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Laser Conoscopic Research Technique For Single Crystals $LiNbO_3$: Mg

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ABSTRACT

Optical homogeneity and subtle features of structural distortions in a series of single crystals of lithium niobate ($LiNbO_3$) of congruent composition doped Mg^{2+} [0.01 - 5.5 mol·%] Investigated by laser conoscopic method using radiation He-Ne laser (λ = 632.8 nm) of less than 1 mW.

Keywords: Lithium Niobate; Conoscopic Patterns; Optical Homogeneity

1. Introduction

Initially, the conoscopic patterns obtained with a polarizing microscope were used in mineralogy in order to identify minerals based on the data on crystal symmetry and orientation [1]. Conoscopic pattern informativity provides for the possibility to determine orientation and nature of optical indicatrix, measure an angle between the optical axes of a biaxial crystal, determine an optical sign of the crystal, detect optical axes dispersion, identify qualitative and quantitative changes in the optical indicatrix in response to external action, etc. [1-11]. Conoscopic method is one way to analyze the properties of optical crystals, which allows to determine their functionality, and which has long and successfully used in scientific research and a variety of optical devices.

In this article, it is proposed to obtain conoscopic patterns using an optical system where diverging laser radiation is let pass through an anisotropic crystal placed between the polarizer and analyzer, rather than using a polarizing microscope. The pattern on the screen is recorded by a digital camera and displayed on a computer.

Where the point symmetry group of the crystal is already known, the practical importance of such conoscopic studies lies in detection and analysis of various distortions of optical elements of actual crystals [10,11]. Modern industrial technology for growing single crystals of lithium niobate doped with different dopants influencing the composition of the crystals and physical properties, allowing them to adjust to a wide range. One of the main criteria is the quality of produced crystals of optical homogeneity. Use as a dopant cations Mg^{2+} provides lower interfering effect photorefraction in lithium niobate single crystals, however, can complicate the structure is strong enough crystal and, as a consequence, lead to the optical inhomogeneity. The possibility of observing conoscopic patterns of large-scale appears when you use the laser system in which divergent wide-beam radiation is obtained through the diffuser placed in front of the front face of the crystal [5].

The significant size of the image allows you to perform a detailed analysis of subtle features of the structural distortions in the crystal, as in the center of the field of view, and in the peripheral region of the conoscopic patterns.

The development of laser conoscopic method also relevant to studies of thin structural distortions, arising in photorefractive crystals, for the detection and investigation of subtle features of structural distortions, as well as micro-and nanostructures, inevitably present in doped single-crystal materials [12].

In this paper, a laser conoscopic method investigated the fine features of structural distortions in a series of single crystals of lithium niobate ($LiNbO_3$) congruent (R = Li/Nb = 0.946), doped with Mg^{2+}, characterized by low effect photorefraction (optical damage), promising as materials for electronics [12,13]. Used as a relatively lightly doped crystal $LiNbO_3$:Mg[0.01 - 1.5 mol·%], and crystals with a high concentration of Mg^{2+} ($LiNbO_3$:Mg [3.0 - 5.5 mol·%]), photorefractive effect in which is almost equal to zero [13].

2. Experimental Technique

The test samples were cut from a single crystal boule grown in the direction of the Z (the polar axis of the crystal). In order to evaluate the optical homogeneity of single crystal boules grown samples were cut from different parts of the boule.

The cylindrical portion of the boule was cut into transverse disks from which the samples were cut into parallelepipeds ~ $8 \times 6 \times 4.7$ mm^3 edges parallel crystallophysical axes X, Y, Z, respectively. Faces of the parallelepiped and plates carefully polished. Methods of crystal growth and preparation of samples for research in more detail in [12].

When conducting an experiment to observe conoscopic patterns of optical crystals with optical system (**Figure 1**), consisting of a source of radiation, polarizer, diffuser, crystal, analyzer and the screen, which allows you to receive conoscopic pattern of considerable size (0.5 meters or more).

Investigated crystal plate is located on the two-coordinate optical mobile stand that allows you to scan the entire plane with a laser beam entrance face and get a series of conoscopic patterns. In the experiments, radiation He-Ne laser (λ = 632.8 nm) power not exceeding 1 mW in order to minimize the possible impact of the photorefractive effect on conoscopic pattern.

To investigate the defect micro and macrostructure single-crystal $LiNbO_3$:Mg was applied high-performance and flexible image analyzer Thixomet ®, based on modern hardware (microscope of *Carl Zeiss* - Axio Observer) and software [14].

