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OPTICAL AND HOLOGRAPHIC PROPERTIES OF A PHOTOREFRACTIVE CRYSTAL OF LITHIUM NIOBATE DOPED WITH IRON IONS

Abstract. The work is devoted to the study of the optical and holographic properties of an iron-doped lithium niobate crystal ($LiNbO_3$: Fe). The research results show that with an increase in the iron impurity concentration, the maximum diffraction efficiency is achieved at a lower exposure. The change in the refractive index of the crystal increased from 10^{-5} to $5\cdot10^{-5}$.

Keywords: Diffraction efficiency, refractive index, photosensitivity, photorefractive crystal.

Introduction

In holography, to form an interference pattern, two coherent beams are required, one of which is called the reference beam, and the other is called the object beam scattered from the object (extraordinary). The resulting interference pattern contains information about the amplitude and phase of the object beam. The intensity of an interference pattern can be recorded by placing an appropriate photosensitive material (such as photographic film or a photorefractive crystal) in the interference region. This recorded stripe pattern or grating is called a hologram. The recorded hologram, when illuminated by the same reference beam, can scatter light in the direction of the object beam. The diffracted beam contains information about the phase and amplitude of the original object beam.

The creation of optical information processing systems and the development of holography with its numerous applications set the task of searching for and developing materials that change their optical properties when exposed to laser radiation. The parameters of optical information processing systems are mainly determined by the characteristics of the recording media. For example, systems for online optical information processing require reversible media, unlike photographic emulsions, which have high sensitivity (10^{-5} j/cm^2) and resolution, but cannot be used multiple times. Recording materials for optical information processing systems should allow high-density recording of information, non-destructive reading, easy rewriting of information (10^7–-10^8 cycles) with sufficient diffraction efficiency to reproduce information [1].

Photorefractive crystals, such as $LiNbO_3$ doped with iron ions in various concentrations, occupy a special place among promising recording materials for creating holographic systems for optical information processing [2]. The recording of information in ferroelectric crystals is based on the effect of a local reversible change in the refractive index in these crystals when illuminated by a laser beam. The photoelectric properties of ferroelectrics are affected by spontaneous polarization, with a change in which, under the influence of light, an internal field appears, which contributes to the redistribution of carriers and the formation of a space charge. The space charge field due to the electro-optical effect causes a change in the refractive indices of the substance.

In a photorefractive single crystal, defects arise under the action of laser radiation in the illuminated region of the crystal, through which the laser beam passes, which is not the case outside the illuminated region of the crystal. These defects are fluctuating micro- and nanostructures with changed physical parameters (such as the refractive index, diffraction efficiency, photo- and electrical conductivity, etc.) [3; 4]. Increasing the sensitivity and speed of recording holographic information can be achieved by changing the composition of the crystal and the features of its structure. The most interesting part of it is the influence of the order of the units of the cationic sublattice along the polar axis on the properties of the photorefractive effect. Note that the order of units of the cationic sublattice determines the magnitude of spontaneous polarization in optically nonlinear crystals with an oxygen octahedral structure, as noted in [5].

Among the large number of photorefractive materials synthesized to date, promising as holographic materials, the oxygen-octahedral lithium niobate ($LiNbO_3$) single crystal stands out for its long-term memory due to high electro-optical and optically nonlinear coefficients [6; 7]. The photorefractive properties of $LiNbO_3$ can be controlled by changing both the stoichiometry (R = Li/Nb ratio) and doping [8; 9]. In this case, as noted above, the order of units of the cationic sublattice, the state of defects, and the magnitude of spontaneous polarization change significantly [9]. $LiNbO_3$ single crystals with a strong photorefractive effect can be obtained by doping with multiply charged transition metal cations. These cations (Fe, Cu, Mn, Ni, etc.) are called "photorefractive" and under the action of laser radiation change their charge in the crystal, improving the photorefractive effect.

The effect of photorefraction, the photo- and electrical conductivity of the $LiNbO_3$ crystal, depending on its composition and the state of defectiveness of the crystal lattice, vary over a very wide range. In this case, a change in the band gap should occur, which for a nominally pure crystal is 3.4 eV, which is close to the value characteristic of wide-gap semiconductors. By reducing the band gap, it is possible to bring the properties of the $LiNbO_3$ crystal closer to semiconductor properties, which makes it possible in principle to develop materials with cross-effects.

Experimental technique

Noting the above, we note that the interest in the studied mechanisms of holographic recording of information on lithium niobate crystals doped with iron ions is beyond doubt.

