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Photoinduced domain origin in Lithium Niobate crystals

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ABSTRACT

Light influence on $LiNbO_3$: Fe crystals has been found to induce appearance of needle-like microdomains with average length of 1-2 mm and diameter of 1 μ. Light scattering by photoinduced microdomain array has been investigated and found to be a seeding source of polarizationally anisotropic photorefractive light scattering. The effect of unlocality of forming the microdomains has been revealed. Photoinduced charge redistribution at the crystal surface has been found to cause the domain formation and rebuilding the crystal structure.

Keywords : domains , photorefraction , surface chardge , light scattering .

1. INTRODUCTION

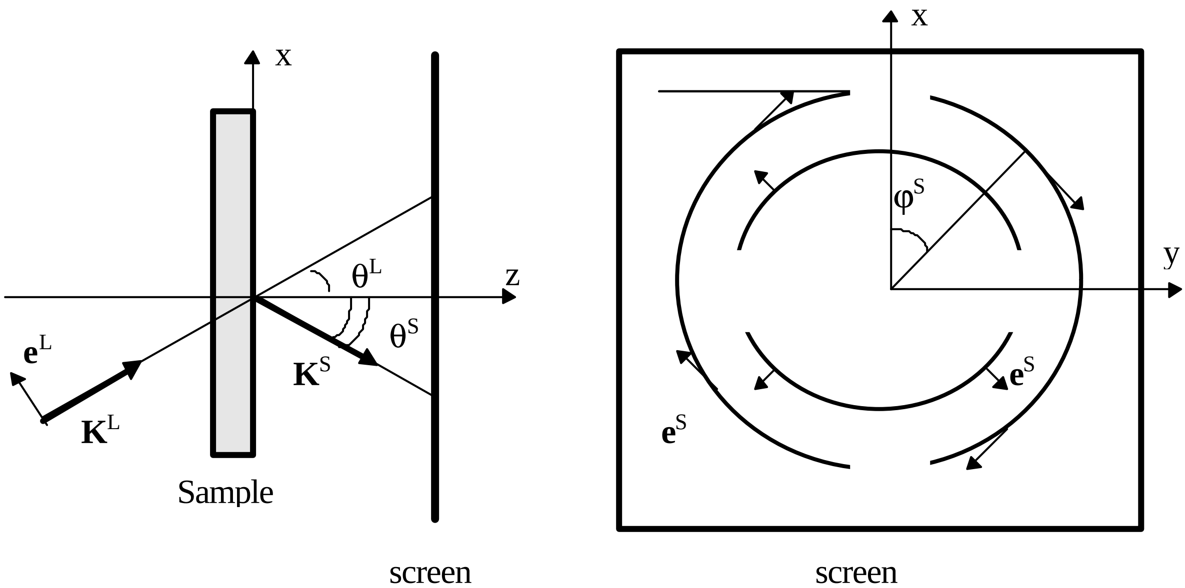
Repolarization of lithium niobate crystals is known to be hampered for large values of both the Curie temperature (T_c=1480^o K) and coercive field (E_c ~ 300 kV/cm) [1,2]. However light influence on the crystals was observed to be able to affect the domain structure [3-5].

In this paper, microdomain structure induced in $LiNbO_3$ by the light flow of small power (10 - 20 mW) is investigated. Besides light scattering by the microdomains is studied and suggested to be a seeding source of photorefractive light scattering [6].

2. EXPERIMENTAL RESULTS AND DISCUSSION

$LiNbO_3$: Fe single - domain crystal plates of z-cut have been used as the experimental samples. The crystals were grown by the Chokhralsky method with typical ratio Li/Nb ~ 0.95 ; the iron concentration was about 0.03 wt.% measured in the melt. The typical dimensions of the samples were $L_x \sim L_y$ ~ 15 - 30 mm, L_z ~ 1.2 -4.0 mm.The experimental setup is shown in Fig.1. He-Cd laser radiation (λ_L =0.44 μ , P_L=20 mW) was used as the pump. The laser beam (with the diameter 2 mm) was directed into the sample at angle of incidence θ^L (relative to z-axis of the crystal). Besides the special light scattering appeared in a few minutes. The scattering indicatrix was located at surface of the cone with the angle of 2 θ^L. The scattering cone axis was always directed exactly along z-axis. Note the scattering can be induced by appropriate incoherent light as well. After stationary state of the scattering was reached, change of pumping angle of incidence up to 30^o only caused noninertial change of the scattering cone angle without any change of the total scattering intensity.

Placing the sample into conductive media made the scattering instantly disappear.



a) b)

Fig.1. Experimental setup (a) and schematic structure of scattering intensity distribution on the screen (b) (S - crystal sample ; $\mathbf{K}^{L,S}$ - wave vectors of pump and scattering light ; $\mathbf{e}^{L,S}$ - orts of polaryzation).

