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Investigation of nonlinearity of photorefractive effect in $LiNbO_3$

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In this paper, the nonlinearities resulting from the simple band transport model of photorefractivity are compared with dependences experimentally obtained in $LiNbO_3$:Fe crystals. The investigation of nonlinearities is performed for both periodic and aperiodic optical fields. The comparison shows that the space-charge field cannot be the main cause of light-induced refractive index changes.

Keywords: photorefractive effect, lithium niobate

1. Introduction

Although the photorefractive effect (PRE) in $LiNbO_3$ crystals has recently been in the focus of interest mostly due to its usefulness for optical memories [1, 2] or construction of elements for optical circuits [3, 4], there is still space for studying its nature, after more than forty years from the first reference to the effect, *e.g.*, [5, 6].

Today, there are several models known describing the photorefractivity of crystals but as default they all consider the electric field built up by redistribution of the charge carriers and electrooptic effect (EOE) as dominant mechanisms responsible for the light-induced refractive index inhomogeneities [7–9]. The decades-long investigations of the kinetics of the recording process by means of illumination with periodic (harmonic) spatial distribution of intensity were carried out in order to come to these conclusions but, to best of our knowledge, only rarely was the attention paid to application of the aperiodic illumination which gives more complex information on the origin of PRE. However, also in the cases where aperiodic illumination is used one has to take care of the proper choice of the spatial distribution of the illumination in order to get as unambiguous and convincing results as possible. For example, an illumination used in [10, 11] was aperiodic one, 2-D spatially distributed in its nature. Thus, the observed results can be due to a combination of several effects which arise in 2-D case but do not occur in 1-D case.

Information about the origin of the refractive index changes can be obtained from study of spatial symmetry of the refractive index distribution (partly done in [12]) and from study of nonlinearity of the effect which is the topic of this paper.

2. Band transport model of PRE

The commonly used models of PRE, *e.g.* [7], are based on rate equations for light--excited charge carriers provided by certain types of donors (impurities or imperfections) into the conduction (and/or valence) band, continuity equation, Poisson's equation and relation describing the connection between electric field and refractive index change. Assuming a one-level model and a simple 1-D case we can write

$$\frac{\partial n_D(t,z)}{\partial t} = -g_D I(z) n_D(t,z) + r_D n_c(t,z)\left[N_D - n_D(t,z)\right] \tag{1}$$

$$\frac{\partial n_c(t,z)}{\partial t} = -\frac{\partial n_D(t,z)}{\partial t} + \frac{1}{e}\frac{\partial i(t,z)}{\partial z} \tag{2}$$

$$i_z = D_z \frac{\partial n_c}{\partial z} + n_c \mu E_z + \beta_z I(z) \tag{3}$$

where n_D is the density of electrons at donor level, n_c is the density of electrons in the conduction band, e is the elementary charge, i_z is the total current density, D_z is the diffusion coefficient in the direction considered, μ is the electron mobility, $I(z)$ is the intensity of illumination, g_D and r_D are the probabilities of generation and trapping of the electrons, respectively. N_D is the total density of donor states, β_z is the photovoltaic constant, and E_z is the strength of the electric field. The subscript z emphasizes 1-D situation, with z referring to direction of optical axis of the crystal. The internal electric field is produced by the space charge ρ, the density of which is given by the continuity equation

$$\frac{\mathrm{d}\rho}{\mathrm{d}t} = -\frac{\mathrm{d}i_z}{\mathrm{d}z} \tag{4}$$

and can be obtained from Poisson's equation which is written in the form

$$\mathrm{div}(D_l) = \rho \tag{5}$$

where D_l is the electric displacement vector related to electric intensity vector by $E_k = \eta_{k,l} D_l$, $\eta_{k,l}$ is the impermittivity tensor and k, $l = 1, 2, 3$. In the models used it is assumed that the refractive index is modulated by the electric field through Pockel's effect. It means that the refractive index can be expressed through impermittivity tensor

$$\eta_{kl} = \eta_{0kl} + r_{jkl} E_l$$

From the previous equation and according to $3m$ symmetry of the $LiNbO_3$ in 1-D case one gets, *e.g.* [13]

$$n_e = n_{e0} + \frac{1}{2} n_{e0}^3 r_{33} E_3 \tag{6}$$

for the refractive index of the beam polarized in parallel to z-axis, where n_e and n_{e0} denote the extraordinary refractive index of the crystal with and without electric field, respectively, and r_{33} is the corresponding electrooptic coefficient.

