Effect of domain structure fluctuations on the photorefractive response of periodically poled lithium niobate

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Abstract

We analyze theoretically the photorefractive response of periodically poled lithium niobate (PPLN) taking into account spatial fluctuations of its domain structure. It is shown that these fluctuations strongly affect the amplitude of space-charge field in the limit of small photorefractive grating vectors. The behaviour of the nonlinear response in this limit is controlled by the root-mean-square of the spatial fluctuations of the domain structure. In the limit of large grating vectors, the nonlinear response is not sensitive to deviations from the ideal periodic geometry. The results obtained can be useful for the interpretation of photorefractive experiments and the formulation of requirements for the fabrication of photorefractive PPLN samples.

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I. INTRODUCTION

During the past several years, the fabrication of high-quality samples of periodically poled lithium niobate (PPLN) has become possible¹⁻⁴. The period of such structures ranges from a few to several tens of micrometers and the number of opposite domains reaches a few thousand. The technological progress was primarily stimulated by the prospects for quasi-phase-matched frequency conversion¹⁻⁴.

Recently it has been shown that PPLN is attractive not only for frequency doubling but also for photorefractive applications⁵⁻⁷. The photorefractive response of PPLN is weak for low spatial frequencies of the exciting light interference patterns and strong for sufficiently small periods of the space-charge gratings. This distinctive feature allows to avoid large-scale distortions of light beams (optical damage) and, at the same time, to exploit the standard schemes of two-wave and four-wave mixing^{8,9} using the advantage of strong photorefractive nonlinearity of lithium niobate. Initial photorefractive experiments have supported these main theoretical conclusions^{10,11}.

Previous theory on photorefractive properties of PPLN^{6,7} was based on the assumption of an ideal periodic domain structure. In practice, the domain structure of PPLN can be far from ideal^{12,13}. Moreover, a number of studies have been made to determine the influence of various deviations from the ideal structure on frequency doubling in PPLN, see e.g., ^{14,15}. The purpose of this paper is to investigate the effect of spatial fluctuations of the domain structure on the photorefractive response of PPLN within the whole range of spatial frequencies relevant to experiments. The problem under study is of practical importance and solutions are needed to formulate the necessary requirements and restrictions for the fabrication of photorefractive PPLN samples.

We develop below two basical statistical models for PPLN domain structure and analyze the photorefractive response of this material for the case of photovoltaic charge transport 16,17 , that is typical of photorefractive LiNbO₃ crystals.

II. FORMULATION OF THE PROBLEM

The geometry of the problem is clear from Fig. 1(a). Two recording light beams with wave vectors $\mathbf{k_1}$ and $\mathbf{k_2}$ are incident symmetrically onto the YZ-face of a PPLN sample. The vector of spontaneous polarization \mathbf{c} is parallel to the Z-axis but it reverses its direction on moving to an adjacent domain (along the coordinate x), though without changing its absolute value. In such a way, we assume a zero width of the domain walls. In average, the PPLN structure is considered symmetric, i.e., positive and negative domains have the same average size $\langle l \rangle$. Such a situation is preferable for photorefractive applications⁷. The thickness of the sample in the x-direction, L, is supposed to be much larger not only than $\langle l \rangle$ but also than the period of the light interference pattern ($\Lambda = 2\pi/K$), where K is the grating vector equal to the difference of the light wave vectors \mathbf{k}_1 and \mathbf{k}_2 , see Fig. 1.

Two basic statistical models of PPLN domain structure (analogous to those used in 15 for analysis of the frequency doubling) will be considered. In the first model (I), the probability for a domain to have size l, which we shall denote by $W_1(l)$, does not depend on the sizes of the other domains. Correspondingly, we have $\int_0^\infty W_1(l) \, dl = 1$ and $\int_0^\infty l W_1(l) \, dl = \langle l \rangle$. This model is relevant to PPLN samples used in 10,11. The structure of inverted domains is expected to be formed during crystal growth and no restrictions are imposed on the dimensions of the samples. This property is favorable for photorefractive applications.