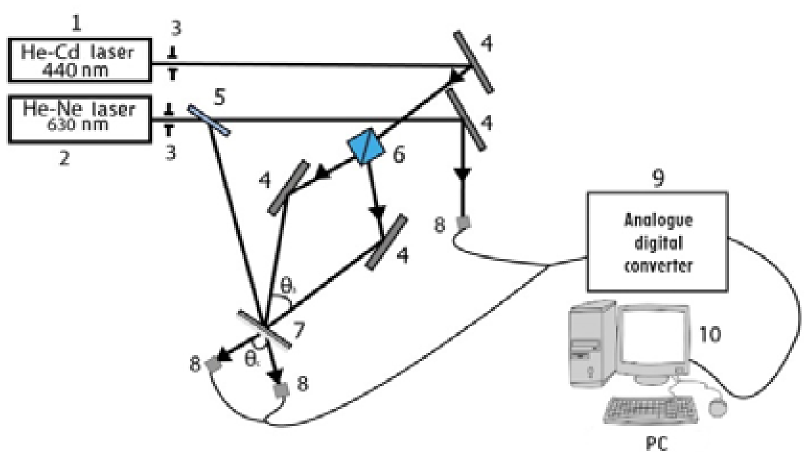

Figure 1. Scheme of the experimental setup: 1 – recording laser (He-Cd) λ = 440 nm, 2 –reading laser (He-Ne) λ = 630 nm, 3 – diaphragm, 4 – mirrors, 5 – filter, 6 – prism Vollaston, 7 – sample, 8 – photodetector, 9 – analog digital converter, 10 – computer

In experiments on the study of optical recording in $LiNbO_3$ crystals, the scheme shown in Figure. 1 was used.

The recording is made by a helium-cadmium (λ = = 440 nm) laser beam. At this wavelength, the crystals are highly sensitive to optical distortion. The reading is performed by a He-Ne laser with λ = = 630 nm, for which the sensitivity of the crystal is negligible, so the reading does not lead to erasure of the hologram. As a result of the superposition of two plane waves in a crystal, an interference pattern appears in the form of light and dark stripes. The diffraction efficiency – η of a sinusoidal grating, when read by an extraordinary beam with a given wavelength – λ, is given by the Kogelnik formula [10].

$$\eta = \sin^2 \left\{ \frac{\pi D \Delta n_e}{\left[\lambda \cos \frac{\theta}{2} \right]} \right\} \qquad (1)$$

where Δn_e is the modulation amplitude of the refractive index of the extraordinary ray, D is the crystal thickness.

Experimentally, diffraction efficiency is defined as the ratio of the intensity of the diffracted readout beam to the intensity of the beam that has passed through the crystal when the hologram is not recorded in the crystal. Our diffraction efficiency results were defined as the ratio of the diffracted beam to the intensity of the reference beam, i.e. without taking into account the reflection and scattering of light in the crystal.

Results and its discussion

The fundamental issue in the analysis of the mechanism of formation of optical damage Δn (changes in the refractive index) is the question of its dependence on the wavelength of the irradiating light λ. This dependence makes it possible to judge the exchange of electrons between excited and unexcited iron ions, with the transition of electrons to the conduction band.

On fig. Figure 2 shows experimental studies of the dependence of the influence of various concentrations of iron ions in $LiNbO_3$ on the diffraction efficiency of holograms – η, recorded (Fig. 1) by a helium-cadmium laser (λ = 440 nm) in the form of a plane wave front (sample 1–0.005 wt.% Fe, sample 2–0.020 wt% Fe).

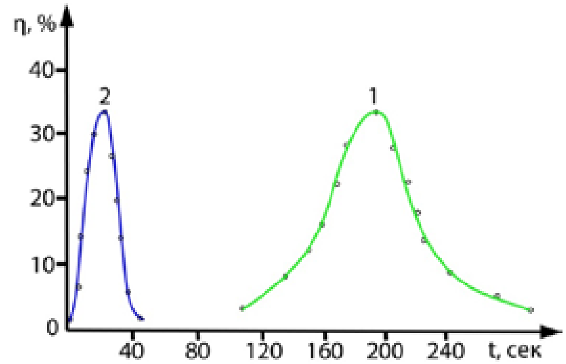

Figure 2. Experimental dependences of the diffraction efficiency of holograms on exposure at λ = 440 nm (sample 1– –0.005 wt.% Fe, sample 2–0.020 wt.% Fe)

The studied dependence of the diffraction efficiency on the concentration of introduced impurities showed that the obtained holograms recorded at a wavelength of λ = 440 nm differ significantly. As can be seen from Figure 2, the photosensitivity, and hence the diffraction efficiency, in sample 2 is 7 times greater than in sample 1. The maximum diffraction efficiency η = 34%, obtained at a wavelength of λ = = 440 nm, is achieved at various expositions.