Following the model of PRE described by Eqs. (1)–(6) the reasons for the nonlinearity may be:

a) a nonlinear dependence of the generation rate of carriers on sample illumination (the light absorption can be a two-photon process),

b) the compensation of the diffusion and/or photovoltaic currents by a drift current induced by electric field which is built up by redistribution of carriers (including carriers on trapping (donor) centers),

c) the change of the charge carrier density on donor or trapping centers, eventually their emptying or filling, during creation of the refractive index changes.

In the system of Eqs. (1)–(6) the illumination distribution $I(z)$ plays the role of an external generating function. However, the current density i which influences the carrier densities n_c and n_D contains a term proportional I (photovoltaic current) and a term dependent on the derivative of I with respect to z (diffusion current). The domination of one term over another can lead to different refractive index distributions. Thus, the spatial distribution of refractive index may be used for obtaining information about dominant mechanism of the internal current generation. When the illumination distribution I is expressed by cosine function both current terms are harmonic (cosinusoidal for photovoltaic current and sinusoidal for diffusion current) and it is difficult to distinguish which one is dominant in this case, mainly when diffraction investigation is used for studying the effect. That is why an aperiodic illumination can give additional information about the mechanisms of PRE.

In the next paragraphs, the experimentally obtained dependences and consequences following from the band transport model of PRE are discussed.

3. Experimental set-up

The samples used for the experimental investigation were congruent $LiNbO_3$ crystals with no additional post-growth treatment. The crystals were of different provenience (Crytur, Czech Republic; Photox Ltd., Great Britain, for example), different thicknesses (from 1 to 20 mm) and different concentrations of impurities. Investigation of symmetry of PRE, *i.e.*, the symmetry of spatial distribution of light-induced refractive index change, has shown that the character of the effect depends on the crystallographic orientation of the sample and does not depend on polarization of the beam used for inducing the refractive index changes (the writing beam) [12]. It also confirmed that the amplitude of the refractive index modulation is the greatest when the gradient of the illumination produced by writing beams is parallel with z-axis of the crystal and the reading beam is of extraordinary polarization. That is why results presented in the contribution were obtained using conditions as mentioned above.

The experimental investigation of the PRE nonlinearities was performed using two approaches:

– the real-time monitoring of the diffraction of light incident upon grating during its recording into $LiNbO_3$:Fe by periodic (harmonic) optical field,

– interference imaging of the refractive index modulation induced by periodic and aperiodic ("a strip-like") illumination.

A scheme of the set-up used for diffraction investigation is shown in Fig. 1**a**. The set-up consists of Ar and He-Ne lasers, a beam splitter (with dividing ratio of approximately 1:1) and a detector. An Ar laser (beam diameter 1 mm, vertical polarization) operating at 488 nm was used for creation of the refractive index changes and a HeNe laser operating at 633 nm was used for reading the changes. The angle α between interfering Ar laser beams was usually lower than one degree. The Figure 1**b** shows the set-up used for creating the refractive index change by strip-like illumination. It consists of the Ar laser, optical expander (3×) and a slot (normally with the width of 0.5 mm) placed just in front of the sample. Such geometry of illumination distribution allows us to compare the observed results with the theory simplified for 1-D case. The set-up used for imaging the refractive index changes is based on the Mach−Zehnder interferometer and its scheme is shown in Fig. 1**c**.

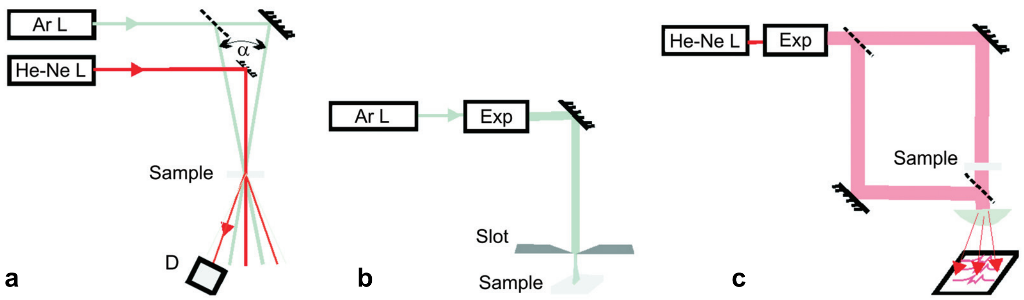

Fig. 1. The set-up for: diffraction investigation of PRE (**a**), recording by the strip-like illumination (**b**), interference imaging of refractive index modulation (**c**).