The second model (II) refers to relatively thin (at least in one dimension) PPLN samples obtained, for example, using electric-field domain inversion and photolithographic masks¹⁻⁴. The position of each domain boundary is shifted from the ideal position by a random variable $\Delta(\sec^{15})$. Let $W_2(\Delta)$ be the probability for a domain wall to have a shift Δ . Correspondingly, we have $\int_{-\infty}^{\infty} W_2(\Delta) d\Delta = 1$ and $\int_{-\infty}^{\infty} \Delta W_2(\Delta) d\Delta = 0$. Note that $W_2(\Delta) = 0$ for $|\Delta| \ge \langle l \rangle/2$ to avoid "negative-size" domains. The sizes of successive domains cannot fluctuate independently within this model.

The density of electric current, responsible for grating formation, is described as:

$$\mathbf{j} = \kappa I \mathbf{E} + \beta I s(x) \mathbf{z} \,, \tag{1}$$

where I is the light intensity, E the electric field vector, κ the specific photoconductivity, β the photovoltaic constant, z a unit vector parallel to the Z-axis, and s(x) is a random alternating function ($s^2 = 1$) whose sign is opposite for positive and negative domains, see Fig. 1(b). In accordance with Fig. 1(a), the light intensity I is a function of z, and is represented as $I = I_0 (1 + m \cos Kz)$, where I_0 is the average intensity and m is the light modulation.

The first and the second terms on the right-hand side of Eq. (1) correspond to the photoconductivity and the photovoltaic effect^{16,17}, respectively. The direction of the photovoltaic
current coincides with the direction of the polar axis \mathbf{c} , i.e., it is opposite in positive and
negative domains. We have omitted the diffusion contribution to the current density in
Eq. (1), because it is small compared to the photovoltaic current. The ratio $E_{pv} = \beta/\kappa$ is a
measurable parameter, the so-called photovoltaic field. This field approaches 10^2 kV/cm in
LiNbO₃, which is much greater than the characteristic diffusion field. Large values of E_{pv} ensure strong photorefractive nonlinearity in lithium niobate crystals ^{16,17}.

The random alternating function s(x) defines not only the sign of the photovoltaic current but also the sign of the electrooptic coefficient. If r is a tabulated electrooptic constant (equal to r_{13} and r_{33} for ordinary and extraordinary light waves, respectively), then the field-induced change of refractive index n is $\delta n(x) = -n^3 r E_z(x) s(x)/2^{8.9}$.

III. PHOTOREFRACTIVE GRATING AMPLITUDE

The introduced basic phenomenological relations are sufficient for further description of the photorefractive response. Our aim is to express the light-induced change $\delta n(x,z)$ through the function s(x) within the standard linear approximation in the light modulation.

In order to find the steady-state distribution of the light-induced space-charge field $\mathbf{E}(\mathbf{x}, \mathbf{z})$, we use the Maxwell relation $\nabla \cdot \mathbf{j} = 0$ for the current density and express the vector \mathbf{E} through the scalar electric potential φ , $\mathbf{E} = -\nabla \varphi$. Then we represent the potential $\varphi = \varphi(x, z)$ as

$$\varphi = \varphi_K(x) e^{iKz} + c. c.$$
 (2)

Using the introduced definitions and Eqs. (1), (2), we obtain for the amplitude $\varphi_K(x)$:

$$\frac{d^2\varphi_K}{dx^2} - K^2\varphi_K = \frac{i}{2} \, m \, K \, E_{pv} \, s(x) \,. \tag{3}$$

From this we can find the relationship between the Fourier components (in x) $\varphi_{K,q}$ and s_q :

$$\varphi_{K,q} = -\frac{i}{2} \frac{mKE_{pv}}{K^2 + q^2} s_q .$$
(4)

Correspondingly, we have for the z-component of the field amplitude $E_{z,K} = -iK\varphi_K$:

$$E_{z,K} = -\frac{m}{2} K^2 E_{pv} \int \frac{s_q \exp(iqx)}{q^2 + K^2} dq .$$
 (5)

