Laser beam control devices on the base of the lithium niobate single crystals

Anatol T. Mikhalevich[1], Andrii O. Matkovskii[1,2], Dymitrij Y. Sugak[1]
Ivan M. Solskii[1], Vladimir M. Gaba[1], Bogdan M. Kopko[1], Marian Kuźma[2]

[1)] RPA "Carat"
202 Stryjska str., Lviv, 290031, Ukraine
[2)] Institute of Physics, WSP
ul. Rejtana, 35-310, Rzeszów, Poland

1. Introduction

Lithium niobate $LiNbO_3$ (LN) single crystals have the unique set of acousto-optic, piezoelectric, electrooptic, nonlinear optical and other characteristics which determine the application of them in varied fields of electronic engineering, including devices for modulation, deviation, laser radiation frequency conversion and also for holographic recording [1, 2]. A great number of publications, including some monographs [3÷5] are devoted to the investigation of the LN single crystals growth processes, their physical-chemical properties and operational parameters.

At the same time there are some difficulties in production of large LN crystals of more than 3 inches in the diameter and 4 inches in length with high degree of optical homogeneity. Absence of a reproducible technology for production of such a kind of crystals hampers to a certain extent spreading its usage in nonlinear and polarization optics, because for production of parametric light generators of infrared range, polarization prisms and other devices of range 0.5÷4.5 μm require large (1÷50 cm^3) and optically homogeneous (inhomogenity of the refractive index no less than $2.5 \cdot 10^{-5}$ per cm of crystals lenth) single crystal blocks [5, 6]. To solve such a problem there are necessary techniques of optical homogeneity control and verification that will allow to choose the quality areas from the volume of a boule.

Studying of LN beam resistance as well as ionizing radiation and annealing effect investigations are great importance, because the irradiation and thermal action significantly influence on optical and other physical properties of LN.

This work is devoted to production of large LN single crystals and determination of their optical homogenity, investigating of their thermo- and radiation induced changes of optical properties as well as the problem of polarization prisms and optical blocks production from the LN crystals for laser equipment.

2. Growth of LN single crystals

With the purpose of large LN crystals production of high degree of optical homogenity allowing for their use in polarization and nonlinear optical devices, a complex set of technological experiments was carried out, including preparation of the load and the growth parameters optimization.

The starting load of stoichiometric compound was preliminary recrystallized which made it possible to provide low content (<1 ppm) of unwanted impurities in the raw, and congruent ratio of oxides $R = Li_2O/Nb_2O_5 \approx 0.96$.

The crystals were grown in air by Czochralski method from platinum crucibles using PHYSITERM (France) equipment with high frequency heating and automatic boule diameter control by weight method.

It is known that the one of the basic factors providing the crystal homogeneity is the stability of temperature gradients in the growth zone and maintaining them in the state that provides flat liquid-solid interface. To achieve this, as well as to maintain the appropriate conditions for aftergrowth annaeling, there was designed a special heating unit with the use of a platinum screen and a double alumina thermal insulation [7].

To control the boule morphology (ie., the length of the conic parts and the diameter) there were used non-stationary growth conditions (rotation and pulling speed). The use of the above mentioned methods made it possible to produce *Z*-grown crystals with 80÷85 mm diameter and the length of the cylindrical part up to 100 mm.

3. Investigations of crystal optical homogeneity

Two techniques for studying of crystal optical homogeneity were used. The first one was based on observation of conoscopic image during passing the light along the crystal optic axis (*Z*) [8], and the second one is based on the phanomena of spontaneous parametric scattering of laser radiation ($\lambda = 488$ nm) in case of light spreading in the direction of crystallographic *X* axis of LN crystals [9].

A device for measuring the crystals optical homogeneity by a conoscopic method consist of a light source, (He-Ne laser with 0.63 μm wavelength), and crossed polarizer and analyzer with a crystal located between them. The crystal was placed on a two-direction movable table which moved perpendicularly to the optical beam. The optical axis of a crystal coincided with the direction of light beam. A glass diffusing plate is placed near the crystal in the incident laser beam. The conoscopic image was observed in the plane behind the analyzer. In such case the conoscopic image is similar to one obtained for crystal with single optical axis observed along this axis [10]. For a high quality area of a crystal, a homogeneous dark cross formed by isogyres was observed.

