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Influence of magnetic field on the photorefractive effect in $LiNbO_3$ crystals

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Abstract. The photorefractive effect has been studied experimentally in single crystals of Fe and Mn-doped lithium niobate as a function of external static magnetic field. A strong dependence of changes in birefringence is observed on the magnitude and direction of the external field.

Keywords. photorefraction; lithium niobates; bulk-photovoltaics.

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1. Introduction

A local inhomogeneity of indices of refraction, giving rise to the photorefractive effect (PRE) (also called optical damage) is known to have been observed in the non-centrosymmetric ferroelectric single crystals such as $LiNbO_3$ (Ashkin *et al* 1966; Chen 1967) and some pyroelectric oxides of the ABO_3 type (Johnston (1970)). This effect is detrimental to the use of these crystals as electro-optic and acousto-optic modulators and second harmonic generators. On the positive side, this phenomenon is of great current interest due to its applicability to high speed, erasable volume of holographic memory.

The influence of magnetic field on the magnitude of the photorefractive effect was detected by Pogosyan *et al* (1982). In this reference, for photorefraction measurements a constant field of 6 kGauss was only employed and hence the effect remains yet to be properly investigated. In the present paper, we report the results of our study on the changes in the birefringence, $\delta\Delta n$, in $LiNbO_3$ using various values of external magnetic field. Also, the changes of Δn with different impurities in the $LiNbO_3$ host lattice are presented.

2. Experimental

The crystals were grown by the Czochralski technique from Fe and Mn-doped melts. The incorporated quantity of Fe or Mn in the host lattice was about 0·05 wt %. The crystals examined were Y-cut plates of 1·22 and 0·98 mm thickness for Fe and Mn-doped $LiNbO_3$ crystals, respectively. The radiation from the LU 832A type He-Ne laser tube ($\lambda = 0{\cdot}63\ \mu$; output power 2 mW) was focussed on to the sample by a spherical lens of focal length 16 cm which produced an illuminated spot of dimension

$1{\cdot}8 \times 10^{-3}$ mm^2. The magnetic field was produced by an electromagnet and could be varied in steps from 0 to 15 kGauss by varying the current from a regulated rectifier to pass through the coil. The magnetic field was reversed by reversing the flow of current through the electromagnet so that the North and South poles got interchanged. The whole experimental arrangement was set in a semi-dark room at a temperature of about 28°C; the set-up is shown in figure 1 which is self-explanatory and the optics of the scheme is quite simple (Chen 1969).

The specimen under test is properly positioned between the two pole pieces with its optic axis parallel to the Babinet-Soleil compensator CP and then subjected to optical damage by the incident laser beam whose direction of propagation is normal to the crystal c- axis. For measurements by the usual polarisation-optical method (Levanyuk and Osipov 1975), the maxima and minima readings on the compensator are determined before (with the diffuser F inserted to allow a feeble-intensity beam $\approx 0{\cdot}01$ mW cm^{-2}) and thenafter (with the diffuser F removed from the path to allow the fully intense beam to be incident on the crystal for about 20 minutes to induce index change) damage to give the path difference

$$X = \frac{\min_{BD} \sim \min_{AD}}{\max_{BD} \sim \max_{AD}}, \tag{1}$$

where the subscripts BD and AD signify before damage and after damage, respectively. The change in birefringence is computed using the relation:

$$\pi X = 2\pi d \delta \Delta n / \lambda, \tag{2}$$

where d is the thickness of the sample and λ is the wavelength involved. It may be noted that during sample exposure by the linearly polarised light, with its plane of polarisation at 45° from the c-axis of the sample, the transmitted light was split into three beams, displaced from each other along the c-axis. The central beam, which was not displaced, was an ordinary ray while the two outer beams were extraordinary rays. This revealed the fact that the optically induced changes diminished the extraordinary index of refraction n_e the most and the ordinary index n_o the least. Since n_o was little affected, $\delta\Delta n$ was approximately the same as the change in n_e, *i.e.* δn_e.

