# THE PHOTOREFRACTIVE RESPONSE OF $LiNbO_3$:Fe:Mn CRYSTAL DEPENDING ON ELECTRICAL PROPERTIES OF ITS SURROUNDINGS

Norbert TARJÁNYI
Department of Physics, Faculty of Electrical Engineering,
University of Žilina, Univerzitná 1, 010 26 Žilina, Slovac Republic, tel.+421 41 513 2368, e-mail: tarjanyi@fyzika.uniza.sk

## ABSTRACT

*The results of experimental investigation of the impact of ambient surroundings on refractive index inhomogeneity induced in $LiNbO_3$:Fe:Mn crystal are presented in the article. The effect of dielectric (ambient air, paraffin oil) and electrically conducting (saturated aqueous solution of $CaCl_2$) media surrounding the sample was investigated. The refractive index inhomogeneity in the sample was created due to photorefractive effect and monitored in real time by means of Mach-Zehnder interferometer. The time dependences of the amplitude of the induced refractive index changes were obtained from captured interferograms. The analysis of the obtained time dependences shows that the temporal behaviour of the inhomogeneity during and after its formation as well as the spatial distribution of the refractive index within the inhomogeneity depends significantly on electric properties of a medium surrounding the crystal sample.*

***Keywords:*** *photorefractivity, lithium niobate, interferometry*

## 1. INTRODUCTION

In 1966 Ashkin *et al*. observed optically induced refractive index change in lithium niobate crystals ($LiNbO_3$) for the first time [1]. Since then $LiNbO_3$ has become one of the best known photorefractive crystals and showed great potential for various applications based on desired and controlled light-induced refractive index change of the material [e.g. 2-4]. Despite the many years of intense research $LiNbO_3$ crystals still offer a vast field for studying the light-matter interaction and related phenomena. In 80's it was observed that above some threshold intensity of light incident on the iron doped $LiNbO_3$ crystal spontaneous and quasi periodic jump-like decrease of the refractive index change occurred in the investigated samples [5]. Such behaviour was attributed to charge carriers recombination at or near the surface of the crystal originated as consequence of the electric field in the bulk material formed due to its illumination, but no further theoretical treatment confirming the suggestion was performed. The influence of the charges present in the environment surrounding the crystal sample on the refractive index inhomogeneity within the sample was experimentally showed and numerically confirmed for static case in [6].

This article deals with investigation of the influence of the electrical properties of the ambient surroundings on the temporal behaviour of the photorefractive inhomogeneity induced in a $LiNbO_3$:Fe:Mn sample in which also the jump-like decrease of the amplitude of the refractive index change was observed if sample was put into dielectric medium. The refractive index changes are observed by means of Mach-Zehnder interferometer.

## 2. BASIC CONCEPT OF PHOTOREFRACTIVITY

The explanation of the photorefractive effect valid for most of the observed photorefractive phenomena in crystals can be introduced by the so called 'standard band-transport model' [7]. The model assumes the photorefractive material contains certain kinds of impurities or defects which give rise to allowed energy states also within band gap of the material. Some of these states may be occupied by electrons while some of them remain empty, primarily acting as traps. The occupied (empty) states can be ionized if electrons in the allowed states (valence band) absorb photons with proper energy. Electrons (holes) are generated into conduction (valence) band where they can move due to diffusion, photogalvanic effect [8] or externally applied electric field and then become captured by ionized states or empty traps. The charge transport mechanisms and recombination of free electrons (holes) with ionized states or traps lead to spatial redistribution of the charge carriers and consequently give rise to the local electric field. This field changes the refractive index via the Pockel's effect. Taking into account the symmetry of the $LiNbO_3$ crystal (*3m*) the refractive index of the beam polarized, e.g. parallel with optical axis, will be

$$n_e = n_{e0} + \frac{1}{2} n_{e0}^3 r_{zz} E_z , \tag{1}$$

where $n_e$ and $n_{e0}$ are the extraordinary refractive indices of the crystal in presence of the electric field and when the electric field is absent, respectively and $r_{zz}$ is the involved electro-optic coefficient.

## 3. EXPERIMENT

The influence of the ambient surroundings on temporal behaviour of the photorefractive inhomogeneity was experimentally investigated for $LiNbO_3$:Fe:Mn sample (*y*-cut with 0.075% Fe and 0.025% Mn) provided by Photox, Ltd.. The both *y* surfaces of the crystal sample were polished and dimensions of the sample were $x \times y \times z$ = $10mm \times 1mm \times 10mm$. All experiments were performed at room temperature.