By multiplying $E_{z,K}$ by the factor $n^3 r \varepsilon(x)/2$, we obtain an expression for the amplitude of the refractive index $\delta n_K(x)$. In the next step, we perform the inverse Fourier transformation from s_q to s(x'). Then, using the identity

$$\int_{-\infty}^{\infty} \frac{\exp(iq\rho)}{q^2 + K^2} dq = \frac{\pi}{K} \exp(-K|\rho|) , \qquad (6)$$

we finally find to the following important relation for $\delta n_K(x)$:

$$\delta n_K = \frac{1}{8} mrn^3 K E_{pv} \int s(x) s(x') \exp(-K|x - x'|) dx'.$$
 (7)

The photorefractive properties of PPLN, such as the diffraction efficiency of the recorded grating and beam coupling, are characterized by the value $\langle \delta n_K \rangle_x$ averaged over x. As follows from Eq. (7), this fundamental characteristic can be presented in a form similar to that for the single-domain case^{8,9},

$$\langle \delta n_K \rangle_x = \frac{1}{4} mrn^3 E_{pv} K \chi_K , \qquad (8)$$

where χ_K is the Laplace transform of the correlation function of the domain structure $\chi(\rho)$,

$$\chi_K = \int_0^\infty \chi(\rho) e^{-K\rho} \, d\rho, \qquad \chi(\rho) = \langle s(x) \, s(x+\rho) \rangle_x \,, \tag{9}$$

where $\langle ... \rangle_x$ denotes the average over x. In such a way, we have expressed $\langle \delta n_K \rangle_x$ through the statistical characteristic $\chi(\rho)$. In the single-domain case, where s(x)=1, we have $K\chi_K=1$. In the case of a periodic domain structure one can find from Eq. (13) of Ref. 7 that

$$K\chi_K = 1 - \frac{2}{Kd} \tanh\left(\frac{Kd}{2}\right) , \qquad (10)$$

where d is the uniform domain size.

Below we express the function $K\chi_K$ in terms of the distribution functions $W_1(l)$ and $W_2(\Delta)$ corresponding to the statistical models introduced in Section 2 and analyze the effect of the spatial fluctuations on the K-dependence of the photorefractive response.

IV. STATISTICAL AVERAGING

As a first step, using Eqs. (9) we represent χ_K in the form:

$$\chi_K = \frac{1}{L} \int_0^L s(x) \, e^{Kx} \int_x^\infty s(\rho) \, e^{-K\rho} \, d\rho \, dx \,. \tag{11}$$

Since s(x) is a step-like function, see Fig. 1(b), we can replace the integral over x by a sum of integrals over all the domains,

$$\int_0^L = \int_0^{x_1} + \int_{x_1}^{x_2} + \dots + \int_{x_{N-1}}^L , \qquad (12)$$

with $\vec{N} \simeq L/\langle l \rangle \gg 1$ being the total number of domains.

Simple calculations lead to the following expression for a particular integral:

$$\int_{x_i}^{x_{i+1}} = \frac{x_{i+1} - x_i}{K} + \frac{2}{K^2} \left(e^{Kx_{i+1}} - e^{Kx_i} \right) \left[\sum_{j=i+1}^{N} (-1)^{(j-i)} e^{-Kx_j} \right] , \tag{13}$$

where $0 \le i \le N-1$, $x_0 = 0$ and $x_N = L$.

Hence, by adding all the contributions given by Eq. (13) from i = 0 to i = N - 1, we may write Eq. (12) as

$$\int_{0}^{L} = \frac{L}{K} + \frac{2}{K^{2}} \left[-N + e^{-Kl_{1}} + 2e^{-Kl_{2}} + 2e^{-Kl_{3}} + \dots + 2e^{-Kl_{N}} - e^{-Kl_{1}} e^{-Kl_{2}} - 2e^{-Kl_{2}} e^{-Kl_{3}} - 2e^{-Kl_{3}} e^{-Kl_{4}} \dots - 2e^{-Kl_{N-1}} e^{-Kl_{N}} + \dots - e^{-Kl_{1}} e^{-Kl_{2}} \dots e^{-Kl_{N}} \right],$$
(14)

where $l_i = x_i - x_{i-1}$ is the size of the i^{th} domain. This relation is valid for any statistical model of PPLN domain structure.