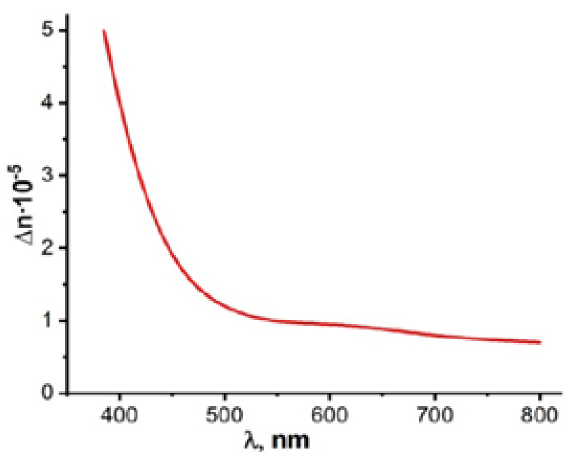

Figure 3. Dependence of the change in the refractive index of $LiNbO_3$: Fe crystals on the wavelength of the recording radiation

In our opinion, this is due to a significant increase in the change in the refractive index of $LiNbO_3$: Fe at $\lambda < 500$ nm, which is shown in Figure 3.

Using formula (1), one can find the change in the refractive index of the crystal:

$$\Delta n_e = \left[\frac{\lambda \cos \frac{\theta}{2}}{\pi D} \right] \text{arc} \sin \sqrt{\eta} \qquad (2)$$

where λ is the wavelength of the reading radiation; θ is the angle between the interfering beams. The values of Δn_e during the recording of information can vary from 10^{-5} to $5 \cdot 10^{-5}$.

Features of holographic recording in $LiNbO_3$: Fe are due to the anisotropy of the properties of the crystal and the specifics of the mechanisms of hologram formation. For practical applications of $LiNbO_3$: Fe crystals, it is important to know the mechanisms of change in the refractive index, which are studied by many researchers.

As is known [3], the use of transition metals as dopants is associated with their ability to reversibly donate d-electrons to the conduction band under the action of radiation. When a crystal is doped with Fe^{3+} ions, the absorption of light in it is caused by ionization. The light sensitivity of iron-doped crystals is determined by the concentration of Fe^{2+} ions, which have a broad absorption band in the lattice with a maximum at about 400 nm. Upon photoexcitation, Fe^{2+} donates a photoelectron to the conduction band, which is captured by the Fe^{3+} ion in the unilluminated region during diffusion. As the Fe^{2+} concentration increases, the absorption at the wavelength at which information is recorded increases, which leads to an increase in the sensitivity of the crystal to light.

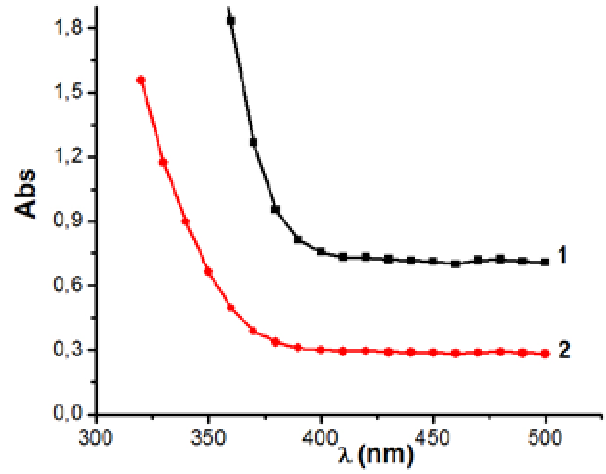

Figure 4. absorption spectra of nominally pure and iron-doped lithium niobate $LiNbO_3$: Fe

For comparison, in Figure 4 shows the absorption spectra of nominally pure and iron-doped lithium niobate $LiNbO_3$: Fe, from which it can be seen that the addition of an impurity significantly increases the absorption. Absorption begins to increase rapidly from 400 nm. Therefore, in the figure 4, the range from the short-wavelength range to 500 nm is taken.

Conclusion

An increase in the iron impurity concentration in lithium niobate leads to an increase in the diffraction efficiency. The maximum diffraction efficiency $\eta = 34\%$, obtained at a wavelength of $\lambda = 440$ nm, is achieved at different exposures, depending on the concentration of iron in the crystal, which is due to a significant increase in the change in the refractive index of $LiNbO_3$: Fe at $\lambda < 500$ nm. The values of the change in the refractive index, during the recording of information, can vary from 10^{-5} to $5 \cdot 10^{-5}$.

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