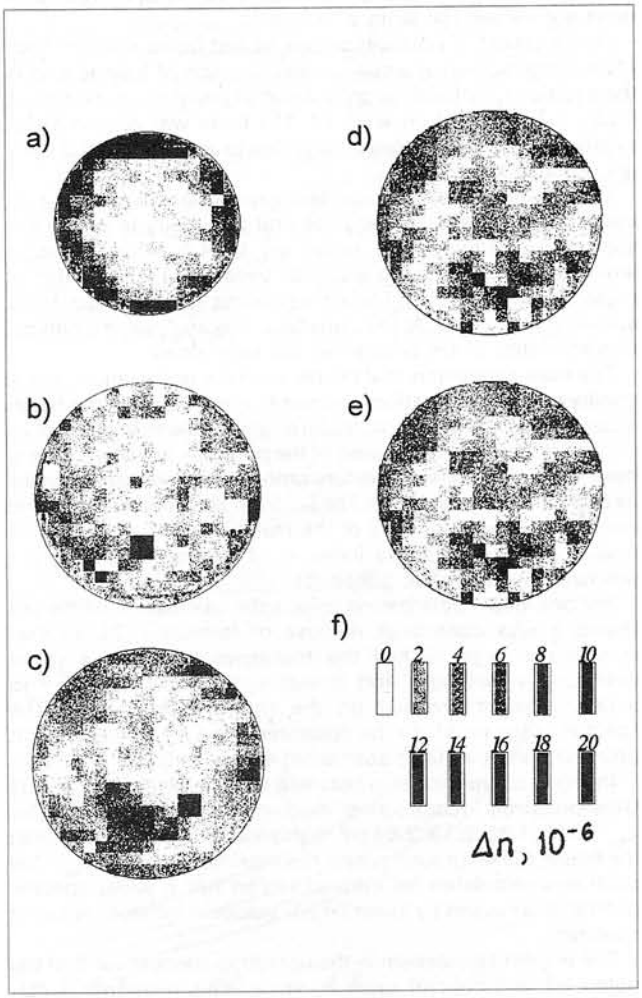

Fig. 1. Topograms of Δn_a values in series crystal crosssections (a – boule beginning, e – boule end). Depth of samples is 20 mm, boule diameter is 80 mm

In the case of optical inhomogeneity, the isogyres are separated and in the centre of the cross a bright spot is observed – the crystals becomes anomalous biaxial. The distance *b* between the isogyre extremum corresponding to the exist of the anamalous axes is directly connected with the magnitude of the angle between these axes (angle of anomalous biaxiality V_a). Value V_a may be found from the equation [4]:

$$V_a = \arcsin \frac{b/L}{n_0[1+(b/2L)^2]^{1/2}}$$

where n_0 is a ordinary refractive index at 0.63 μm and *L* is a distance between the crystals and the observation plane. The value of the anomalous birefrigence Δn_a is related with the V_a:

$$\sin V_a = \left(\frac{\Delta n_a}{n_0 - n_e}\right)^{1/2}$$

and may be calculated from the equation:

$$\Delta n_a = \frac{n_0 - n_e}{n_0^2} \sin^2 (\operatorname{arc\,tg}\, b/2L)$$

where n_e is the extraordinary refractive index.

The use of the scanning table allowed to find Δn_a value through the entire crystal surface perpendicular to the optical axis. The step-by-step investigation of the boule cross-sections starting from the upper cone to the lower one showed the general distribution of optical inhomogeneities in the crystals. The examples of Δn_a topograms are shown in Fig. 1. The thickness of the investigated sample was 20 mm. The Δn_a reaches its extrema in the cone and near surface areas of the boule ($10 \cdot 10^{-6} \div 50 \cdot 10^{-6}$). The most homogeneous are the central parts of the crystal, the least homogeneous are a and b cross-sections of the boule. The major part of the boule is characterized by the value $\Delta n_a = (1 \div 6) \cdot 10^{-6}$.

Thus, the conoscopic investigations allows to study optical homogeneity of LN crystals, proceeding from the value of anomalous birefringence, and to determine not only absolute values of Δn_a but also the change of its sign, to get the topograms of Δn_a and change of its sings and in such a way to make the selection of the areas best quality for production of optic blocks.