A. Model I

Let us consider now the case of independently fluctuating domain sizes. The total number of exponents (like e^{-Kl_i}) in the first line of Eq. (14) is 2N-1. If we divide the sum of these exponents by N, the result is equal, with high accuracy, to

$$2\int e^{-Kl} W_1(l) dl \equiv 2W_1(K) . {15}$$

i.e., to the Laplace transform of the distribution function multiplied by a factor 2.

The second line contains products of two exponents, like $e^{-Kl_2}e^{-Kl_3}$. The total number of such terms is 2N-3. Since the probability for a i^{th} domain to have a length l does not depend on the sizes of other domains, the sum of the terms of the second line divided by N is $-2W_1^2(K)$ with high accuracy. Analogously, the sum (divided by N) of the terms including products of three exponents is $2W_1^3(K)$. The generalization to the sum of products of p exponents, like $e^{-Kl_1}e^{-Kl_2}\cdots e^{-Kl_p}$, yields $2(-1)^{p+1}W_1^p(K)$ with an error of the order of $\sim p/N$, which becomes significant for $p \lesssim N$. It is natural to assume, however, that the number of essential terms in the geometric progression $W_1(K) - W_1^2(K) + W_1^3(K) - \cdots$ is much less than $N \equiv L/\langle l \rangle$ (we refer to the appendix for further details of the whole procedure). With this approximation, we obtain from Eqs. (11) and (14) the following explicit expression for $K\chi_K$:

$$K\chi_K = 1 - \frac{2}{K\langle l \rangle} \left[\frac{1 - W_1(K)}{1 + W_1(K)} \right]$$
 (16)

B. Model II

For the case of fluctuating domain wall positions, we represent the wall position x_i as $x_i = i\langle l \rangle + \Delta_i$. By substituting this ansatz into Eq. (14), separating the products of different exponents (like e^{Δ_i} , e^{Δ_i} e^{Δ_j}), and keeping in mind that the deviations Δ_i fluctuate independently, we find the expression

$$K\chi_K = 1 - \frac{2}{K\langle l \rangle} \left[1 - \frac{2W_2(K)W_2(-K)}{1 + e^{K\langle l \rangle}} \right] , \qquad (17)$$

where $W_2(K) = \int_{-\infty}^{\infty} e^{-K\Delta} W_2(\Delta) d\Delta$.

Equations (16) and (17) are the results we were looking for.

V. DISCUSSION

Let us consider first some general properties of expression (16) for $K\chi_K$. If we assume $W_1(l) = \delta(l-d)$, which corresponds to the ideal periodic case with all domain sizes $d = \langle l \rangle$ (δ is the Dirac function), we return immediately to the known result given by Eq (10).

Since the ratio $(1-W_1(K))/(1+W_1(K))$ in Eq. (16) is smaller than 1, the product $K\chi_K$ becomes close to 1 for $K\langle l\rangle\gg 1$. In other words, for sufficiently small grating spacings the photorefractive properties of the random structure are not much different from those of single-domain crystals or PPLN samples with an ideal periodic structure.

In the opposite case of small grating vectors, $K\langle l\rangle\ll 1$, we can restrict ourselves to the first terms of the Taylor expansion of the exponential in the integrand of Eq. (15). Then $W_1(K)=1-K\langle l\rangle+0.5K^2\langle l^2\rangle\cdots$, and one can find from Eq. (16) that

$$K\chi_K \simeq \frac{K}{2} \frac{\langle l^2 \rangle - \langle l \rangle^2}{\langle l \rangle} \,.$$
 (18)

We see that the K-dependence of the photorefractive response is linear in the limit of small grating vectors, whereas for the ideal periodic structure it is quadratic (see Eq. (10)). The slope of the linear function of K given by Eq. (18) is fully defined by the root-mean-square deviation of the domain size.