The sample was put into a glassy cuvette and placed on to a holder situated in one arm of Mach-Zehnder interferometer (Fig. 1).

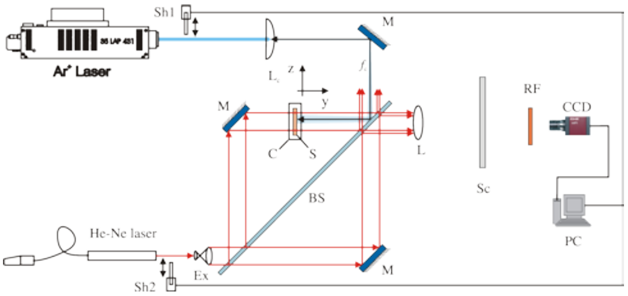


**Fig. 1** Experimental setup: Ar laser, $L_c$ - cylindrical lens, Mach-Zehnder interferometer with sample (BS – beam splitter, M – mirror, C – cuvette, S – sample, L - objective), CCD – CCD camera, Sc – screen, RF – red filter, Sh1 and Sh2 – electromechanical shutters, PC – computer, $f_c$ – focal length

The refractive index inhomogeneity in the sample was induced by $Ar^+$ laser beam with wavelength 488*nm* and vertical linear polarization. The laser beam was modified by a cylindrical lens. Gaussian radii of the modified laser beam measured in the place of the sample seating were $R_z = 0.325$*mm* and $R_x = 2.90$*mm* (Fig. 2).

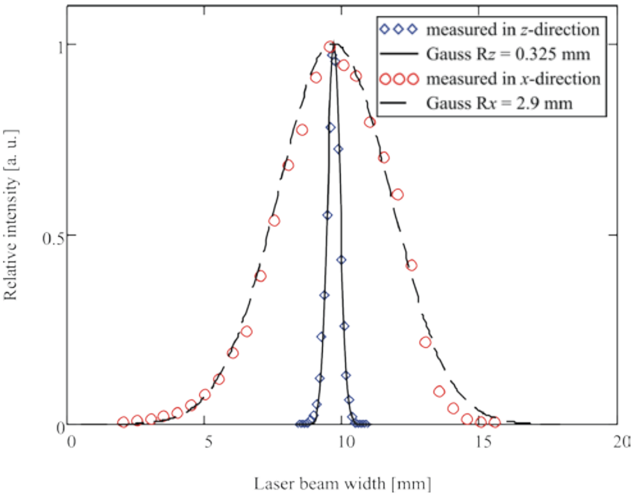


**Fig. 2** Intensity distribution in the middle of the laser beam measured in *z*- and *x*-directions

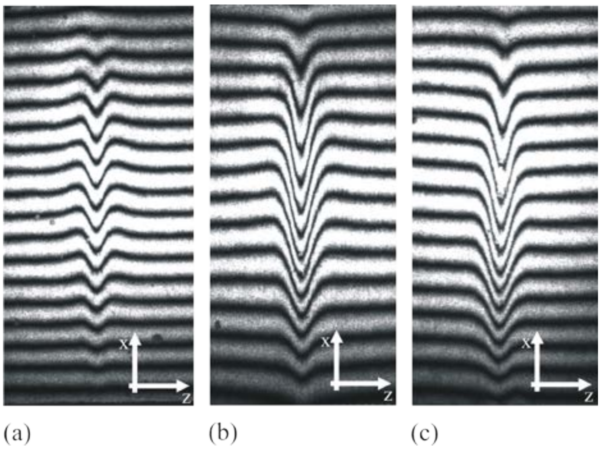
A beam from He-Ne laser (wavelength 633*nm*, horizontal linear polarization) expanded by 20 times illuminated the crystal in order to display the photorefractive inhomogeneity by means of interference imaging [9]. The interference field of the object and the reference waves was imaged by an objective put at the output of the interferometer. A red filter put between interferometer's output and the screen avoided the blue light from $Ar^+$ laser reflecting from the front and back surfaces of the sample its incidence on a projecting screen. A computer controlled CCD camera monitored the evolution of the interferogram carrying information on space-time distribution of the refractive index in the inhomogeneity. Two electromechanical shutters allowed controlling the conditions of the sample's illumination.

The created refractive index inhomogeneity was usually erased by heating the sample in an oven (air atmosphere) at 110 °C for 2 hours.

