Investigation of spatially nonuniform nonlinear response of a lithium niobate crystal sample at low light intensity

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Abstract. The spatial distribution of nonlinear optical response over a bulk of lithium niobate sample is experimentally studied through the distortions of the two-dimensional light beam intensity patterns at the sample output surface caused by the beam spatial self-action. The compensation of these distortions and the linear light beam divergence by means of the pyroelectric effect contribution into the nonlinear optical response of the crystal are also studied. The results obtained for the light wavelength of 532 nm and beam waist diameter of 13 µm demonstrate the partial or total compensation of the beam divergence depending on light power and a temperature increase at the sample heating.

1. Introduction
The homogeneity of physical properties of dielectric and semiconductor materials is a critical requirement for production of large aperture optical and acousto-optical components, electro-optical modulators and switches, wavelength converters and many other optical elements [1 - 4]. Light beam propagation in nonlinear optical medium may result in the transformation of its spatial and spectral structure due to the self-action effects. In turn, the efficiency and magnitude of distortions appearing during light propagation in the medium depend on the local concentrations of impurity atoms, on the presence of defects and on small variations of the crystal structure itself. In some known electrooptic crystals like lithium niobate (LiNbO₃), lithium tantalate (LiTaO₃) and strontium – barium niobate (SrₓBa₁₋ₓNb₂O₆) the spatial self-action of light is caused by the photorefractive effect and may be observed at the light powers of a microwatt range that is very useful for the experimental investigations [5 - 7]. The high optical nonlinearity results in the slow speed of the medium nonlinear response that also brings the advantages to the distortion accumulation study because it requires using simple and not too expensive equipment.

The most interesting among the spatial conversion effects of optical beams is the light localization in the form of spatial solitons or light field parts which demonstrate diffraction-free propagation in nonlinear optical media. The spatial soliton effects make possible design of all-optical photonic elements and circuits with light controlled by light. Another result of similar effects relates to generation inside the nonlinear medium optical waveguides and regular diffractive elements which also can be used for the light control needs. It should be noted that the boundaries of the nonlinear medium or interfaces between various materials may sufficiently vary the self-action characteristics of light fields. For example, generation of regular light patterns due to the self-action effect becomes possible in nonlinear optical cavities where the optical feedback comes into play. Similar effects have
been observed in photorefractive crystals and other nonlinear media with both, self-focusing and self-defocusing nonlinear response [8 - 10].

In this work, we study the spatial distributions of nonlinear optical properties within LiNbO$_3$ bulk sample testing the distortions of light beam profiles in conditions of normal room temperature, the photorefractive effect influence and at the contribution of thermo-optic effect at different elevated temperatures and light wavelength of 532 nm.

2. Experimental setup and conditions

The schematic of the experimental setup used to study the uniformity of linear and nonlinear properties of the LiNbO$_3$ sample over its cross-section is shown in Figure 1. A continuous-wave YAG:Nd$^{3+}$ laser (1) with radiation wavelength of 532 nm and output power up to 50 mW is used as the light source in our experiments. The light polarization corresponds to the extraordinary wave of a crystal. The light beam is focused onto the sample input surface by a lens (2) with focal length of 40 mm. It provides the diameter of light spot at the input surface about 13 µm for the half-maximum intensity level. The dimensions of LiNbO$_3$ sample (3) are 4×10×4 mm$^3$ along X, Y and Z axes. The sample is allocated on the Peltier element (4) which provides the crystal heating up to 100ºC. Its temperature is measured with a thermocouple or a non-contact infrared thermometer. Light beam propagates within the crystal along its Y-axis running the distance ~10 mm. The spherical lens (5) images light pictures at the input or output surfaces of the crystal sample onto the sensor plane of a laser beam analyzer (6). The corresponding light fields at these surfaces are studied visually and their images are saved and analyzed with a computer.