The results obtained by conoscopic method correlate well with the results of measurements in which the spontaneous parametric light scattering method was used (the measurements were carried out at INP of Belorussian University by M. Livshits and Yu. Voitukevich). This method allowed to measure the absolute changes of the refraction index along the light direction coinciding with crystallographic *X* axis. The measurements were made using a sample with the following dimension: 75 mm (*Y*), 85 mm (*Z*), 23 mm (*X*). The topogram of the value δn obtained with a 4 mm step in *YZ* plane is shown in Fig. 2. In the same figure the dependences of δn change along the boule diameter in the upper, middle and lower parts, as well as change of δn in the crystal centre along the growth axis (*Z*) are shown. As it may be seen on the picture, the most homogeneous areas of a crystal are located in one third and in the centre of the boule. The least homogeneous areas are located near the surface of the crystal and in the lower cone. In general, the worst values $\partial n_z/\partial x$ for this crystal is equal to $2.5 \cdot 10^{-5}$ cm^{-1}, that corresponds to the optic homogeneity criterium given in [5].

It should be noticed that the topogram of δn value has a certain axis symmetry and most propably it presents a crossection of *YZ* surface of the planes of the same composition which are determined by isothermic surfaces in each moment of crystal growth.

Thus, the investigations ahowed that the produced LN crystals despite their large dimensions have high degree of optical homogeneity, which is sufficient for selection of crystals areas of the required quality.