In the arrangement used for interference imaging the He-Ne laser beam is expanded (15×) and the back surface of the sample is projected on a screen. Here, the wave passing through the sample interferes with the reference wave coming from the other arm of the interferometer. The changes of the refractive index of the sample lead to the change of interference field. Consequently, knowing the sample thickness as well as wavelength of the light-source used the interference pattern allows us to determine the magnitude of the refractive index change in particular places of the sample. The phase difference between the object and the reference waves in a given place depends on the integral of the phase constant distribution (distribution of refractive index modulation) along the path of the optical beam in the sample. However, as the observed refractive index modulation is of the order of 10^{-3}, the deflection of the beam in samples with the thickness of about 1 mm is of the order of few micrometers. Thus, the distortion of the wave passing through the refractive index inhomogeneity which is of the order of 1 mm can be neglected.

The investigation performed can be divided into two groups: investigation of the linearity of the carrier generation and investigation of the linearity of the process of creation of the refractive index changes.

4. Linearity of carrier generation

With the goal of checking the linearity of carrier generation we measured the time dependence of the diffraction efficiency for the first diffraction maximum of grating created by interference of two coherent Gaussian beams (writing beams). The dependences were measured for intensities of the writing beams in the range from 0.2 $mWmm^{-2}$ to 8.7 $mWmm^{-2}$ in the $LiNbO_3$:Fe sample with 0.025 wt.% Fe, thickness 1.1 mm. The obtained dependences are plotted in Fig. 2**a**. The value of maximal diffraction efficiency as well as character of the measured dependences indicate that the grating behaves like a phase grating. This confirms the assumption that the illumination of the sample with intensity periodically dependent on the coordinate leads to modulation of the refractive index with periodic dependence on the coordinate. In Figure 2**b** there are the same dependences as in Fig. 2**a** shown but they are plotted as functions of the exposure (function of the product of intensity and duration of the applied illumination).

The range of intensities used is more than one order wide but the differences among measured dependences are small, comparable with the error of the measurement. This implies that within the range of intensities used the changes of refractive index are linearly dependent on the intensity of illumination.

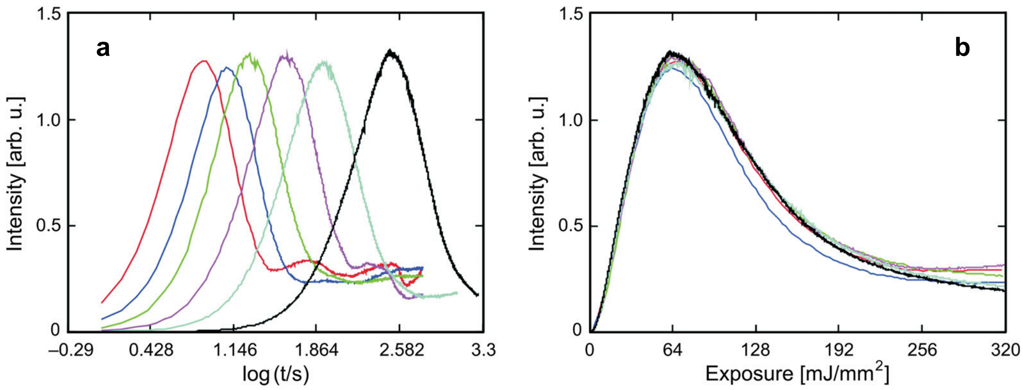

Fig. 2. The time dependences of the intensity of the first diffraction maximum for various intensities of recording beams (**a**). The same data plotted as a function of exposure (**b**).