Let us now consider the statistical case designated as model II. The general properties of the photorefractive response given by Eq. (17) are similar to those described above. To return to the ideal periodic case, we should set $W_2(\Delta) = \delta(\Delta)$. In the limit of small grating spacings we have again $K\chi_K \simeq 1$ and for large grating spacings, $K\langle l \rangle \ll 1$, we have

$$K\chi_K \simeq \frac{2K\langle \Delta^2 \rangle}{\langle l \rangle}$$
 (19)

where the mean-square value $\langle \Delta^2 \rangle = \int \Delta^2 W_2(\Delta) d\Delta$ characterizes the dispersion of the positions of the domain walls. Note that the slope of the linear dependence (in K) is here more sensitive to the mean-square value than in the previous model (compare Eq. (19) with Eq. (18)). On the other hand, the typical values of $\langle \Delta^2 \rangle$ are expected to be smaller than those of $\langle l^2 \rangle - \langle l \rangle^2$ in agreement with the explanations given in the Introduction.

The increase of photorefractive response in the limit of small grating vectors, caused by domain-size dispersion, has a simple physical explanation. Because of the dispersion, relatively large domains (with sizes greater than $\langle l \rangle$) are present within each grating period $\Lambda = 2\pi/K$ and these domains increase the response because of its strong spectral dependence⁷. The larger Λ , the stronger is this enhancement.

If we consider a particular distribution $W_1(l)$ (or $W_2(\Delta)$), we can analyze the dependence $K\chi_K$ in the whole range of grating vectors. Let us consider first the model of fluctuating domain sizes and choose $W_1(l)$ in the form

$$W_1(l) = \frac{(Ql/\langle l \rangle)^Q}{l\Gamma(Q)} \exp\left(-Ql/\langle l \rangle\right), \qquad (20)$$

where Q is a variable parameter larger than 1 and $\Gamma(Q)$ is the Gamma function of Q. In this particular case it is possible a complete analytical study. It is easy to verify that $W_1(0) = 0$, $\int_0^\infty W_1(l) dl = 1$, $\int_0^\infty l W_1(l) dl = \langle l \rangle$, and

$$Q^{-1} = \frac{\langle l^2 \rangle - \langle l \rangle^2}{\langle l \rangle^2} \,. \tag{21}$$

In such a way, Q^{-1} is the relative dispersion of the domain sizes. In the limit $Q \to \infty$ the distribution function $W_1(l)$ tends to the delta function $\delta(l-\langle l \rangle)$. Figure 2 shows the function

 $W_1(l)$ for several representative values of dispersion. Using Eq. (21), it is not difficult to calculate the Laplace transform of the distribution function,

$$W_1(K) = \left(1 + \frac{K\langle l \rangle}{Q}\right)^{-Q} . \tag{22}$$

In the limit of small K it produces the known result for $K\chi_K$, see Eq. (18).

Figure 3 shows the normalized dependence of the photorefractive response of PPLN for several values of Q within a wide interval of $K\langle l\rangle$. The lower curve corresponds to the periodic case. One sees that the larger the dispersion Q^{-1} the higher is the curve. At the same time, in the region of sufficiently large grating vectors $(K\langle l\rangle \gtrsim 10)$ the difference between periodic and random structures is very small even for strong fluctuations (large dispersion). However, in the limit $K\langle l\rangle \ll 1$, the response of a random domain structure is much higher than that of PPLN.

In the case of fluctuating positions of domain boundaries (model II), we use the particular distribution $W_2(\Delta) = \Theta(\Delta_0^2 - \Delta^2)/2\Delta_0$, where $\Theta(x) = 1$ for x > 0 and 0 for x < 0, thus $W_2(l) = (\sinh K\Delta_0)/K\Delta_0$. The parameter Δ_0 does not exceed $\langle l \rangle/2$ and $\langle \Delta^2 \rangle = \Delta_0^2/3$. Figure 4 shows the normalized K-dependence of the photorefractive response for several values of Δ_0 . Qualitatively, the main features of this dependence are similar to those presented in Fig. 3. For $\Delta_0 \lesssim 0.2 \langle l \rangle$, which is the case of most PPLN samples obtained with the use of photolithographic masks, the effect of fluctuations is significant only for very large gratings periods, $\Lambda \gtrsim (5-6)\langle l \rangle$.

VI. CONCLUSIONS

Using two representative models for random domain structure of LiNbO₃, we have expressed the photorefractive response of PPLN through the relevant statistical characteristics and investigated the effect of spatial fluctuations within a wide range of grating vectors. We have found that these spatial fluctuations strongly increase the response in the limit of small grating vectors whereas a negligible effect is produced for sufficiently large grating vectors. Our results indicate that the requirements for the quality of PPLN samples,

necessary for photorefractive applications, are considerably less strict than those for the quasi-phase-matched frequency conversion.