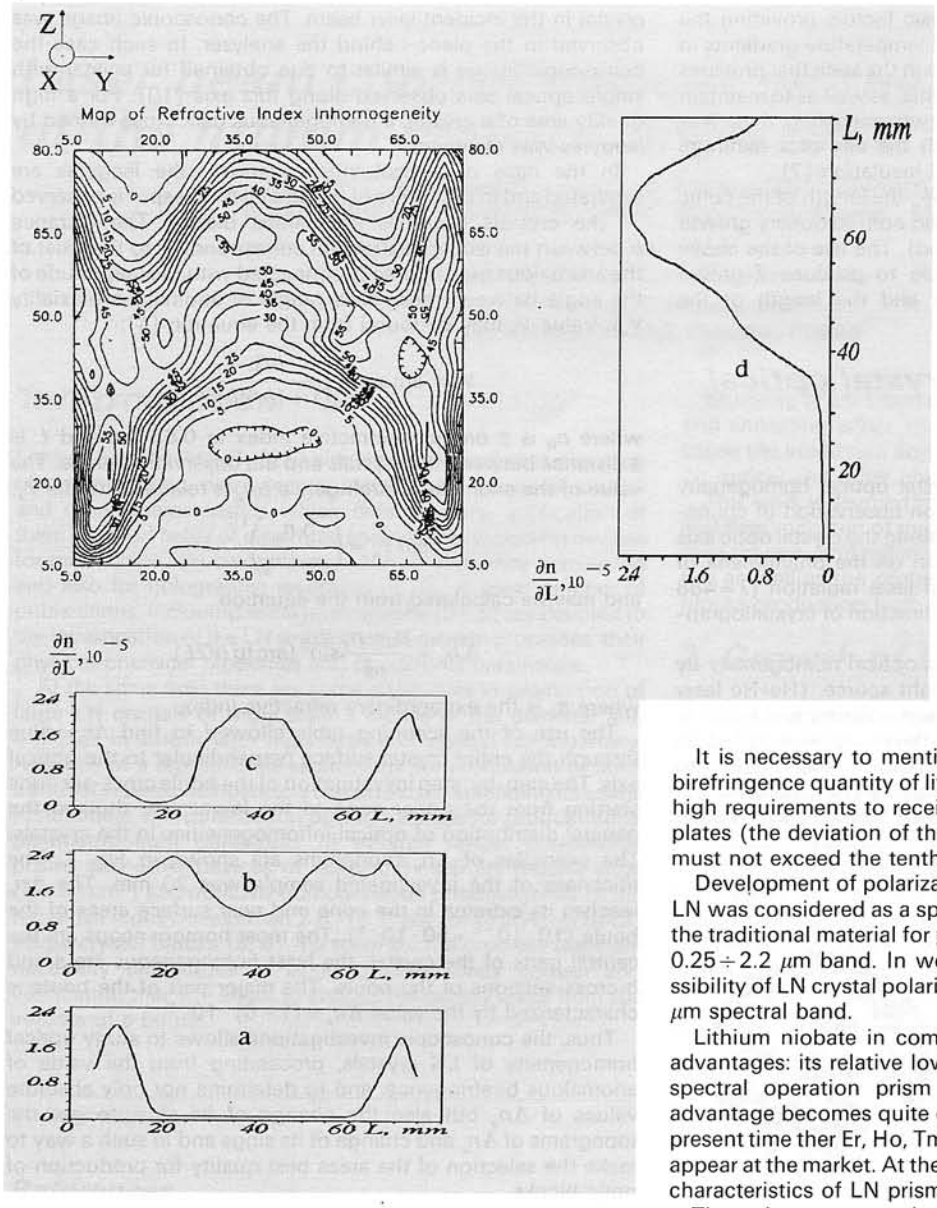

Fig. 2. Map of refractive index inhomogeneity δn ($\delta n \cdot 10^{-6}$, sample depth is 23 mm). Changes n_e to the boule diameter at the distance 15 mm (a), 40 mm (b) and 65 mm (c) from the boule beginning. Changes n_e along growth axis at the center of boule (d)

4. Q-switch active elements, wave plates and polarizers

A series of the grown crystals traditional optical elements for laser Q-switches were produced. The elements have the following dimensions of the entrance face in XY plane 3×3; 6×6; 9×9 mm^2 and the length in Z direction $20 \div 35$ mm. Due to the special selection of the crystal regions, the contrast that was measured as light intensity relation that had passed through an optic block at parallel and crossed polarizers comprized a value not worse than 1:250 throughout the whole entrance face.

The high optical quality of the obtained crystals in which swirls are absent, and with significant dimensions crystals areas having the birefringence constant quality over a surface and wide region of spectral transparence, permits to produce wave plates for the phase shift of the orthogonally polarized beams of $\lambda/2$ and $\lambda/4$ in the middle IR range (up to 4.5 μm).

$LiNbO_3$ crystals was cut in such way, that the optical axis was situated in the wave plate plane. After that, a standart grinding and polishing were done, so that the waveplates finishing (thick $\sim$1 mm) was realized using optical contact to the glass plate on the special technological equipment. The waveplates $\lambda/2$ and $\lambda/4$ with 0.63, 1.064 and 2.94 μm wavelength were obtained. The waveplates thickness of $\lambda/4$, for example for $\lambda = 2.94$ μm, was equal to 999.1 μm (order 22). The phase shift error for the obtained wave plates was not excuded $2 \div 4$%.

It is necessary to mention that the significant value of the birefringence quantity of lithium niobate causes the sufficiently high requirements to receive the necessary thickness of wave plates (the deviation of the thickness from the optimum value must not exceed the tenth of micron).

Development of polarization prisms and beam-splitters from LN was considered as a special task. Crystals of Iceland spar is the traditional material for production of polarizers operating in $0.25 \div 2.2$ μm band. In work [7, 11] there was shown a possibility of LN crystal polarization prism production for $0.5 \div 4.5$ μm spectral band.

Lithium niobate in comparison with Iceland spar has two advantages: its relative low price and possibility to widen the spectral operation prism range up to 4.5 μm. The latter advantage becomes quite essential, because of the fact that at present time ther Er, Ho, Tm lasers emitting in the $2 \div 3$ μm band appear at the market. At the same time, any analysis of technical characteristics of LN prisms has not been done.

The main parameters that characterize the polarization prisms quality are the polarization contrast P, operation spectral band, acceptance angle w, the deviation angle γ of the light beam of its original direction after passing of the polarizar, linear aperture α, base length to the linear aperture ratio L/a, the beams divergence angle β for beamsplitters. The polarizer specifications depend upon optical homogeneity of the material, prismatic elements quality and the refractive index n_c of media connecting the prismatic elements (air, adhesive).

For one-beam polarization prisms the calculation of the cut angles α was done with the use of formula [12], so that acceptance angle ω had the maximum value for a given optimized wavelength and it was symmetrical relatively to a perpendicular dropped on the entrance prism face. The spectral range in which the optimized for a given wavelength polarizer can operate, is connected with ω value.

In Fig. 3 the relationship between w and wavelength for LN Glan-polarizers (connecting medium - air), optimized for $\lambda_{opt} = 0.63$; 1.15; 2.13; 2.94 μm, is shown. As it may be seen from the figure, the w for such prisms has maximum value at λ_{opt}. The polarizers calculated for infrared region has a wider spectral band that is caused by lower $\partial n_e/\partial\lambda$ values in infrared region of spectrum.