The peculiarities pointed out above as well as the effect of space unlocality of the scattering to be described bellow testify that the conic scattering substantially differ from such well known types of scattering as photorefractive light scattering, beam fanning or parametric effects[7-9]. The lasts disappear unless appropriate wave synchronism conditions are realized. But such conditions are being broken under change of pumping angle of incidence.

The conic scattering features can be explained within the supposition that a set of needle-like microdomains oriented along z-axis of the crystals are induced under light influence on the crystal. Experiments with chemical etching the crystal surfaces have really displayed the microdomain butt-ends. Indeed, such microdomains can be formed in lithium niobate crystals due to the symmetry properties [10].If the domains of transversal dimension $a_d \sim \lambda_L$ are long enough $z_d >> \lambda_L$ (the previous investigations[5] brought the following results : a_d=0.9 μ m,z_d=1200 μ m,surface density of the domains $\sigma = 10^6$ cm^{-2}) the light scattering will take place under the synchronism conditions $K^l_s = (K^s_o)_z = (K^s_e)_z$, where K^L, K^S are wave vectors of the incident light and the scattering accordingly, “o” and “e” mean ordinary and extraordinary scattering components polarization. The synchronism conditions determine two scattering cones at angles θ^S_o, θ^S_e.

At small $\theta^{L,}$ finding $\Delta\theta$ is not complicated : $\Delta\theta = \theta^S_o - \theta^S_e = 2\,\Delta n_{oe} / (n_o + n_e)$. Using the values of refractive indexes $n_o = 2.297$, $n_e = 2.208$ for lithium niobate crystals [10] one can get $\Delta\theta = 0.042\ \theta^{L.}$

The azimutal dependence of scattering intensity is mainly determined by both polarization types of pump and scattering (o - ordinary, e - extraordinary) and appropriate orts $\mathbf{e}^{L,S}(\theta,\varphi)$ viz. $I_S(\pi_L, \pi_S) \sim z_d\, P_L(e^L, e^S)\, P_L(\vec{e}^{\,L}, \vec{e}^{\,S})$. Finding the dependence for scattering at z-axis is not complicated :

$$\begin{aligned} I_S(o,o) \approx I_S(e,o) \approx C_1 \cdot \sin^2 \varphi, \\ I_S(e,e) \approx I_S(o,e) \approx C_1 \cdot \cos^2 \varphi \end{aligned} \qquad (1)$$

(φ – is the angle between x-axis and projection of $\vec{k}_S$ on XY plane). The obtained dependence agree with experimental data (Fig. 1 b).

The correct polarization measurements brought the experimental value of $\Delta\theta$:

$$\Delta\theta = (0.040 \pm 0.006)\ \theta^L \qquad (2)$$

that is in a good agreement with the above. Note that integral intensity of the conical scattering weakly depend on the pump beam polarization $\mathbf{e}^L$.

As the conic scattering instantly disappears after placing the crystal into conductive media, the role of surface charges is considered to be important for domain forming. Direct measurement of integral surface charge value Q brought such a result : $Q = 1.7\ 10^{-10}$ coul independently on P_L at $P_L \geq 10$mW. The appropriate internal field reached the value of E_f = 70 kV/cm.

Note another peculiarity of both the domain forming and accompanying conic scattering to be further named “the effect of unlocality “. Under testing the photoexited crystal by an inactive light beam ($\lambda_T = 0.63\ \mu$ m, $P_T = 1$ mW), the conic scattering of the testing light was found to appeared first far enough (~ 10 mm) from the region illuminated by the pump (with diameter of 2 mm) and only a few minutes latter the conic scattering appeared in the crystal region illuminated by the pump.

Investigation of surface charge density distribution σ(x,y) was performed by electrographic development method[14] to study the effect of unlocality. At “-z” surface the positive charge has been found only to concentrate in the pump localization area. At the same time at “+z” surface, the negative charge appeared to be localized at the centre of the pump illumination area making a circle with the diameter of 4 - 5 mm. Besides the positive charge formed a ring - shaped zone with average diameter of ~12 mm. Just in that zone did the conic scattering of testing beams (and consequently, microdomains) appear first of all.

The results described above can be explained within the supposition that the photogalvanic effect[2,10] is the main cause of the charge generation at z-surfaces of the crystal. Direction of photoinduced electron motion due to the photogalvanic effect (from "-z" side to "+z" side) confirms the supposition. Besides at "-z" surface, positively charged deep traps (Fe^{3+} ions) remain motionless and only localized in the area illuminated by the pump. At "+z" surface, mobile photoinduced electrons having filled up all the deep traps, begin shifting under influence of the Coulomb forces and finally form a circle with the diameter to be bigger than exitant beam one. At the same time the positive charge carriers (for instance, oxygen vacancies or surplus ions Li^{+})[2,10,] begin flowing from the crystal edge to the negative charge localization area making the positive charged ring pointed out above. At that ring-shaped area, 180^o - repolarization is energetically profitable as negatively charged butt-ends of the microdomains reduce both the surface charge density and surface energy density $U \sim \iint \sigma^2(x,y)dxdy$. At central negatively charged circle-shaped zone 180^o - repolarization only could increase the surface charge density. However 180^o - microdomain forming at that zone is profitable from another point of view : it brings to decrease of photogalvanic current value since repolarized regions generate the photogalvanic current in invert direction. Of course, such repolarization takes place at less favourable conditions compared with positively charged ring-shaped zone.