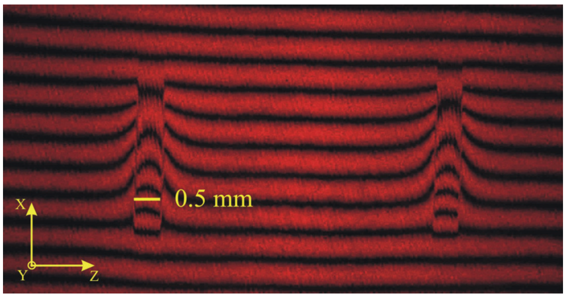

Fig. 3. Interferogram showing the refractive index distribution in a 1.1 mm thick sample of $LiNbO_3$: Fe (0.025%), after applying the strip-like illumination with different intensities, but the same exposure.

The interferogram in Fig. 3 shows the refractive index modulation after applying the strip-like illumination of intensity 7 mW/mm^2 for 6 minutes (closer to the left margin) and 21 mW/mm^2 for 2 minutes (closer to the right margin). It can be seen that the change of the refractive index in both cases is practically the same. This confirms the result obtained from measuring the time dependence of diffraction efficiency that the light absorption is (dominantly) a one-photon process. This is in agreement with assumption made in the model considered (Eq. (1)).

It can be seen from the figure that the change of the refractive index is practically the same in both cases. This confirms the result obtained from measuring the time dependence of diffraction efficiency that the light absorption is (dominantly) a one--photon process. This is in agreement with assumption made in the model (Eq. (1)).

5. Nonlinearity of the creation the refractive index changes

The nonlinearity of the process of changing the refractive index is reflected in the time dependence of the diffraction efficiency (Fig. 4).

During the experiment the diffraction efficiency was recorded with sampling rate 1 Hz, but in Fig. 4**a** there are values plotted which were measured every 190 seconds for red curve (sample 1 – pure $LiNbO_3$, thickness 8.4 mm), 15 seconds for blue curve (sample 2 – $LiNbO_3$ with no specified impurities, thickness 14.1 mm), and 10 seconds for green curve (sample 3 – $LiNbO_3$:0.05% Fe, thickness 1.1 mm). These intervals were chosen according to different sensitivities of the samples. Black curve in Fig. 4**b** represents the diffraction efficiency calculated for harmonic phase grating with fitted parameters.

Figures 4**a** and 4**b** show the same dependences but plotted as a function of the time of exposure and modulation of the wave phase $\partial\phi$ after its passing through the sample ($\partial\phi = 2\pi\partial n l/\lambda$), respectively.

The measured time dependences of the diffraction efficiency shown in Fig. 4**b** imply that:

– the amplitude of refractive index modulation is nonlinear during exposure and reaches a steady state with the maximal amplitude of refractive index modulation.

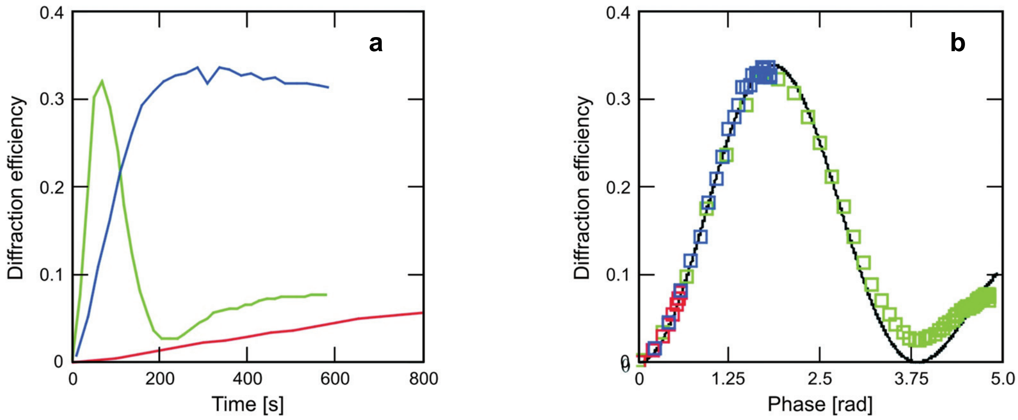

Fig. 4. Time dependence of the diffraction efficiency measured in various samples. Records were made in the same conditions (recording light intensity 11 mW/mm^2) – **a**. The same values as in (**a**) plotted as a function of modulation wave phase after its passing through the sample – **b**.

An exponential character of the process is documented by gradually increasing density of measured points,

– the maximal modulation of refractive index is different for each sample.

