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Influence of surroundings on photorefractive effect in lithium niobate crystals

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ABSTRACT

In the paper results of the investigation of the influence of electric properties of the environment surrounding $LiNbO_3$ crystals on photorefractive effect induced in these crystals by Gaussian Ar^+ laser beam with various intensities are presented. We show spatial and temporal dependences of changes of the refractive index obtained experimentally in $LiNbO_3$: Fe and $LiNbO_3$: Fe:Mn samples surrounded by media with different electric conductivities and different permittivities (water, air, water solution of $CaCl_2$). The space and time dependences of the refractive index changes induced by the Ar^+ laser beam are observed by means of the Mach–Zehnder interferometer using light from HeNe laser. The experimentally obtained results are in a good agreement with those following from numerical calculations using the manifold mirroring method. The agreement between calculated and experimental results indicates that the polarization charge at the photorefractive crystal/surrounding medium boundary significantly influences the photorefractive process in the crystal. The experimentally observed slow spontaneous decrease of the refractive index change in a sample placed into a slightly conducting medium (air) after switching off the beam also indicates that the polarization charge in the sample's surroundings affects the photorefraction.

1. Introduction

The mechanism of the photorefractive effect (PRE) in ferroelectric crystals lies in spatial redistribution of charge carriers excited into conduction band due to the exposure of the material to light (internal photoelectric effect) and the subsequent change of the refractive index of the medium invoked by the electric field originated due to the charge redistribution (electro-optic effect) [1]. Throughout decades many researchers have contributed to the knowledge about the photorefractive effect in $LiNbO_3$ (LN) and other crystals by investigating the influence of doping [2,3], stoichiometry [4–8], intensity of light [9,10] and other kinds of radiation [11–13], or the temperature of the environment [14–16] on it. The effect of an applied magnetic field [17,18] and ac/dc electric fields [19–21] has also been a matter of a number of studies. Despite the variety of conditions under which the experiments were usually performed we found only a few works dealing with the effect of the electrical properties of the environment which surrounds the crystal on the optically induced refractive index change within the sample [22–25]. The main focus of these works was to present the observed rapid spontaneous decrease of the photorefraction (quasi breakdown) depending on the intensity of illumination and the environment, or both, rather than to provide a more general study of the origin of the relationship between the electrical properties of the medium surrounding the crystal and the photorefraction.

The present article deals with the influence of the electrical properties (σ and ε) of the surroundings of the LN crystals on the refractive index change induced due to their exposure to light. If a sample of a photorefractive material has finite dimensions and it is exposed to light, in addition to the space charge arising in the volume of the material also a polarization charge on the sample surfaces originates due to different electrical properties of the sample surroundings that influences the electric field inside the sample.

The charges produced in this way may under certain circumstances significantly contribute to the refractive index change inside the sample of the photorefractive material. Since the magnitude and the distribution of the charge at the 'investigated material/ambient surroundings' interface depend on the properties of these media, also the refractive index change will depend on the properties of the photorefractive material surroundings.

2. Calculation of electric field distribution

The electric charge density created in a photoconductive medium due to its exposure to light can be expressed in terms

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of electric current density by the equation:

$$\frac{\partial \rho}{\partial t} = \mathrm{div}(\vec{i}\,), \tag{1}$$

where $\vec{i}$ is the electric current density invoked by electric field, carriers diffusion or photogalvanic effect [26,27] that are due to the illumination of the material. In a one-dimensional case (applicable when dimensions of the sample are big enough and the intensity of illumination is a function of only one coordinate) the determination of the distribution of the electric field intensity (and thus also the refractive index modulation) is simple. This holds especially for short time just after commencement of illumination when the electric field intensity is so small that it affects the electric current density only negligibly. Then the final electric field is proportional to the current density distribution. In a multidimensional case (e.g. the dimensions of the sample are comparable with the dimensions of the area in which the charge has been induced by light) the calculation of the electric field intensity must take into account also the polarization charge that arises at the boundary between the photorefractive material and the ambient surroundings. In case the electric charge of density $\rho(\xi,\eta,\zeta)$ (ξ,η,ζ are the space coordinates of a particular point) is created in a plane parallel plate placed in a homogeneous medium, the distribution of the electric field induced by the polarization charge can be determined using the so called manifold mirroring method. This method is based on substituting the field produced by charges induced at the boundary between the two involved media by the field of virtual electric charges ${}^{n}\rho$ 'created' by mirroring on planes confining the sample [28] as shown in Fig. 1. The distribution of the virtual charge created by nth mirroring on a boundary with positive ($n>0$) or negative ($n<0$) coordinate is expressed by the formula:

$${}^{n}\rho(\xi,\eta,\zeta) = \varepsilon^{|n|}\rho((-1)^{n}\xi,(\eta-2na),\zeta). \tag{2}$$

Thus the total field is given by the sum of the fields produced by the charges ${}^{n}\rho$ (${}^{0}\rho$ is equal $\rho(\xi,\eta,\zeta)$) and $\varepsilon=-1$ when there is a conductive surrounding medium on both sides of the photorefractive medium. The parameter a in Eq. (2) denotes the half width of the sample. The method of manifold mirroring can be used also in case when the sample is sandwiched between two dielectric materials. In such a case, the parameter ε in Eq. (2) is given by the equation [29]:

$$\varepsilon = \frac{\varepsilon_1-\varepsilon_2}{\varepsilon_1+\varepsilon_2} \tag{3}$$

where ε_1 and ε_2 are the dielectric constants of the sample material and the ambient surroundings, respectively.