![Figure 1. Schematic of the experimental setup: 1 – laser; 2, 5 – lenses; 3 – LiNbO$_3$ crystal; 4 – Peltier heater; 6 – laser beam analyzer.](image-url)

We test in experiments distortions of focused light beam probing different regions of a crystal bulk. The schematic of the incident beam positions (white circles) over the sample entrance surface is shown in Figure 2. To precisely move the sample with respect to the light beam, the three-coordinate positioning stage is used. Light beam freely diffracts while propagation through the sample in the linear regime, e.g. at very low light intensity and short time periods after light switching on. At the first set of experiments we study the light beam patterns at the exit surface of the sample that informs on the uniformity of crystal sample optical properties together with a quality of optical polishing of its input and output surfaces. The spatial inhomogeneity of nonlinear optical properties of the sample is studied due to comparison of light beam images at its output surface in the initial moment after light switching on and after development of beam distortions because of the spatial self-action effects. The different mechanisms of nonlinear optical response may contribute to the beam self-action in LiNbO$_3$. These are the photorefractive effect, the light absorption and the thermo-optic effect which includes the pyroelectric effect and the sample thermal expansion. The thermo-optic contribution may vary by the sample heating and for LiNbO$_3$ such a heating can compensate the photorefractive beam distortions [11, 12]. We use the sample heating procedure to check the possible compensation of light beam divergence as at low as at higher light intensity.
3. Experimental results and discussions

The results of the linear diffraction of light beam at its propagation within the crystal sample, the distortions of the beam cross-section at the sample output surface due to the nonlinear diffraction caused by the photorefractive effect, and the total compensation of linear and nonlinear diffraction of the probing light beam are illustrated by Figure 3. The light power is 0.2 mW in this experiment. The first column shows the light pattern at the input surface of the sample and the intensity profile of this field along direction parallel to the crystal optical axis (Z axis). The linear diffraction broadens this beam about four times on the sample length and corresponding light pattern and intensity profile are shown in the second column. In some time the diffraction divergence of this beam becomes larger due to the uncontrolled photorefraction of the crystal. The crystal sample is nominally un-doped with special photorefractive impurities like iron or copper [5 - 7]. However, the photorefraction within it is notable due to the existence of intrinsic defect states and some dose of uncontrolled impurities. Thus, the space-charge electric field, directed primarily along the crystal polar axis, develops within the sample because of the light influence. The photorefractive nonlinearity of LiNbO₃ is of self-defocusing kind and this electric field induces a negative nonlinear lens within the illuminated area due to the linear electrooptic effect. It results in the anisotropic distortions of intensity distribution at the output surface of the sample with strong broadening of light pattern along the optical axis (Figure 3, column 3). This is a nonlinear diffraction broadening of a light beam.

Both, the linear and nonlinear diffraction may be suppressed in experiments by the crystal heating using Peltier element. Light pattern and its intensity profile (Figure 3, column 4) confirm the total compensation of the beam diffraction which is obtained in this experiment at the crystal temperature ~60°C. The effect of the beam diffraction compensation is generally caused by the pyroelectric field $E_{pyro}$ arising within a ferroelectric crystal bulk due to its uniform heating accompanied with the spontaneous polarization $P_s$ change [12, 13]:

![Figure 2. Schematic of probing beam locations over LiNbO₃ sample input surface.](image)

![Figure 3. Light patterns (top row) and intensity profiles (bottom row) at the sample input surface (1), output surface in the linear regime (2), output surface in 10 minutes after light switching on (3) and at output surface and sample heating to 60°C (4).](image)
\[ E_{\text{pyro}} = -\frac{1}{\varepsilon_0 \varepsilon_r} \frac{dP_S}{dT} \Delta T, \]

where \( \varepsilon_0 \) and \( \varepsilon_r \) are the dielectric constants of vacuum and the crystal, and \( \Delta T \) is a temperature change. This field decreases the refractive indices of the whole crystal due to the linear electro-optic effect with the decreasing value \( \Delta n = 0.5n' r_{\text{eff}} E_{\text{pyro}} \). Here \( n \) is average refractive index, and \( r_{\text{eff}} \) is effective electro-optic constant. However, the pyroelectric field \( E_{\text{pyro}} \) becomes screened in the illuminated region of the crystal due to the redistribution of charge carriers existing within illuminated bulk because of the photoexcitation. Thus, the interplay of the photorefractive and the pyroelectric effects may result at some conditions in the formation of two-dimensional bright spatial soliton in the crystal bulk. The inhomogeneity of light beam distortions is experimentally studied in further our experiments in conditions of low and higher light intensity that can change the results of interplay of photorefractive and pyroelectric effects.