It means that the maximal refractive index modulation is not determined by the general parameters of single crystals, but depends on specific parameters of each sample. The smallest value was observed in the sample declared as pure $LiNbO_3$ and the highest in the sample possessing the highest concentration of dopants (Fe). These properties coincide with the idea that the limitation of the magnitude of the refractive index modulation is directly related to changes of the occupancy of the donor centers [14].

The existence of the steady state can also be demonstrated by investigating the refractive index modulation in the case of strip-like illumination. Figure 5 shows that the increase of refractive index modulation significantly slows down during illumination.

As stated in the first paragraph, the nonlinearity of the process of changing the refractive index can be either due to significant changes of occupancy of donor centers or due to the mutual compensation of the current involved by illumination and by the drift current generated by the electric space-charge field. However, the current induced by the sample illumination can be involved by the photovoltaic effect or due to the diffusion of the induced carriers. These mechanisms alone imply quite different results. If the photovoltaic effect is the main reason for free carrier redistribution, the electric field should be homogenous everywhere inside the illuminated strip and zero outside it [11]. However, our experiment does not confirm it. Moreover, the interferograms presented in Figs. 3 and 5 show that the refractive index modulation changes its sign at the boundary of the illuminated strip. Such a behavior in 1-D case cannot be explained assuming the photovoltaic current to be the main driving force

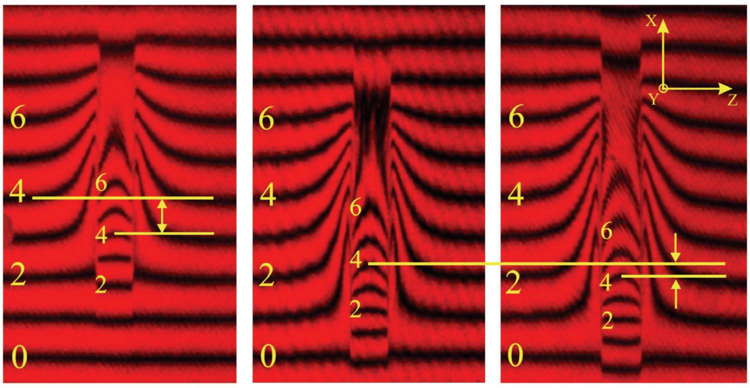

Fig. 5. Interferograms illustrating the slowing down of the increase of refractive index modulation for strip-like illumination in $LiNbO_3$:Fe:Mn with 0.075% Fe, 0.01% Mn. The width of the strip is 0.5 mm.

for the carriers. That leads us to the assumption that the diffusion current dominates in the carrier redistribution in the crystals used for the investigation. The validity of the assumption is justified also by the observed independence of the saturated value of the refractive index modulation on the illumination intensity (Figs. 2 and 3). If the photovoltaic effect played an important role in the process of the carrier redistribution in the samples the maximal value of the refractive index modulation would depend on the illumination intensity as the space charge voltage depends on the intensity of the sample illumination [15, 16].

6. Refractive index modulation in a steady state

The reason for the nonlinearities of PRE can also be deduced from the investigation of the refractive index distribution in the steady state reached after a long persisting illumination. The advantage of such investigation follows from the possibility of expressing the spatial distribution of the refractive index when the steady state is either the result of emptying the donor centers, or it is the result of the compensation of the diffusion current by the drift current.

6.1. Compensation of the diffusion current by the drift current

When neglecting the influence of the photovoltaic current the increasing of the amplitude of the refractive index change stops either due to mutual compensation of diffusion and drift currents (Eq. (3)) or due to emptying the donor centers (Eq. (1)). In the former case, using some simplifying assumptions it follows from Eqs. (1)–(3) that for an illumination with harmonic spatial distribution of intensity, expressed as

$$I(z) = I_0 + I_1 \cos(\Omega z), \quad I_0 > I_1 \tag{7}$$

the intensity of electric field after reaching the steady state is expressed by the relation

$$E(z) = \frac{D}{\mu}\frac{\Omega I_1 \sin(\Omega z)}{I_0 + I_1 \cos(\Omega z)} \tag{8}$$

Using the Einstein relationship between D and μ

$$D = \frac{\mu k_B T}{e}$$

in Eq. (8) we get for the spatial dependence of the intensity of electric field

$$E(z) = \frac{k_B T}{e}\frac{\Omega I_1 \sin(\Omega z)}{I_0 + I_1 \cos(\Omega z)} \tag{9}$$

which corresponds to the relation given in [7]. In Eq. (9), k_B is the Boltzmann constant and T temperature in Kelvin scale.