## 3.1. SPATIAL DISTRIBUTION OF THE REFRACTIVE INDEX WITHIN INHOMOGENEITY

The influence of the ambient surroundings on the light-induced refractive index change in $LiNbO_3$:Fe:Mn sample was investigated for two electrically distinguished cases: conducting environment (saturated aqueous solution of $CaCl_2$, $n = 1.4495$ at 20 ºC) and dielectric one (ambient air and paraffin oil, $n = 1.4670$ at 21 ºC). In all three cases the sample was put in the cuvette, placed in the interferometer's arm and illuminated by light from $Ar^+$ laser. The intensities of light in the sample were 27.9$mWmm^{-2}$ for ambient air as the surrounding, 32.3$mWmm^{-2}$ in case the sample was immersed in $CaCl_2$ solution and 33.8$mWmm^{-2}$ when sample was dipped into oil. The temporal evolution of the photorefractive inhomogeneity was observed by means of Mach-Zehnder interferometer which produced the interference pattern and imaged it by an objective on the screen. The camera captured the interference pattern every second while the sample was illuminated by $Ar^+$ laser beam. The beam illuminating the sample was blocked by means of electromechanical shutter 300 seconds after commencement of the recording. Simultaneously, the beam from He-Ne laser was also blocked out in order to reduce any damaging influence of the light from He-Ne laser on the induced refractive index inhomogeneity.

The impact of the surrounding medium on the induced inhomogeneity can be seen via the spatial distribution of the refractive index. The interferograms in Fig. 3 show the spatial distribution of the refractive index in the inhomogeneity after almost the same exposure.



(a) (b) (c)

**Fig. 3** Spatial distribution of the refractive index in the $LiNbO_3$:Fe:Mn sample placed in: (a) aqueous solution of $CaCl_2$, $t = 52s$; (b) air, $t = 60s$ and (c) paraffin oil, $t = 50$s

## 3.2. TIME DEPENDENCE OF THE AMPLITUDE OF THE REFRACTIVE INDEX CHANGE WITHIN INHOMOGENEITY

The refractive index change within inhomogeneity was evaluated using a program processing the interferograms made in Mathcad 14 software environment. The developed program estimates the value of the amplitude of the refractive index change with an error $2\times10^{-5}$. The temporal behaviour of the amplitude of the refractive index change during illumination of the sample can be seen in Fig. 4a for three investigated cases. Obviously, a notable influence of the medium surrounding the sample on the refractive index change during illumination of the sample can be seen from measured temporal dependences. In case of electrically conducting ambient medium the amplitude of the refractive index change reached after 300 seconds of applying the illumination is the lowest one compared to cases of the dielectric ambient surroundings (Fig. 4a). The measured data can be clearly fitted by exponential time dependence as it implies from the standard band-transport model [7].

It is known that in $LiNbO_3$ crystals the life-time of the light-induced refractive index inhomogeneity can be in the range from several months to several years [e.g. 10] depending on storage conditions of the crystal. The critical parameters of the environment having direct and cardinal impact on the record's life-time are the temperature and light ranging from UV to middle-VIS region. However, the physical properties of the crystal itself play some role, too. Mainly, it is the level of doping which influences the dark conductivity (caused by mechanisms of tunnelling) of the crystal which, in turn, causes the decay of the refractive index inhomogeneity. It can be seen in Fig. 4b that if the conducting medium is the sample's surroundings the amplitude of the refractive index change reached in the sample after 300 seconds does not change in time after the illumination is turned off. If air and paraffin oil are the ambient surroundings a slow decrease of the amplitude of the refractive index change with time is observed. Moreover, also these two cases of dielectric media are not equivalent. The spontaneous decay of the amplitude of the refractive index inhomogeneity seems to be mono-exponential in case of air as ambient surroundings while two-exponential if the sample is put into paraffin oil.

Since the life-time of the photorefractive inhomogeneity created in the crystal is expected to be at room temperature several months at least, the significant decay of the amplitude of the refractive index change observable practically within hours cannot be attributed to dark conductivity responsible for spontaneous erasure of the inhomogeneity. The observed temporal behaviour of the photorefractive inhomogeneity's decay when sample is put into dielectric media indicates there is a significant influence of polarization charge originated due to light-induced electric field in the sample.