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APPENDIX A

This appendix is devoted to a more detailed derivation of Eq. (16). From Eqs. (11) and (14) we have

$$K\chi_{K} = 1 + \frac{2}{KL} \left\{ -N + \left[e^{-Kl_{1}} + 2\sum_{i=2}^{N} e^{-Kl_{i}} \right] - \left[e^{-Kl_{1}} e^{-Kl_{2}} + 2\sum_{i=3}^{N} e^{-Kl_{i-1}} e^{-Kl_{i}} \right] + \dots + \left(-1 \right)^{j+1} \left[\prod_{r=1}^{j} e^{-Kl_{r}} + 2\sum_{i=j+1}^{N} \left(\prod_{r=i}^{j+i-1} e^{-Kl_{r}} \right) \right] + \dots + \left(-1 \right)^{N+1} \prod_{r=1}^{N} e^{-Kl_{r}} \right\}.$$
(A1)

Let us consider the terms inside the first square brackets in Eq. (A1):

$$e^{-Kl_1} + 2\sum_{i=2}^{N} e^{-Kl_i} = -e^{-Kl_1} + 2\sum_{i=1}^{N} e^{-Kl_i} \simeq -e^{-Kl_1} + 2N\int_0^\infty e^{-Kl} W_1(l) dl$$
 (A2)

Owing to the fact that N is very large the sum can be replaced by an integral containing $W_1(l)$ which is the distribution function of the sizes of domains. This integral, as was mentioned above, corresponds to the Laplace transform $W_1(K)$.

Let us now take into account the terms inside the second square brackets in Eq. (A1). Since l_i and l_{i-1} are random variables, we can write

$$\sum_{i=2}^{N} e^{-Kl_{i-1}} e^{-Kl_i} \simeq (N-1) \int_0^\infty \int_0^\infty e^{-Kl} e^{-Kl'} W_1^{(2)}(l,l') \, dl \, dl' \,, \tag{A3}$$

where $W_1^{(2)}(l, l')$ is the joint distribution function of l and l'. If the continuous random variables l and l' are independent, which is equivalent to say that the probability for a

certain domain i to have a given length does not depend on the sizes of other domains, then $W_1^{(2)}(l,l')$ can be expressed as a product of the corresponding distribution functions $W_1(l)$ and $W_1(l')$ of l and l', respectively. Therefore, the double integral in Eq. (A3) is equal to $W_1^2(K)$, and

$$e^{-Kl_1} e^{-Kl_2} + 2 \sum_{i=3}^{N} e^{-Kl_{i-1}} e^{-Kl_i} \simeq -e^{-Kl_1} e^{-Kl_2} + 2(N-1)W_1^2(K)$$
 (A4)

Combining the first and the second square brackets we have

$$\left[e^{-Kl_1} + 2\sum_{i=2}^{N} e^{-Kl_i}\right] - \left[e^{-Kl_1} e^{-Kl_2} + 2\sum_{i=3}^{N} e^{-Kl_{i-1}} e^{-Kl_i}\right]$$

$$\approx 2\left[NW_1(K) - (N-1)W_1^2(K)\right] - (1 - e^{-Kl_2}) e^{-Kl_1}. \tag{A5}$$

Extending the above procedure to other pairs of consecutive terms j and j+1 we obtain

$$\left[\prod_{r=1}^{j} e^{-Kl_r} + 2 \sum_{i=j+1}^{N} \left(\prod_{r=i}^{j+i-1} e^{-Kl_r} \right) \right] - \left[\prod_{r=1}^{j+1} e^{-Kl_r} e^{-Kl_2} + 2 \sum_{i=j+2}^{N} \left(\prod_{r=i}^{j+i} e^{-Kl_r} \right) \right]
\simeq 2 \left[(N-j+1)W_K^j - (N-j)W_1^{j+1}(K) \right] - (1-e^{-Kl_{j+1}}) \prod_{r=1}^{j} e^{-Kl_r} .$$
(A6)

We see from Eq. (A6) that the approximation $\sum_{i=j+1}^{N} (\prod_{r=i}^{j+i-1} e^{-Kl_r}) \simeq (N-j+1)W_1^j(K)$ is getting worse as j increases. However, it is obvious that its relative importance decreases because the number of members in the sum is smaller and, in addition, the approximation $\prod_{r=i}^{j+i-1} e^{-Kl_r} \simeq e^{-jK\langle l \rangle}$ (which tends to be more accurate) becomes much smaller than 1 owing to the exponent j.