The dependences following from Eq. (9) are, for various ratios I_0/I_1 and in $k_B T\Omega/e$ units, shown in Fig. 6**a**. For practically accessible modulation of the spatial distribution of the intensity of illumination the maximal value of amplitude of electric field, according to Eq. (9), is of the order of $k_B T\Omega/e$, which is about 10^3 V/m (for spatial frequencies Ω being of the order of 10^5 m^{-1} which were used). Since Eq. (9) is derived from the band transport model, the dependences shown in Fig. 6**a** should also represent the character of refractive index modulation.

Figure 6**b** shows the interferogram of the sample after long-time (600 s) illumination by light with harmonic spatial distribution of intensity. As follows from Eq. (9) the spatial character of the refractive index is not tangent-like. The dependence of refractive index has the shape of function $1/(A + \cos(\Omega z))$ which is plotted in Fig. 6**c** for A equal 1.05, 1.2 and 2. In Figure 6**b**, there is the above mentioned function with $A = 1.8$ compared to observed spatial dependence of the refractive index. The comparison implies that the function fits much better the experimentally obtained dependences than the tangent-like function does.

The amplitude of phase modulation which can be red off from interferograms presented in Fig. 6**b** and Fig. 5 is more than 4π. Since the thickness of the samples was about 1 mm and the wavelength of the reading beam was 633 nm the maximal refractive index modulation in these samples is more than 10^{-3}. According to the model considered, the relationship between extraordinary refractive index and electric field E for the applied orientation of gradient of illumination (parallel with optic axis) is determined by electrooptic coefficient r_{33}, the value of which is $31.8 \cdot 10^{-12}$ m/V for $LiNbO_3$ [17]. This means that the electric field strength giving such a modulation of the refractive index should be of the order of 10^7 V/m. This value is in disagreement with the value of 10^3 V/m resulting from Eq. (9).

The disagreement between values of the observed change of refractive index and the change calculated according to the model as well as the difference between