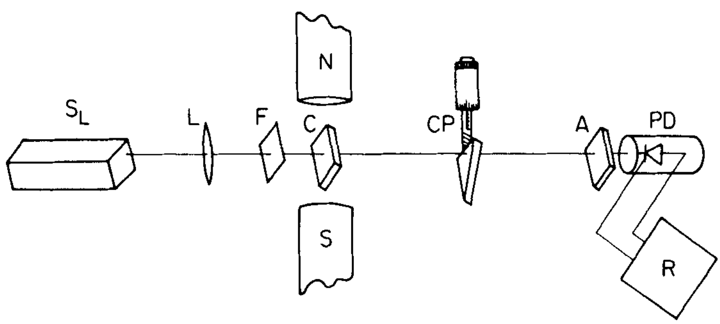

Figure 1. Schematic diagram of the experimental set-up. S_L—laser source; L—focussing lens; F—diffuser; C—crystal; NS—pole pieces of the electromagnet; CP—compensating plate; A—analyser; PD—photodiode; R—XY-recorder.

3. Observation and discussion

The change in birefringence $\delta\Delta n$ was first probed in the absence of the magnetic field as mentioned above. It was found that part of the induced change faded within a few hours but the remaining part, referred to as the integrating component (Ashkin 1966; Chen 1969), stayed unaltered for several days until the crystal was heated. Therefore, to erase the induced index change, the crystal was annealed at a constant temperature of 200°C for about 15 min. This annealed crystal was again properly positioned and the magnetic field B switched on, and thus $\delta\Delta n$ was determined for different known values of B. The observed experimental dependence of $\delta\Delta n$ on B is shown in figures 2 and 3 for $LiNbO_3$: Fe and $LiNbO_3$: Mn respectively. Both the curves reveal a characteristic decrease in $\delta\Delta n$ with increase in applied magnetic field. The rate of decrease, however, is different for the two samples in the examined range of 0–15 kGauss of the field. In the

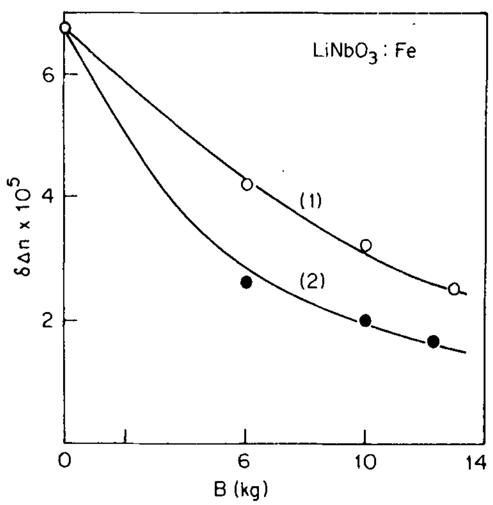

Figure 2. Plot of $\delta\Delta n$ as a function of B for $LiNbO_3$: Fe sample. Curve 1—direct field, Curve 2—reversed field.

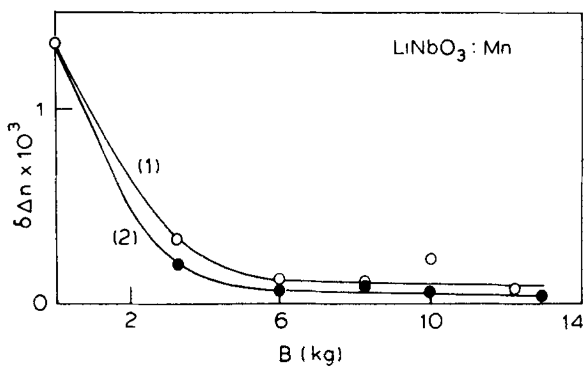

Figure 3. Plot of $\delta\Delta n$ as a function of B for $LiNbO_3$: Mn sample. Curve 1—direct field, Curve 2—reversed field.