Hence, Eq. (A1) can be cast in the form

$$K\chi_K \simeq 1 + \frac{2}{KL} \left\{ -N + 2 \sum_{i=1}^{N-1} \left[(-1)^{j+1} \left(N - j + 1 \right) W_1^j(K) \right] - \Delta_K^{(N)} \right\} ,$$
 (A7)

where $\Delta_K^{(N)}$ denotes a small contribution coming from the sum of each of the last terms in Eq. (A6):

$$\Delta_{K}^{(N)} = (1 - e^{-Kl_{2}}) e^{-Kl_{1}} + (1 - e^{-Kl_{4}}) e^{-Kl_{1}} e^{-Kl_{2}} e^{-Kl_{3}} + \dots +$$

$$+ (1 - e^{-Kl_{2j}}) \prod_{r=1}^{2j-1} e^{-Kl_{r}} + \dots + (-1)^{N+1} \prod_{r=1}^{N} e^{-Kl_{r}}$$

$$< (1 - e^{-Kl_{2}}) e^{-Kl_{1}} + (1 - e^{-Kl_{3}}) e^{-Kl_{1}} e^{-Kl_{2}}$$

$$+ (1 - e^{-Kl_{4}}) e^{-Kl_{1}} e^{-Kl_{2}} e^{-Kl_{3}} + (1 - e^{-Kl_{5}}) e^{-Kl_{1}} e^{-Kl_{2}} e^{-Kl_{3}} e^{-Kl_{4}} + \dots$$

$$= e^{-Kl_{1}} + (-1)^{N+1} \prod_{r=1}^{N} e^{-Kl_{r}} < 1. \tag{A8}$$

Therefore, by neglecting $\Delta_K^{(N)}$ in Eq. (A7) and calculating the geometric series, we easily find

$$K\chi_K \simeq 1 + \frac{2}{KL} \left\{ N - \frac{2N}{1 + W_1(K)} - \frac{2W_1(K)}{(1 + W_1(K))^2} \right\} ,$$
 (A9)

where we have omitted terms $NW_1(K)^N$ (because $W_1(K) < 1$). Finally, by using the equality $\langle l \rangle = L/N$ we obtain Eq. (16).

