations of the domain thickness, we have $C(q_0) = \exp[-2(\pi\Delta z/\Lambda)^2]$. Analysis showed that decrease in the intensity of interacting waves in a NPC with a randomly violated periodicity is caused by the decrease of the parameter ε depending on $C(q_0)$ (8). This is illustrated in Fig. 5, where the results of analytic calculations and numerical simulation are presented. One can see that the results obtained by these two methods are consistent.

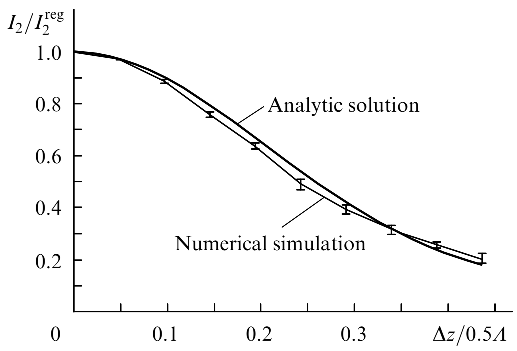

Figure 5. Average intensity of the idler wave normalised to the intensity obtained in the ideal periodic NPC as a function of $\Delta z/(0.5\Lambda)$.

Figure 6 shows how the gain of the signal wave decreases with increasing fluctuations of the domain width and the interaction length. By specifying the crystal length and the level of the gain decrease, we can determine the requirements to the accuracy of crystal fabrication.

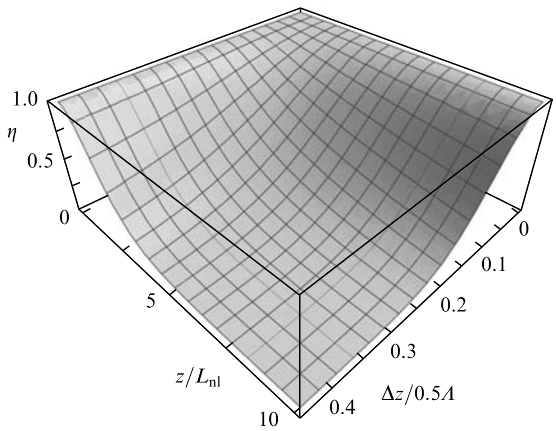

Figure 6. Signal-wave gain $\eta = I_1(z)/I_1^{\mathrm{reg}}(z)$ in the real NPC normalised to the gain in the ideal crystal as a function of the interaction length z and fluctuations of the domain width Δz; $L_{\mathrm{nl}} = \gamma_1^{-1}$.

5. Conclusions

We have studied the influence of fluctuations of the domain width on the parametric interaction of waves in a periodic NPC. The model with correlated fluctuations of the widths of adjacent domains was used, which describes adequately the case of NPCs fabricated by the repolarisation method. The nonlinear process was analysed by the stochastic method both in the undepleted-pump field approximation and by integrating numerically the system of nonlinear equations in the general case, including the nonlinear regime of the interaction of waves. Analysis of the behaviour of the average intensities of the waves has shown that a random deviation of a nonlinear lattice from a periodic lattice does not change the type of energy exchange between the interacting waves, by reducing only the effective interaction length. The intensity fluctuations of the waves were also investigated by using numerical simulations.

The results of the numerical experiment for the average intensities of the waves confirm the validity of the analytic stochastic approach developed in the paper. Analytic results clearly show that the requirements to the accuracy of manufacturing a periodic structure increase with decreasing the nonlinear structure period and increasing the crystal length [see (14), expression for $C(q_0)$, and Fig. 6] and the quasi-phase matching order. The results obtained in the paper can be used to estimate the accuracy required for manufacturing the nonlinear structure of crystals. By specifying the crystal length and the required parametric conversion efficiency, the maximum root-mean-square deviation of the position of the domain walls in NPCs can be determined by the dependences in Figs 3 and 4.

The numerical approach and analytic stochastic method developed in the paper can be used for studying other three-frequency nonlinear-optical processes and various multi-wave processes proceeding in crystals with randomly violated periodicity of the nonlinear structure.

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