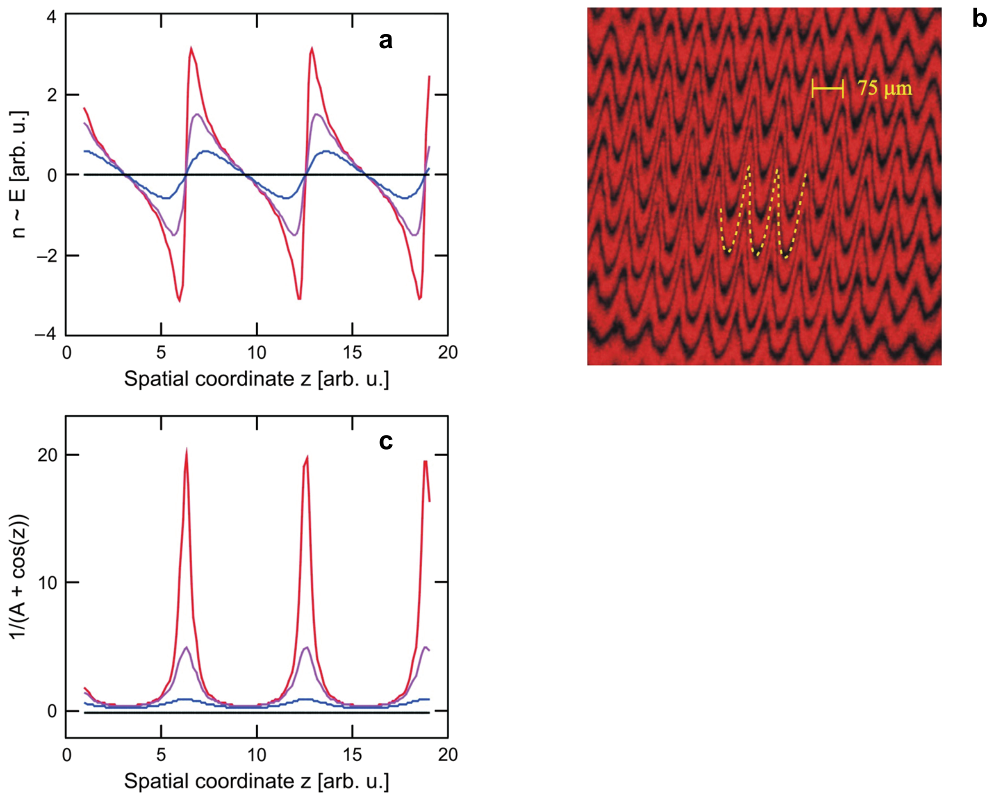

Fig. 6. A steady-state refractive index distribution in the case of diffusion, and the drift currents' mutual compensation for I_0/I_1 equal 1.05, 1.2 and 2 (**a**). Interferogram showing the refractive index modulation after long persisting harmonic illumination of a 1 mm thick sample (**b**). Function $1/(A + \cos(\Omega z))$ plotted for A equal 1.05, 1.2, 2 (**c**).

the shape of functions describing the electric field and the observed spatial distribution of the refractive index modulation leads to the proposition that the refractive index modulation is not due to the electrooptic effect.

6.2. Change of donor centers occupation

The steady state of the refractive index can be reached also by significant change of the density of electrons on donors. The steady state occurs when the density of electrons in conduction band n_{c0} is constant along the whole sample. For this state we get from Eq. (1)

$$n_D(z) = r\, n_{c0}\, N_D \frac{1}{\dfrac{gI_0 + rn_{c0}}{gI_1} + \cos(\Omega z)} \tag{10}$$

Such a distribution of the donor occupancy is, surprisingly, in agreement with the spatial dependence of the refractive index modulation shown in Fig. 6**c** if in expression $1/(A + \cos(\Omega z))$ the value A were equal $(gI_0 + rn_{c0})/gI_1$.

It follows from Fig. 5 that the characteristic features of the refractive index modulation in the case of strip-like illumination are the change of refractive index from positive to negative value at the edge of the illuminated region and the discontinuity of spatial distribution of the refractive index modulation at the edge. These features were observed in all investigated crystals independently of concentration of impurities and crystals' provider. The discontinuity of spatial distribution of the refractive index modulation makes us assume that the changes of the refractive index are not caused directly by internal electric field coming from redistribution of the carriers. It immediately follows from the fact that, according to basic equations of the theory of electromagnetic field, the electric field strength E should be continual function of the coordinate.

Comparison of the observation with the dependences resulting from the band transport model implies that the observed refractive index distribution cannot be explained by models of photorefractivity based on electrooptic effect. Thus the question about the mechanism lying behind the observed refractive index distribution should be answered. Even if providing the answer to this question is beyond the scope of the paper the results presented here give some hints that the diffusion of the charged carriers should be the main driving force leading to the observed refractive index distribution. However, the simplified band transport model uses just electrons as mobile carriers. In fact, there is an evidence that also different ions are mobile in a wide range of temperatures in $LiNbO_3$ [18, 19]. Potentially, the redistribution of these ions can give the refractive index modulation similarly as it is in the case of the planar waveguides prepared by in-diffusion of proper ions [20]. The laser-induced local-ion out-diffusion or interdiffusion in $LiNbO_3$ was suggested by Mailis *et al.* [3] as mechanism responsible for the observed changes of the refractive index when writing a waveguide in $LiNbO_3$. To be able to say whether the above mentioned mechanism takes part, further investigation of the problem is necessary.

7. Conclusions

The results of the experimental investigation performed imply that a general conceptual model of photorefractivity of $LiNbO_3$ should be modified. The investigation showed that the shape of the observed spatial distribution of the light-induced refractive index changes caused by the 1-D strip-like illumination cannot be explained by the assumption that the photovoltaic current is the dominant mechanism responsible for the carrier redistribution. Neither the observed values of the refractive index modulation in $LiNbO_3$ after long exposures, nor the shape of spatial dependences of the refractive index modulation found by the interference imaging of the illuminated samples can be explained by the refractive index modulation caused by the electrooptic effect.

The simple analysis of experimental results gives some hints as to the possible mechanisms which a new model should take into account. The investigation performed has shown that the shape of the refractive index modulation is similar to the spatial distribution of occupancy of donor centers. The result holds for both the periodic and aperiodic spatial distributions of illumination.

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