3. Experimental Results and Discussion

Conoscopic picture perfect uniaxial crystals obtained with linearly polarized radiation is well known, explained and described in the literature [1,6,10,11].

This picture of the propagation of a diverging beam of light along the optical axis is composed of concentric rings centered at the output of the optical axis. Rings superimposed on the characteristic intensity distribution - black "maltese cross" In this case, each ring is the same line of the phase shift and the cone of rays with the same angle of incidence at the coincidence of the axis of the conical radiation beam with the optical axis of the crystal.

Izohrom form depends on the orientation of the optical axis with respect to the input face of the crystal. With some of the angle between the optical axis and the normal to the entrance face of the ring transformed into ellipses. With significant corners form izohrom approaching hyperbole.

Branch of the "maltese cross", consisting of two isogyre minimum intensity, intersect in the center of the visual field, perpendicular to each other and coincide with the axes of transmission of the polarizer and the analyzer.

The characteristic feature of arising anomalous optical biaxiality in which there is a deformation of the optical indicatrix of the crystal is the rupture of black "maltese cross" in two parts with the enlightenment in the center of the visual field.

In our experiments, for samples $LiNbO_3$:Mg[0.01 - 1.5 mol·%] were observed conoscopic pattern of the standard form, in which the black "maltese cross"preserves the integrity of the center of the field of view, and isochromes have the form of concentric circles.

For samples with the same thickness in the direction of the optical axis, but with a different concentration of dopant Mg, for example, $LiNbO_3$:Mg [0.5 mol·%] and the $LiNbO_3$:Mg [1.0 mol·%] general view of conoscopic patterns has coincided **Figure 2** with preservation of diameter ring-izohrom.

Conoscopic pattern crystal $LiNbO_3$:Mg [0.01 - 1.5 mol·%] and $LiNbO_3$:Mg [3.0 - 5.5 mol·%] are very different.

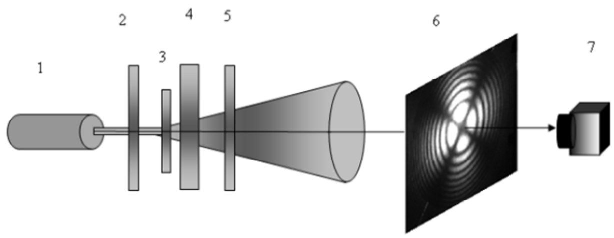
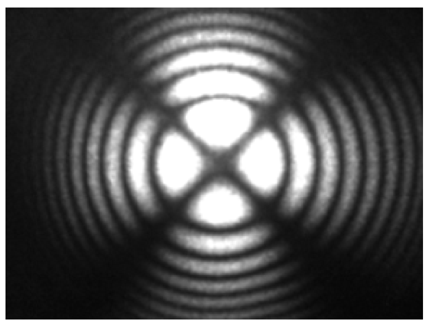
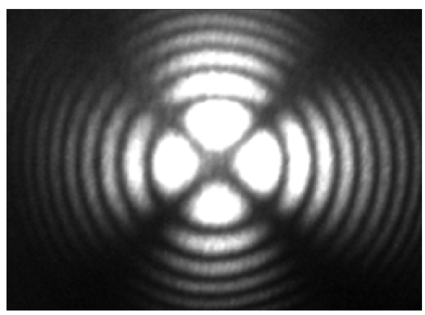


Figure 1. Diagram of the optical sign identifying system: 1–He-Ne laser; 2–polarizer; 3–diffuser; 4–investigated crystal plate; 5–analyzer crossed with polarizer; 6–screen; 7–camera.



(a)



(b)

Figure 2. Conoscopic pattern of single crystals of $LiNbO_3$: Mg:(a)–[0.5 mol·%]; (b)–[1.0 mol· %].

When scanning the plane entrance face with a rather high concentration of the impurities $LiNbO_3$:Mg [3.0 mol· %], in addition to standard patterns and similar in appearance (**Figure 3(a)**) were observed and the distorted conoscopic pattern (**Figures 3(b)-(e)**).

On conoscopic pattern (**Figure 3(b)**) black"maltese cross" cut in half with the enlightenment in the center field of view. The azimuthal direction of displacement on parts of "maltese cross" amounts to the angle of ~ 10° - 13° clockwise from the vertical. Isochromen keep integrity, but some extend in the direction of displacement of the fragments of the cross and take the form of ellipses with the attitude of the minor and major axes of ~ 0.9:1.