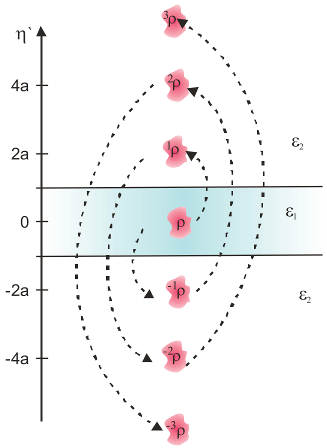

Fig. 1. Virtual electric charges ${}^{n}\rho$ created by mirroring on planes confining the sample.

The space electric charge in a photorefractive medium is due to electric current of density:

$$\vec{i}\,(\xi,\eta,\zeta) = \beta I\vec{k} + eD\,\mathrm{grad}(n_c) + \vec{E}\sigma, \tag{4}$$

where β is a particular element of the photogalvanic tensor expressing the density of the photogalvanic current in the investigated z axis (optical axis) direction for used polarization of light [30], I is the light intensity absorbed at a particular point in the sample, $\vec{k}$ is the unit vector in the z axis direction, e is the elementary charge, D is the diffusion constant, n_c is the density of the carriers in the conduction band, σ is the conductivity of the medium, and $\vec{E}$ is the electric field intensity. If it is assumed that there is no external electric field applied to the photorefractive material, then the electric field originated due to the inhomogeneity of the current inside the material is (for short time after commencement of the sample exposure to light) proportional to time. This means that for sufficiently short times of illumination the current density expressed by $\vec{E}\sigma$ is negligible compared to the photogalvanic and/or diffusion currents and the electric field intensity, as well as the amplitude of the refractive index change, will grow linearly with time.

In accordance with literature [31–33] we assumed that the diffusion current in used samples was negligibly small when compared to the photogalvanic current. Thus when ρ grows linearly with time, we can write:

$$\rho(t) = -\mathrm{div}(\beta\vec{k}I(\xi,\eta,\zeta))t. \tag{5}$$

On the other hand, if we assume the absorption of light is so small that the loss of intensity in the sample does not have to be taken into account and the distribution of the electric field in the sample is induced by a Gaussian beam, then using Eq. (2) we get for the z component of the electric field intensity:

$$E_z(x,y,z) = \sum_{-n}^{n}\int\int\int \frac{\rho(\xi,\eta,\zeta)}{4\pi\varepsilon_{33}} \times \frac{(-1)^{n}(\zeta-z)}{((\xi-x)^2+((-1)^{n}\cdot(\eta-2na)-y)^2+(\zeta-z)^2)^{3/2}}\,d\xi d\eta d\zeta, \tag{6}$$

where ε_{33} is the permittivity of the sample material corresponding to the direction of the optical axis of the crystal.

Since the contributions of ${}^{n}\rho$ to the field with respect to n decreases faster than $1/(2na)^2$, so that a fast convergence of the series (6) can be expected. By performing numerical calculations of the distribution of the electric field intensity we proved that the series converges uniformly and fast enough and that for $n=10$ the difference between each two subsequent terms is negligibly small. We also assume that diffusion of the carriers in $\mathrm{LiNbO_3}$ is negligibly small. It means that the photogalvanic current gives a dominant contribution to the distribution of the charge density.[1] The photogalvanic current in $\mathrm{LiNbO_3}$ is dominant in z axis direction [34] for considered gradient of optical field. In this case the electric charge distribution depends on the derivative with respect to the z coordinate even if the intensity of illumination is not a function of the z coordinate only.

The spatial dependences of the electric field calculated according to the method of multiple mirroring using Eqs. (5) and (6) for a 1 mm thick $\mathrm{LiNbO_3}$ sample illuminated by (elliptical) Gaussian laser beam with radii $R_z=0.325$ mm and $R_x=2.9$ mm are depicted

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[1] It is assumed that during illumination of the sample the free carriers are captured by traps or donor centers. Consequently, the gradient of the free carriers' density is lower than the gradient of the total charge density. Therefore, the assumption that the diffusion current is negligible compared to the photogalvanic current may be correct also for relatively large refractive index change.