The ω (and consequently, the operation spectral band of the polarizer) will depend upon n_c value. This relationship calculated for Glan-Thompson type polarizers with λ_{opt} of 0.63; 1.15; 2.13 and 2.94 μm is shown on Fig. 4. As it may be seen from the figure, the w significantly increases in this case.

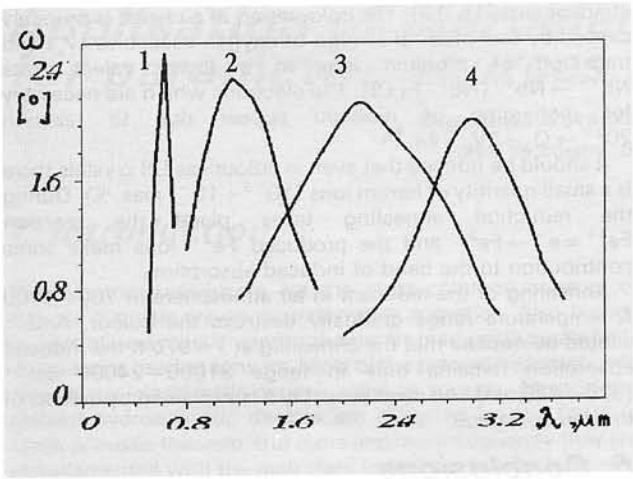

Fig. 3. Dependence of acceptance angle ω on wavelength λ for LN Glan polarizers (with air interlayer) optimized for wavelength 0.63 (1); 1.15 (2); 2.13 (3) and 2.94 μm (4)

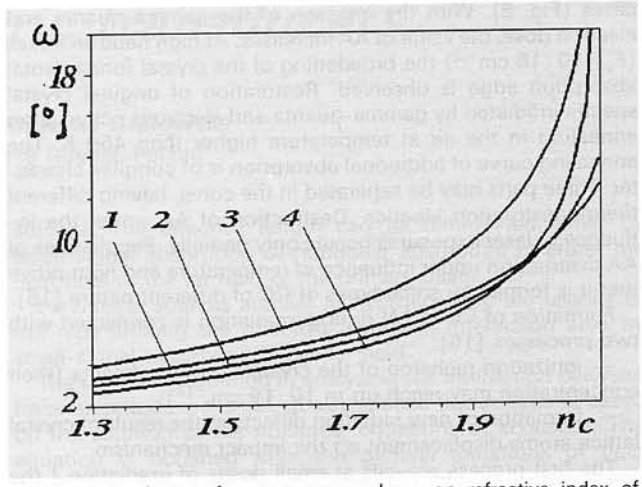

Fig. 4. Dependence of acceptance angle w on refractive index of adhesive interlayer n_e for LN Glan-Thompson polarizers optimized for wavelength 0.63 (1); 1.15 (2); 2.13 (3) and 2.94 μm (4)

For beam-splitters (adhesive joint by Canada balsam) the values of maximum divergence angles β and corresponding them cutting angles a calculated for λ_{opt} of 0.63 and 1.15 μm are given in the table.

Due to the decreasing of the cutting angle it is possible to decrease β angle. For Wollaston prism with adhesive joint of Canada balsam and calculated for $\lambda_{opt}=0.63$ μm at $\alpha=10°$ the $\beta=1.6°$.

In Fig. 5 the calculated relationship between boundary β angles Wollaston prisms with λ_{opt} equal to 0.63; 1.15; 2.13 and 2.94 μm and refractive index nc is given. As it may be seen from the figure, the β angle increases with the approach of n_e to the values of LN refractive index, however L/a value increases as well.

In comparison with Iceland spar the values of n_o and n_e for LN are much higher, while the value of natural birefringence is much lower: $n_o=2.28$, $n_e=2.20$, $\Delta n=0.08$ *for* $\lambda=0.63$ μm.

Relating this to the polarizer made of LN, much more rigid requirements are set on technological tolerance in the process of their production and on the accuracy of cutting angle a and un-parallelity of adhesive layer. For example, for Glan-Thompson type polarizers the same error of cutting in the angles of prismatic elements of LN and Island spar will bring to that γ angle of LN polarizer will be 2.5 times larger than that of Iceland spar.

Because of great values of LN refractive index the light reflection from the entrance face of polarizer reaches 14% and therefore, to decrease light loss it is necessary to give an anti-reflective coating of the prism faces.