Note that optically undistinguishable (under usual conditions) 180^o domains can be distinguishable under influence of the photogalvanic electrostatic field $\mathbf{E}^g$ due to electrooptical effect[2,10].Besides, accordingly to the surface charge distribution, the field $\mathbf{E}^g(\mathbf{r})$ has significant transversal components $E^g_{x,y}$ that brings to the rotation of the passing light polarization plane. The appropriate angle of turning of $\mathbf{e}^L$ reaches the value about $\pi/2$ in the thick crystals ($l_z \geq 1$mm) making the conic scattering practically independent on initial light polarization outside of the crystal.

The repolarization processes in ferroelectrics are known to be depend on temperature. In this connection, temperature investigation of the conic scattering have been performed. The scattering has been find to have two maxima at both temperature range T_1^{max}= 70 - 80^o C and T_2^{max}= 110 - 120^o C.Temperature dependence of the polarization jumps rate in $LiNbO_3$ is characterized by two maxima at both of the temperature ranges as well[11]. Other characteristics of $LiNbO_3$ (e.g. electrooptical index, double refraction index, inner rubbing coefficient) are also characterized by anomalies at the temperatures pointed out. By the way, the special structural transition without crystal symmetry change in $LiNbO_3$ at T=75^o C takes place [12]. In the last case a neighbouring oxygen triangles turn by jump in opposite directions, i.e. an original phase transition is realized. Shift of Li and Nb ions being relieved under conditions of unsteady equilibrium of the crystal structure, the probability of repolarization with 180^o - microdomain formation increases at the temperature ranges pointed out above.

Redistribution of photoinduced charge carriers take place not only under illumination of z-cuts of $LiNbO_3$ crystals, but under photoexitation of both x- and y- cuts also. In the last case surplus charge carriers are localized in the depth of the crystal at illuminated region border. Microdomains appear at the border as well. They have been displayed by testing photoexited (along x- or y- axis) region by inactive light beam propagating at small angle to z-axis. Conic scattering of the testing beams showed that the microdomains had the same parameters as those under photoexitation of z-cuts.

The pump could be also scattered by the microdomains. The microdomains being oriented perpendicularly to the pump propagation direction, the appropriate conic scattering should have indicatrix localized at XY plane. Besides, the scattering contains both ordinary and extraordinary polarization components. The same characteristics have polarizationally anisotropic photorefractive light scattering (oe-PRLS) in $LiNbO_3$:Fe[13]. Taking the above into consideration, the conic scattering of the pump could be a seeding source of oe-PRLS. To verify the supposition, the kinetics of both PRLS (of different kinds) and accompanying conic scattering have been studied (Fig. 2). The obtained data showed both oe-PRLS and the conic scattering to appear at the same time. Besides, both of the processes are characterized with the same delay time t_o too.