3.1. Light beam distortions at low optical intensity

As the result of LiNbO\(_3\) sample testing using its probing with a low intensity laser beam, we observe a map of light images at the output surface of the crystal sample which points to inhomogeneous distribution of internal optical properties over the crystal bulk (Figure 4). The optical power is 0.1 mW and the exposure time is ten minutes in this case. We probe the input surface in nine points with distances of 1.5 mm between them in directions parallel to X- and Z- axis (see Figure 2). These images correspond to photorefractive-distorted initially radial symmetric light patterns with dimensions observed at the light switching on moment (see Figure 3, column 2). Their comparison demonstrates slightly different nonlinear diffraction broadening of initial images corresponding to the linear diffraction of probe beam in different points of the input surface. It points to inhomogeneity of photorefractive properties over the sample bulk. Two main subjects may reason that. First, it is the inhomogeneous distribution of impurities and defects responsible for the crystal photorefractive nonlinearity. And the second subject is possible drift of some ions within the sample bulk along direction of crystal polar axis caused by some cycles of sample heating, cooling and illumination in different previous experiments [15].

![Figure 4](image_url)

Figure 4. Light beam images at the output surface of the crystal sample in 10 minutes after light «switching on» at light power 0.1 mW.
The sample heating to the temperature 35°C results in the partial compensation of the light beam diffraction. It is illustrated by intensity profiles of light patterns at the output surface (Figure 5) after 10 minutes exposure at room temperature (PR) and after sample heating to 35°C (H). It should be noted that we observe only compensation of nonlinear diffraction of light beam in these conditions because the pyroelectric field value is not enough to compensate both, linear and nonlinear diffraction [13].

![Intensity profiles](image)

**Figure 5.** Light intensity profiles at the crystal output surface for optical power of 0.1 mW: profiles PR correspond to beam broadening due to the photorefractive effect; profiles H show partial compensation of light broadening in conditions of crystal heating to 35°C.

### 3.2. Light beam distortions at high optical intensity

We also use the same approach to test the influence of the thermo-optic effect on the light beam diffraction in conditions of the high optical intensity. Light power is 5.5 mW in this case and the crystal sample is heated to 60°C temperature. As in the case of low light intensity and sample temperature of 35°C, the total compensation of light beam divergence is not obtained at these conditions. The experimental results for the point (7) of laser beam location at the sample input surface (see Figure 2) are shown in Figure 6. The only nonlinear contribution to the beam divergence is compensated here at the crystal sample heating. Obviously, the pyroelectric field value is not
enough in this case as well to compensate as linear as nonlinear diffraction of light beam [13, 14]. Similar results are observed for other locations of probing optical beam at the input surface.

![Figure 6](image1)

**Figure 6.** Light fields: at the input sample surface (a); at its output surface (b); at the output surface with photorefractive broadening (c) and after partial compensation of nonlinear diffraction broadening at crystal sample heating to 60°C (d).

### 3.3. Discussions

The comparison of light beam intensity profiles at the output sample surface in conditions of uncontrolled photorefractive effect and in regime of its compensation by the pyroelectric effect points to the possible improvement of light field distributions due to the crystal heating. Indeed, beam intensity profiles with photorefractive induced distortions demonstrate noisy envelopes even at near to perfect optical polishing of the sample surfaces. Ten degrees temperature increase transforms these noisy envelopes to smooth Gaussian-like shapes. Experiments demonstrate that the total compensation of narrow optical beam divergence including their linear and nonlinear parts requires taking into account the crystal quality, composition and content of different impurities.

### Conclusion

The inhomogeneity of photorefractive optical nonlinearity can sufficiently distort the light beam profiles even in linear regime within lithium niobate samples used for operation in the green range of visible, especially at requirements of large apertures of crystal elements. These distortions can be suppressed at the controlled elevation of crystal sample temperature for some degrees. It is important that contribution of pyroelectric effect may result in the diffraction free propagation of light beams with diameters of micrometer range value in this crystal that may be used in creation of all-optical elements with light controlled by light.

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