On conoscopic pattern (**Figures 3(c), (d), (e)**) the black "maltese cross" in the center of the field of view, on the contrary, is an integer, and retain the form isochromen rings. However, in the periphery of the field of view at a considerable angular distance from the center of the picture, starting with a 5 - 6th isochromen, in only one branch of the "maltese cross" is observed by imposing additional interference structure. While the remaining three branches "maltese cross" retain their usual form.

All observed by scanning the plane entrance face conoscopic pattern $LiNbO_3$:Mg [5,0 mol.%] Characteristic of uniaxial crystals, as indicated by the black "maltese cross" on the background of the rings-izohrom (**Figures 4(a)-(e)**). However, on some conoscopic patterns on a small angular distance from the center of one of the four branches of the "maltese cross" there is the imposition of additional distinct interference fringes (**Figures 4(b)-(e)**).

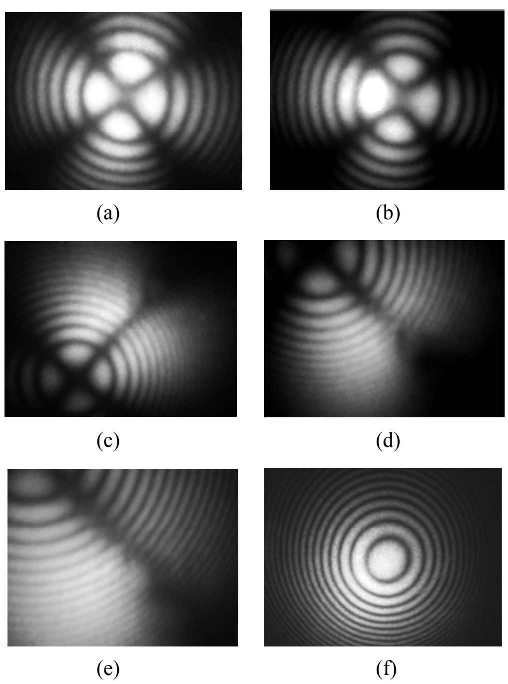


Figure 3. Conoscopic pattern of single crystal of $LiNbO_3$: Mg:[3.0 mol· %]

Conoscopic pattern samples with the highest concentration of the dopant $LiNbO_3$:Mg[5.5 mol· %]. Characteristic of uniaxial crystals (**Figures 5(a)-(e)**), but at some point the input face light up with some pictures of conoscopic anomalies. One type of anomaly is a superposition of additional interference pattern at an angular distance from the center, corresponding to a 3 - 4th isochromen, inone branch of the "maltese cross" (**Figures 5(b)-(c)**).

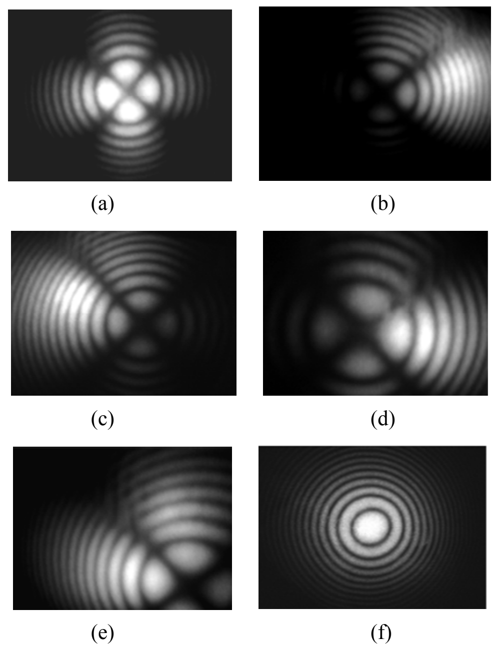


Figure 4. Conoscopic pattern of single crystals of $LiNbO_3$: Mg[5.0 mol· %].