Using the results of the analysis there were made some experimental Glan, Glan-Thompson and Wollaston prisms rated for wavelengths of 0.63; 0.85: 1.05; 1.15 and 2.94 μm, with linear apperture range from 5 to 20 mm.

For prism production the LN crystals areas were selected, having $\Delta n_a < 5 \cdot 10^{-6}$. With careful observation of technology, giving anti-reflecting MgF_2, SiO_2 and Y_2O_3 coatings and other high melting oxides, there were produced prism with polarization contrast P not worse than 0.9999. For GlanThompson and Wollaston prisms adhesive joint with Canada balsam $P=0.99998$, the γ angle did not exceed 10', and the value of reflection from the entrance face was not greater than 2%.

Thus a LN single crystal of high optical homogeneity may be quite suitable for production of polarizers and beam spitters mainly for middle infrared spectral range. At the same time, in comparison with polarizer made of Island spar, the production of polarizer of LN requires much careful observation of technology conditions and tolerances.

Table

λ_{opt}	β	α
0.63	8.2	42.5
1.15	7.8	43.0

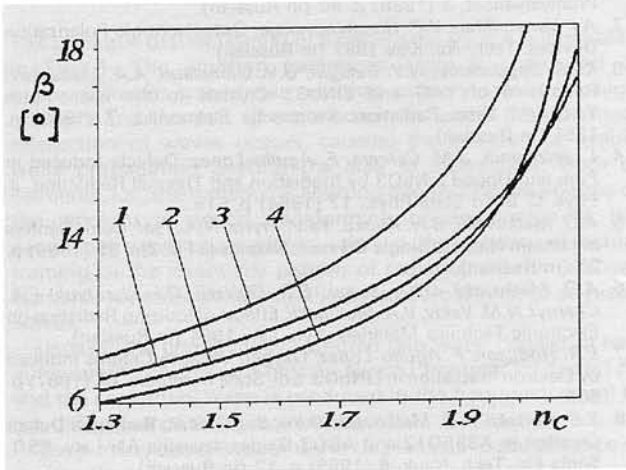

Fig. 5. Dependence of beam divergence angle β on refractive index of adhesive interlayer nc for LN Wollaston prisms optimized for wavelength 0.63 (1); 1.15 (2); 2.13 (3) and 2.94 μm (4)

5. Influence ionizing radiation and thermal annealing on LN crystal properties

The damage threshold of LN induced by laser radiation at wavelength 1.06 μm depending on crystal's optical homogeneity is $(1 \div 3) \cdot 10^8$ Wcm^{-2} and at 2.94 μm it is equal to $10 \cdot 10$ Wcm^{-2} [13].

LN single crystals are transparent in $31000 \div 2200$ cm^{-1} ($320 \div 4500$ nm) region. Under the influence of UV light and Xray beams, color centers (CC) in LN crystals arise, leading to the appearance of additional absorption (AA) bands and to change of refractive indices (photorefraction, X-ray refraction). These phenomena cause that laser optical information recording on LN crystals is possible [1, 2].

Significant change of LN crystal optical properties occur also as the result of the crystal annealing in different atmospheres [14]. This must be taken into account in production of optical elements.

Influence of high energy ionizing radiation also causes significiant changes of crystal optical properties. After irradiation of LN crystals by gamma-radiation, electrons and neutrons, a wide AA band with two maxima at 26500 (375 nm) and 22000 cm^{-1} (455 nm) appear in the absorption spectra. It should be noticed that the first maximum is prevailing in all the

cases (Fig. 6). With the increase of the gamma-quanta and electron dose, the value of AA increases. At high neutron fluxes ($F_n > 10 \cdot 16$ cm^{-2}) the broadening of the crystal fundamental absorption edge is observed. Restoration of original crystal spectra irradiated by gamma-quanta and electrons occurs after annealing in the air at temperature higher than 450 K. The annealing curve of additional absorption is of complex character. Three parts may be separated in the curve, having different thermodestruction kinetics. Destruction of AA under the influence of laser exposures occurs only partially. Peculiarities of AA destruction under influence of temperature and light prove that it is formed by some types of CC of different nature [15].

Formation of CC in LN during irradiation is connected with two processes [16]:

– ionization recharge of the crystals growth defects (their concentration may reach up to $10 \cdot 19$ cm^{-13});

– formation of new radiation defects in the result of crystal lattice atoms displacement on the impact mechanism.