Mn doped sample, a saturation is achieved around 3 kGauss; however, such an effect is not observed for Fe doped sample even up to 15 kGauss. Further, the fact that the magnitude of $\delta\Delta n$ for the Fe-doped lattice is 2 orders greater than that for the Mn-doped sample is perhaps due to radical difference in photovoltaic currents. This, of course, needs to be examined in depth.

The characteristic difference in the behaviour of $\delta\Delta n$ *vs* B curves in figures 2 and 3 can be attributed to the fact that the mechanism of PRE (Glass *et al* 1974) is closely connected with photovoltaic effect. It is suggested that the electrons excited by the incident laser beam will drift through the irradiated region of the crystal not due to the inherent internal field (Chen 1969; Yasojima 1973) but due to a bulk photovoltaic effect (BPVE) originating from an asymmetry of the transition probability in the carrier excitation process. The distance from the impurity Fe or Mn atoms to neighbouring Nb atoms in the $+c$-direction of doped $LiNbO_3$ matrix is different from that in the $-c$ direction. Consequently, the probabilities of charge transfer by optical transition from Fe^{2+} or Mn^{2+} to Nb^{5+} (conduction band) in positive and negative directions differ, implying the appearance of net photovoltaic effect. It may be mentioned that this mechanism of BPVE to influence PRE has been well established in $LiNbO_3$: Fe (Levanyuk *et al* 1980; Pogosyan *et al* 1980; Fridkin and Popov 1978).

Based on close link between BPVE and PRE, the essential dependence of $\delta\Delta n$ on the illumination intensity can be given by (Levanyuk *et al* 1980; Glass *et al* 1974),

$$\delta\Delta n = r_{ij}\frac{Q\phi}{\sigma_{\text{ph}}+\sigma_d}, \tag{3}$$

where r_{ij} is the effective electro-optic coefficient, $Q(=k\alpha$, k being termed the Glass constant and α the absorption coefficient) the photovoltaic constant which is probably most sensitive to the nature of the dopant, since it is calculated by the magnitude of photocurrent which is different for different lattice compositions, the product $Q\phi$ being the photovoltaic current J_{ph}, ϕ the illumination intensity, σ_{ph} the photoconductivity and σ_d the dark conductivity. Evidently from (3), if one postulates Q to decrease more rapidly than $\sigma_{\text{ph}}(\sigma_d \approx 10^{-14}$ is, of course, independent of the magnetic field, as it depends on only the kind of sample), the continuous decrease of $\delta\Delta n$ with increasing B in Fe doped $LiNbO_3$ can be easily understood. In fact, for $LiNbO_3$: Fe the ratio Q/σ_{ph} is continuously falling for higher values of B and consequently, the photorefraction is closely connected with the photovoltaic effect. For $LiNbO_3$: Mn, the magnetic field below 3 kGauss shows the relationship between photovoltaics and photorefraction. But, for higher magnetic fields, the change of Q tends to be compensated by the changes in σ_{ph} so that the ratio Q/σ_{ph} remains unaffected. Clearly then, the BPVE fails to account fully for PRE of the crystal. There can be found experimental data in literature (Volk *et al* 1977; Pashkov *et al* 1979; Levanyuk *et al* 1981) which disprove the existence of BPVE and which bring in additional nonphotovoltaic contribution to PRE. Possibly, the crystal structure change, in particular the photoinduced change in spontaneous polarisation (Levanyuk 1976) in the presence of an external magnetic field, influences much the nature of photorefraction in $LiNbO_3$: Mn, in which case the dipole moment is saturated and does not exhibit dependence on light illumination. Support to this mechanism of non-field (polarisation) contribution directly to optical distortion or PRE will follow from further work in this direction, using an elliptical illuminated spot and more elaborate and precise instrumentation.

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