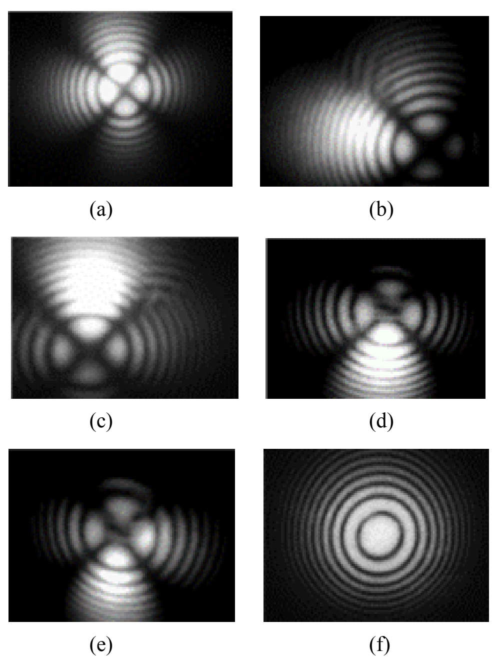


Figure 5. Conoscopic pattern of single crystals of $LiNbO_3$: Mg [5.5 mol· %].

Another kind of anomaly is manifested as additional interference pattern, but in the center of the field of view of a conoscopic pattern on the background of black crossing branches “maltese cross” It should be noted that the conoscopic patterns of each of the three samples $LiNbO_3$:Mg[3.0 - 5.5 mol· %] with circular polarizer and the analyzer, which allows you to remove beclouding “maltese cross” have a standard form of rings and show no noticeable distortion (**Figures 3(f), 4(f) and 5(f)**).

Results conoscopic method study of crystals $LiNbO_3$, doped Mg cations to varying concentrations of interest, grown under different conditions, show that lightly doped lithium niobate samples containing Mg [0.003 - 1.0 mol· %] have a high optical homogeneity.

Analysis of the effect of the dopant Mg on the form the conoscopic pattern $LiNbO_3$:Mg showed that when the concentration of Mg dopant in the samples with the same geometric parameters of the scale of the conoscopic pattern, intensity distribution, shape and size of the “maltese cross” and izohrom saved.

Conoscopic technique to study samples of lithium niobate with the content of Mg [3.0 - 5.5 mol· %] suggests that a stronger doping Mg cations while maintaining overall uniaxial crystal leads to the appearance of local birefringent inclusions, which are recorded in the form of additional interference pattern on the background main conoscopic pattern in the center of the field of view, and in its peripheral region.

Small anomalous biaxiality in a bounded domain is registered for a sample $LiNbO_3$:Mg[3.0 mol· %], which is confirmed by the break and enlightenment “maltese cross” in the center of the conoscopic pattern of the crystal.

The differences in conoscopic patterns of single crystals of $LiNbO_3$: Mg [0.01 – 1.5 mol· %] and $LiNbO_3$:Mg [3.0 - 5· 5 mol· %] can be explained as follows.

Feature of lithium niobate single crystals doped with cations Mg^{2+} at relatively high ($\geq$ 3 mol· %) dopant concentration is uneven of impurity [13, 14] and, therefore, the appearance of growth bands associated with gradients of dopant concentration, as in plane perpendicular to and in the plane parallel to the growth axis (**Figures 6-7**).

Banding is accompanied by the growth of microdefects in the form of dislocations, microdomains of domain walls and block structure, especially in the high impurity concentration gradients at the boundaries of growth bands (**Figure 8**).

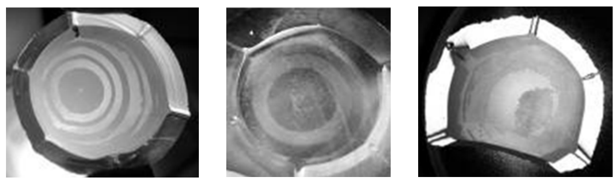
Growth bands, the gradient of the impurity concentration, the concentration of microdefects leads to a local change of the elastic characteristics of the crystal and appearance of mechanical stress [14], locally distorting the optical indicatrix of optically uniaxial crystal.

This leads to a distortion of the conoscopic patterns (**Figures 3-5**). Moreover, the maximum distortion is observed for the conoscopic patterns on the borders growth bands, where the concentration of structural defects and the dopant concentration gradients are maximized.