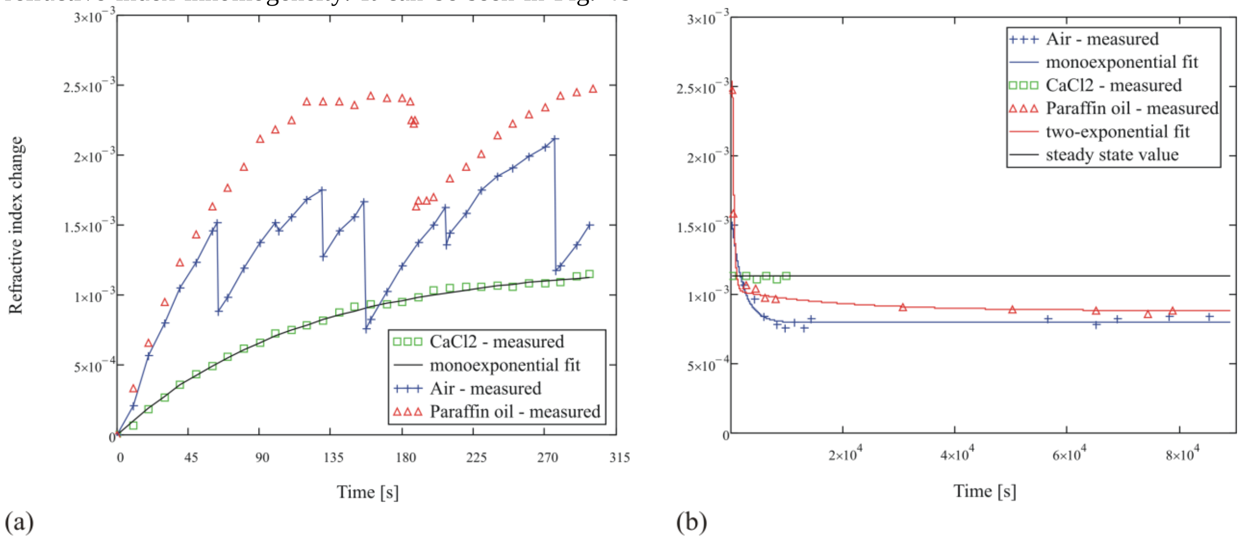


**Fig. 4** a) Temporal evolution of the light-induced refractive index change in $LiNbO_3$:Fe:Mn sample measured for three different media surrounding the sample during recording. b) Temporal evolution of the light-induced refractive index change in $LiNbO_3$:Fe:Mn sample measured in dark for three different media surrounding the sample

If the sample was surrounded by the ambient air or paraffin oil, spontaneous jump-like change of the refractive index of the sample was observed (Fig. 4a and Fig. 5). In case of air as ambient surroundings the jumps, which were observed in the whole illuminated area of the sample at once, showed some quasi-periodicity within the time of investigation. The amplitudes of the single abrupt refractive index changes were not the same. A similar behaviour of the refractive index inhomogeneity was also observed when paraffin oil was used as the medium surrounding the sample. The difference was that several local and small in amplitude jumps preceded the bigger one, which occurred around 188*s* after commencement of the sample's illumination and was only one well identified big change of the refractive index within the time of investigation. The small local jumps occurred in the range between 180*s* and 188*s* and they are responsible for slight decrease of the amplitude of the refractive index change within this time interval.

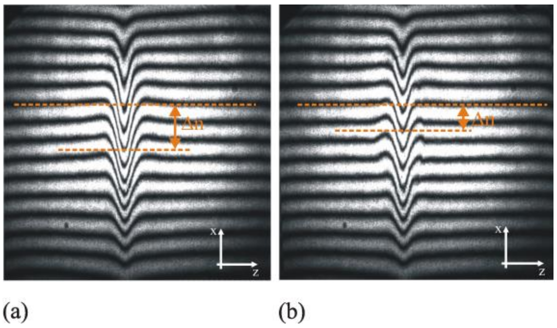


**Fig. 5** Interferograms taken at 63*s* (a) and 64*s* (b) showing the sudden change of the refractive index during recording in $LiNbO_3$:Fe:Mn sample surrounded by air

While recording the inhomogeneity a slight ripple of the interference pattern was observed before the jumps started occurring and the ripple vanished just after the jump. The ripple of the interference pattern was not observed when ambient air was the sample's surroundings. Such deformation of the interference fringes means that a change of optical properties of the paraffin oil occurs. The change might be due to a local change of the oil's temperature probably caused by the absorption of light in the crystal which, in turn, heats the surrounding oil due to heat exchange. However, the change of optical properties of the oil observed during illumination might be also due to interaction of the space charge field in the sample and the charges in the sample's immediate surroundings.