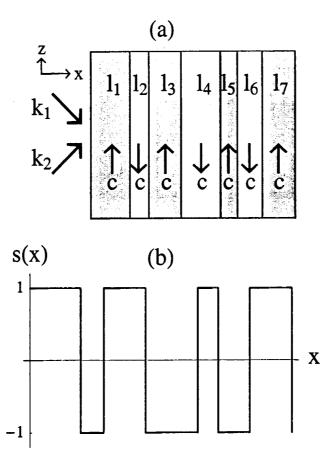
REFERENCES

- ¹ M. Yamada, N. Nada, M. Saitoh, and K. Watanable, Appl. Phys. Lett. 62, 435 (1993).
- ² W. K. Burns, W. McElhanon, and L. Goldberg, IEEE Photonics Technol. Lett. **6**, 252 (1994).
- ³ J. Webjörn, V. Pruneri, P. St. Russell, J. R. M. Barr, and D. C. Hanna, Electron. Lett. **30**, 894 (1994).
- ⁴ L. E. Myers, R. C. Eckardt, N. M. Fejer, R. L. Byer, W. R. Bosenberg, and J. R. Pierce, J. Opt. Soc. Am. B **12**, 2102 (1995).
- ⁵ V. Pruneri, P. G. Kazansky, J. Webjörn, P. St. Russel, and D. Hanna, Appl. Phys. Lett. **67**, 1957 (1995).
- ⁶ B. Sturman, M. Aguilar, F. Agulló-López, V. Pruneri, P. Kazansky, and D. C. Hanna, Appl. Phys. Lett. 69, 1349 (1996).
- ⁷ B. Sturman, M. Aguilar, F. Agulló-López, V. Pruneri, and P. Kazansky, J. Opt. Soc. Am. B 14, 2641 (1997).
- ⁸ P. Günter and J.-P. Huignard, eds. in *Photorefractive Materials and Their Applications I*, Vol. 61 of *Topics in Applied Physics* (Springer-Verlag, Berlin, 1988).
- ⁹ L. Solymar, D. J. Webb, and Grunnet-Jepsen, in *The Physics and Applications of Photorefractive Materials* (Claredon, Oxford, 1996).
- ¹⁰ S. Odoulov, T. Tarabrova, A. Shumelyuk, I. I. Naumova, and T. O. Chaplina, Phys. Rev. Lett. 84, 3294 (2000).
- ¹¹ S. Odoulov, T. Tarabrova, A. Shumelyuk, I. I. Naumova, and T. O. Chaplina, Proceedings of E-MRS Meeting on Photorefractive Materials (Strasbourg, France, 2000).
- ¹²G. Arvidsson and B. Jaskorzynska, AIP Conf. Ser. 103, 47 (1989).

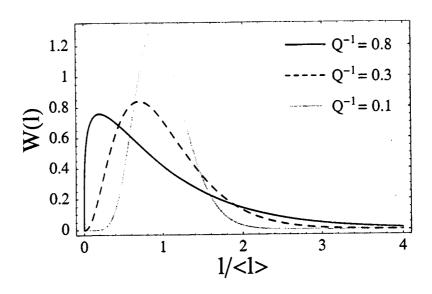
- ¹³ I. I. Naumova, N. F. Elanova, S. A. Blokhin, and S. V. Lavrishchev, J. Crystal Growth, 187, 102 (1998).
- ¹⁴ T. Suhara and H. Nishihara, IEEE J. Quantum Electron. 26, 1265 (1990).
- $^{15}\,\mathrm{S.}$ Helmfrid and G. Arvidsson, J. Opt. Soc. Am. B 8, 797, 1991.
- ¹⁶ A. M. Glass, D. Von der Linde, and T. J. Negran, Appl. Phys. Lett. **25**, 233 (1974).
- ¹⁷ B. I. Sturman and V. M. Fridkin, The Photovoltaic and Photorefractive Effects in Noncentrosymmetric Materials (Gordon and Breach, Philadelphia, 1992).

Figure captions

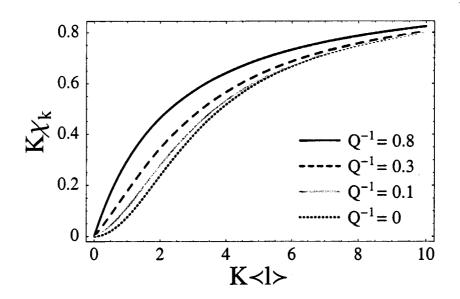
- Fig. 1. (a) Geometrical scheme of a photorefractive experiment using PPLN. (b) Schematic dependence s(x) for a random domain structure.
- Fig. 2. $W_1(l)$ as a function of l for different values of dispersion Q^{-1} .
- Fig. 3. Spectral dependence of the photorefractive response for the case of fluctuating domain sizes (model I). The lower curve corresponds to the periodic case, $Q^{-1} = 0$.
- Fig. 4. Spectral dependence of the photorefractive response for the case of fluctuating positions of the domain walls (model II). The lower curve is plotted for the periodic case, $\Delta_0 = 0$.



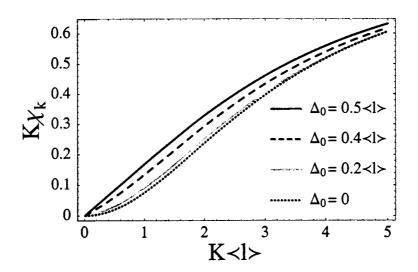
PRBPodivilov et al. Figure 1



PRBPodivilov et al. Figure 2



PRBPodivilov et al. Figure 3



PRBPodivilov et al. Figure 4