The first process prevails at small doses of irradiation (the absorbed dose less than 10^6 Gy); at high doses of irradiation the constribution of the second process becomes noticable, CC absorption bands formed according to the first and the second mechanism are probably found in nearby spectral regions, that does not make it possible to distinct their contribution and brings to the absence of the saturation of AA dose dependencies.

Using known value of oxygen ion threshold displacement energy in the structure LN, $T_d = 52$ eV [17], there was made an attempt to estimate the displacement defects concentration in the oxygen sublattice of the crystal. At neutron fluences $F_n 10^{16}$ cm^{-2} and electron fluences ($E = 4$ MeV) $F_e > 10^{17}$ cm^{-2}, the number of the formed oxygen vacancies exceeds 10^{17} cm^{-3}, that is sufficient for arising of sample AA [18].

Annealing in vacuum at T = 870 K brings to colouration of crystal and arising of a wide absorption band in the range $31000 \div 12000$ cm^{-1} ($320 \div 800$ nm) (see Fig. 7). Analogous absorption also appears in crystals annealed in other reduction

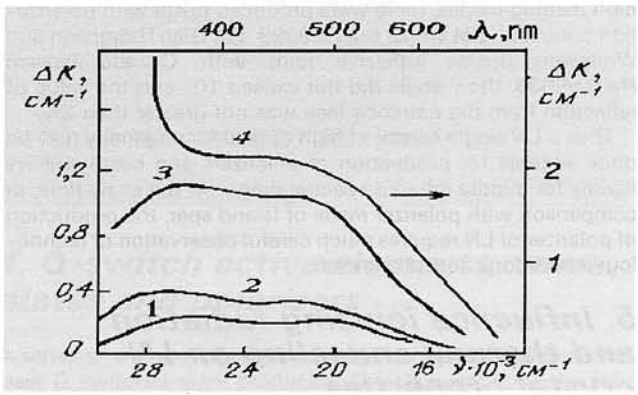

Fig. 6. The additional absorption spectra for LN crystals irradiated by γ-quanta with dose 10^5 Gy (1), electrons with energy 1.3 MeV and dose 10^6 Gy (2), electrons with energy 4 MeV and dose $5 \cdot 10^6$ Gy (3) and reactor neutrons with fluence 10^{18} cm^{-2} (4)

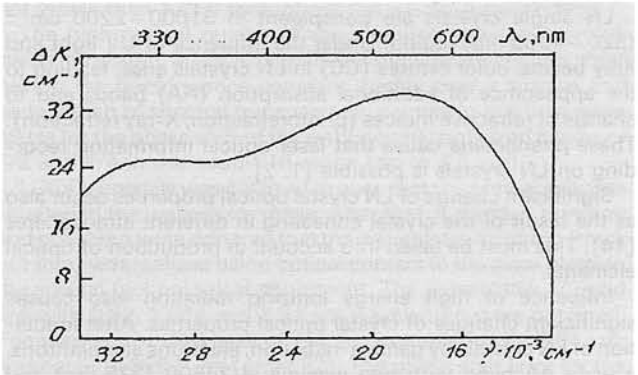

Fig. 7. The additional absorption spectra for LN crystals annealed in vacuum at the 870 K

atmospheres [15, 19]. The colouration of a crystal is propably caused by formation of foreign of oxygen vacancies (V_o) and transition of niobium ions in a lower valent state $Nb^{5+} \rightarrow Nb^{4+}(Nb^{3+})$ [19]. The electrons which are necessary for recharging of niobium appear due to reaction $2O^{2-} \rightarrow O_2 + 2V_0 + 4e^-$.

It should be noticed that even in colourless LN crystals there is a small quantity of ferrum ions ($10^{-3} \div 10^{-4}$ mas. %). During the reduction annealing takes place the reaction $Fe^{3+} = e^- \rightarrow Fe^{2+}$ and the produced Fe^{2+} ions make some contribution to the band of induced absorption.

Annealing of the reducent in air atmosphere in $700 \div 1100$ K temperature range gradually destroys the colour. And it should be noticed that the annealing at $T = 970$ K the induced absorption remains only in range $31000 \div 24000$ cm^{-1} ($320 \div 420$ nm) and most propably it is explained by defects of oxygen sublattice.

6. Conclusions

The production of lithium niobate of high optical homogeneity permit to use it successfully for the manufacturing the active elements of Q-switches, waveplates, polarization prisms mainly for IR range (up to 4.5 μm).

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