## 4. CONCLUSIONS

In the paper there are presented results of investigation of the influence of the medium surrounding $LiNbO_3$:Fe:Mn sample on temporal behaviour of the photorefractive inhomogeneity. The influence of electrically conducting and dielectric surroundings was investigated. The results are that the influence of the electric properties of the medium in which $LiNbO_3$ is situated on photorefractive inhomogeneity is significant during recording as well as in case the sample with inhomogeneity is in the dark. The sudden decrease of the refractive index was observed during recording if sample was in the air or dipped into paraffin oil. No such decrease was observed for sample immersed into conducting liquid. Also spatial distribution as well as the amplitude of the refractive index change reached during recording differs depending on the surrounding medium. The obtained knowledge on temporal behaviour of the photorefractive inhomogeneity induced in $LiNbO_3$:Fe:Mn crystals can be used for definition of the storage conditions for holograms in holographic memory devices based on photorefractive crystals and for studying the processes connected to dielectric breakdown in oils and oil-based liquids as well.

## ACKNOWLEDGMENTS

This work was partly supported by the Slovak National Grant Agency Nos. VEGA 1/0528/12 and 1/1058/11. The support of the R&D operational program Centre of Excellence of power electronics systems and materials for their components II. No. OPVaV-2009/2.1/02-SORO, ITMS 26220120046 funded by European regional development fund (ERDF) is also acknowledged.

## REFERENCES

[1] ASHKIN, A. et al.: Optically-induced refractive index inhomogeneities in $LiNbO_3$ and $LiTaO_3$, *Appl. Phys. Lett.*, vol. 9, no. 1, pp. 72–74, 1966.

[2] SUN, Q. et al.: Recognition of color patterns using a photorefractive $LiNbO_3$: Fe crystal, *Opt. Commun.*, vol. 157, no. 1-6, pp. 23–26, 1998.

[3] WOOTEN, E. L. et al.: A Review of Lithium Niobate Modulators for Fiber-Optic Communications Systems, *IEEE J. Sel. Top. Quantum Electron.*, vol. 6, no. 1, pp. 69–82, 2000.

[4] ZAJTSEV, A. K. – LIN, S. H. – KEN, Y. H.: Sidelobe suppression of spectral response in holographic optical filter, *Opt. Commun.*, vol. 190, no. 1-6, pp. 103–108, 2001.

[5] AUGUSTOV, P. A. – SHVARTS, K. K.: Surface Recombination and Photorefraction in $LiNbO_3$-Fe Crystals, *Appl. Phys. A*, vol. 18, no. 4, pp. 399–401, 1979.

[6] TARJÁNYI, N. – TUREK, I.: Influence of surroundings on photorefractive effect in lithium niobate crystals, *Physica B*, vol. 407, pp. 4347-4353, 2012.

[7] YEH, P.: *Introduction to photorefractive nonlinear optics*, New York: John Wiley & Sons, Inc.1993, pp. 82–114.

[8] HEYSZENAU, H.: Electron transport in the bulk photovoltaic effect, *Phys. Rev. B*, vol. 18, no. 4, pp. 1586–1592, 1978.

[9] TUREK, I. – TARJÁNYI, N.: Interference imaging of photorefractive record in thin sample of $LiNbO_3$ crystal, 14th Slovak-Czech-Polish Conference on Wave and Quantum Aspects of Contemporary Optics, Nitra, Slovakia, Proc. SPIE, vol. 5945, pp. 59450J-1–59450J-6 , Sept. 2004.

[10] NEE, I. – MÜLLER, M. – BUSE, K. – KRÄTZIG, E.: Role of iron in lithium-niobate crystals for the dark-storage time of holograms, *J. Appl. Phys.*, vol. 88, no. 7, pp. 4282–4286, 2000.

Received November 27, 2012 , accepted January 15, 2013

## BIOGRAPHY

**Norbert Tarjányi** was born on 26.5.1976. In 1999 he graduated (MSc) at the department of Telecommunications of the Faculty of Electrical Engineering at University of Žilina. He defended his PhD in the field of Physics of Condensed Matter and Acoustics in 2004. Since 2002 he is working as a teacher with the Department of Physics. His scientific research is focusing on studying light-matter interaction and questions related to applications of the records of structured optical fields made in photorefractive $LiNbO_3$, photopolymers, photoresists and magnetic fluids.