In the series of crystals investigated by us striation of samples, in general, decreases with increasing impurity concentration from 3.0 to 5.5 mol· % (**Figures 6-7**). In the same row is somewhat reduced degree of distortion conoscopic patterns (**Figures 4-5).**

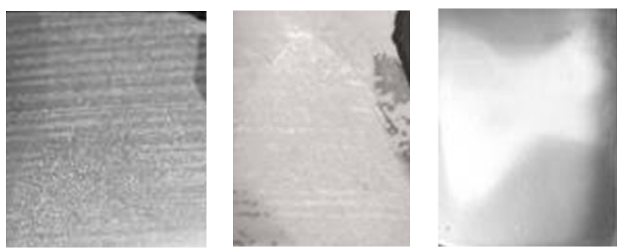
Thus, the deficiency of the crystal associated with the inhomogeneity of admixture disposition, passes through a maximum at a certain concentration of ~3 mol· % Mg^{2+}. The latter may be due to a change in the mechanism of admixture disposition when changing dopant concentration [13, 15].

In particular, the research methods of microanalysis found a reduction ratio R=Li/Nb (0,94) at a concentration in the crystal Mg^{2+} ~ 3% [16]. With such a concentration of Mg^{2+} defects Nb_{Li} (cation Nb^{5+}, are in the positions of lithium of the ideal structure of the stoichiometric composition) completely forced out in the cation sublattice [13].



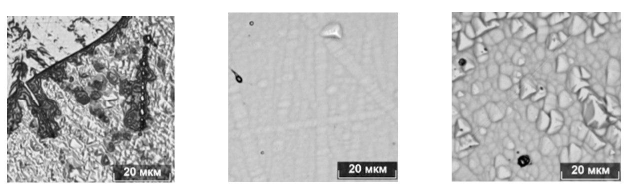
(a) (b) (c)

Figure 6. Bands of crystal growth LiNbO3: Mg in the plane perpendicular to the growth: (a)–[3,0 mol.%]; b–[5,0 mol.%]; c–[5. mol· %].



(a) (b) (c)

Figure 7. Bands of crystal growth LiNbO3: Mg in the plane parallel to the growth:(a)–[3.0 mol· %];(b)–[5.0 mol· %];(c) – [5.5 mol· %].



(a) (b) (c)

Figure 8. Microdefects at the boundaries of crystal growth bands $LiNbO_3$: Mg in the plane perpendicular to the axis of growth:(a)-3.0 mol· %];(b) -[5.0 mol· %];(c)-5.5 mol· %].

At a concentration of $Mg^{2+} > 3\%$ are replaced by base cations Li^+, accompanied by an increase of defects V_{Li} (vacant oxygen octahedrons, which in an ideal structure stoichiometric composition should be located the cations Li^+) [13,16].

When approaching the value R = Li/Nb to the value of 0.84 [$Mg^{2+} \geq 8\%$], corresponding to the stability boundary $LiNbO_3$ phase in the phase diagram [13], the cations Mg^{2+} are included in both (lithium and niobium) position of the ideal structure of stoichiometric composition with a simultaneous decrease concentration of compensating defects V_{Li} [13,16].

4. Conclusions

Results conoscopic method study of lithium niobate crystals doped with cations Mg^{2+}, show that samples containing Mg^{2+} (0.01 - 1.5 mol · %), even cut from different crystals, grown under different conditions, have high optical homogeneity.

And the scale of the conoscopic pattern, intensity distribution, shape and size of the "maltese cross"and izohrom are fully maintained for samples with the same geometric parameters.

Stronger doping cations Mg^{2+} (3.0 - 5.5 mol · %), while maintaining overall uniaxial crystal leads to the appearance of local birefringent inclusions, which are recorded as additional interference pattern on the background main conoscopic pattern in the center of the field of view and in its peripheral region.

A small anomalous biaxiality in a limited region is registered for a sample containing Mg^{2+} 3.0 mol · %, which confirmed the break and enlightenment "maltese cross"in the center of the conoscopic pattern of the crystal.

When the concentration of the dopant appears uneven of impurity, accompanied by the appearance of growth bands and microdefects in the form of dislocations, microdomains of domain walls and block structure, especially in the high impurity concentration gradients at the boundaries of growth bands.

Defect structure of the crystals, associated with irregular occurrence impurities leads to a local change of the elastic characteristics of the crystal and the appearance of mechanical stresses that cause distortion of the conoscopic patterns. And the imperfection of the crystal samples and the degree of distortion conoscopic patterns, generally decrease with increasing impurity concentration from 3.0 to 5.5 mol · %.

Thus, laser conoscopic research technique on the proposed scheme in this paper to identify subtle changes in the optical properties of the crystal at its doping. Conoscopic same study samples under a polarizing microscope, in which the light source is used as a incandescent lamp, make it possible to observe a picture of the crystal as a whole, without detailing the fine features of the structure in the form of local distortions.

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