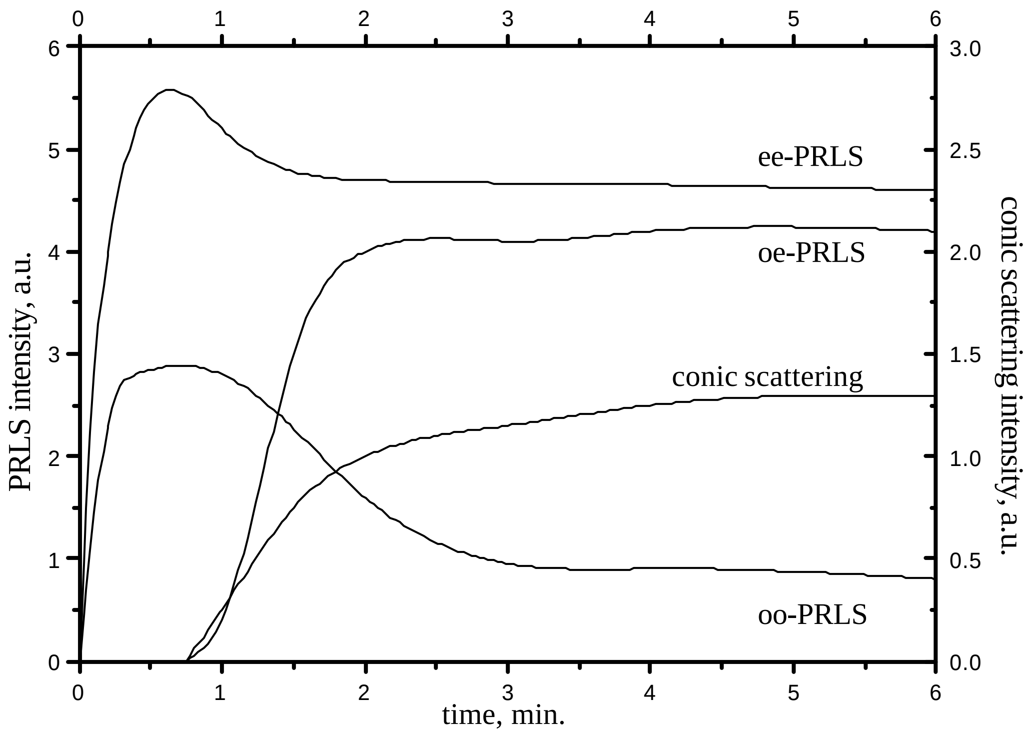


Fig.2.Kinetics of photorefractive light scattering (PRLS) and conic (microdomains) scattering. (indexes denotes polarization of pump and scattering beams).

Note that under conditions which make appearance of photoinduced domains impossible (e.g. at homogeneous illumination of the crystal placed into conductive liquid), oe-PRLS appears considerably later (the delay time is 3-4 times more then t_o). Besides PRLS intensity increases slower.

Thus the conclusion may be drawn that the conic scattering can be an effective seeding sources of polarizationally anisotropic light scattering in ferroelectric crystals.

3. ACKNOWLEDGMENTS

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4. REFERENCES

1. F.Jona and G.Shirane, Ferroelectric crystals, Pergamon Press, Oxford, 1962.

2. M.E.Lines and A.M.Glass, Principles and applications of ferroelectrics and related materials, Clarendon Press Oxford, 1977.

3. V.I.Kovalevich, L.A.Shuvalov and T.R.Volk, "Spontaneous polarization reversal and photorefractive effect in single domain $LiNbO_3$:Fe crystals", Phys. Stat. Sol.(a), Vol. 45, N 1, pp.249 -252, 1978.

4. F.Kahmann, R.Pankrath, R.A.Rupp, "Photoassisted generation of ferroelectric domain grating in SBN ", Optics Communications, Vol. 107, N 1, pp. 6 - 10, 1994.

5. V.V.Lemeshko, V.V.Obukhovsky and A.V.Stoyanov, "Photomicrodomain light scattering in lithium niobate crystals ", Photorefractive Materials, Effects and Divices. Techn. Digest, pp. 351 - 353, Kiev, August 1993.

6. R.Magnusson and T.K.Gaylord, "Light scattering induced holograms in lithium niobate", Appl. Optics, Vol. 13, N 7, pp. 1545 - 1548, 1974.

7. S.Odoulov, B.Sturman, L.Holtmann, E.Krätzig, "Nonlinear scattering in $BaTiO_3$ induced by two ortogonally polarized waves ", Appl. Phys., Vol. B52, N 2, pp. 317 - 322, 1991.

8. J.Marotz, K.H.Ringhofer, R.A.Rupp, S.Treichel, "Light induced scattering in photorefractive crystals ", IEEE Journ. Quant. Electronics, Vol. 22, N 10, pp. 1376 - 1383, 1986.

9. M.Segev, D.Engin, A.Yariv,"Temporal evolution of fanning in photorefractive media ", Optics Letters, Vol. 18, n 12, pp. 956 - 958, 1993.

10. Yu.S.Kuzminov, Electrooptical and nonlinear crystals - lithium niobate ", Nauka, Moscow, 1987.

11. V.P.Kamentsev, A.V.Nekrasov, B.B.Pedko and B.M.Rudiak, "Investigation of physical properties of lithium niobate single - crystal within temperature range of 20 - 200^0 C ", Izv. Akad. Nauk SSSR,Ser. Fiz., Vol. 47, N 4, pp. 791 -793, 1983.

12. J.S.Zhu and X.H.Yang, " SHG study of the anomalous properties of $LiNbO_3$ crystals near 75^0 C ", Chinese Phys., Vol. 5, N 4, pp.980 -984, 1985.

13. S.G.Odoulov and M.S.Soskin "Amplification, osscillation and light induced scattering in photorefractive crystals " in : "Topic in Applied Physics, Vol. 62,"Photorefractive materials and their applications II ", edit. by P.Gunter and J.-P.Huignard, Springer - Verlag, Berlin, 1988.

14. D.M.Pai and B.E.Springett, "Physics of electrography ", Rev. Mod. Phys., V. 65, N 1, pp. 163 - 211,1993.