PHD THESISES

Z-scan study of the nonlinear optical properties of $LiNbO_3$

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I. SCIENTIFIC BACKGROUND, AIMS

The interest on the $LiNbO_3$ (LN) as a nonlinear optical material has arisen in the recent years. One of the reasons is that it was shown the material can be periodically poled. This means that in the crystal a periodical domain structure can be created in the micrometer range by growth or by posterior poling. Such crystals are called periodically poled (PPLN) crystals. The LN crystal is used for various applications, one of these fields is the application in optical frequency-converters. The efficiency of the conversion is determined partly by the effective value of the nonlinear optical tensor. In case of PPLN crystal we can effectuate the quasi-phase matching technique, which has the advantage contrary to the conventional geometrical phase-matching that it makes possible to utilise the largest d_{33} element of the nonlinear optical tensor.

The second reason, which explains the boom is the good quality stoichiometric crystals were successfully produces lately. The stoichiometric LN has the advantage contrary to the congruent that in case of this composition the coercitive force is lower with two orders of magnitude. Because of this it is enough to use a significantly weaker poling field to generate the periodic structure. This makes the poling process easier and more accurate and also makes possible to produce thicker PPLN crystals.

LN has a disadvantageous property from the point of the frequency conversion, namely the photorefraction. Due to this in case of undoped LN for relative low intensities significant distortion of the beams can be observed. In course of application in frequency-converters any effect resulting beam distortions have to be avoided. From the literature it is known, that doping by Mg the photorefraction can be significantly reduced. Our long-term aim is the domestic production of PPLN crystals for application in frequency-converters. This implicates the study of the light-induced changes of refraction in LN. During my PhD studies I participated in a project where I examined the light induced change of refraction, and its origin in pure, Fe impured, and Mg doped both in stoichiometric and congruent LN

The experimental results of the dissertation were achieved at the Department of Experimental Physics, University of Pécs and at the Research Institute for Solid State Physics and Optics of the Hungarian Academy of Sciences.

II. MEASUREMENT METHODS

In 1990 M. SHEIK-BAHAE established a measurement method with the purpose of determining the nonlinear refraction of thin samples. This is called the *Z*-scan technique, since the nonlinear sample is scanned along a focused Gaussian-beam through its focal plane. The nonlinear refraction of the sample can be obtained by monitoring the far-field on-axis intensity of the beam versus the sample position.

For the measurements the Gaussian-beam was supported by an all lines visible Ar-ion laser with maximum power of 2 W. The detector placed at the far-field to examine the middle of the beam was a photodiode, the other detector, which measured the total beam power was a thermic detector. For scanning a manual or a piezoelectric translator was used.

I completed the *Z*-scan measurements with the detailed analysis of cross section of the beam passing through the sample. For this a CCD or a digital camera was used. The temporal evolution of the beam distortions were also examined, for this – depending on the time constant – an oscilloscope was used.

The theoretical calculations and fittings were performed with the help of the MathCad software.

III. NEW SCIENTIFIC RESULTS

1. I demonstrated that in order to decide the applicability of the *Z*-scan technique as a qualitative method some accessory measurements have to be performed, mainly in those cases, when the origin of the light-induced change of refraction was not clarified. One of these measurements is the detailed examination of the cross section of the beam passing through the sample. Concerning to the $LiNbO_3$ (LN) crystal, the beam cross-section was examined by digital recordings. In the case of not significant distortions the characteristics of the variation was clarified by the detailed analysis of the intensity-distribution. In case of the 5.0 mol% of Mg doped stoichiometric LN the beam cross-section was isotropic, while in case of the congruent composition doped on the same Mg level it was found to be elongated into the direction of the crystal-optic axis. From the *Z*-scan curves in the case of 5.0 mol% Mg doped congruent crystal I concluded for positive, and in the case of 5.0 mol% Mg doped congruent for negative, but with one order of magnitude larger nonlinearity than the stoichiometric. [1]

2. While in case of the 5.0 mol% Mg doped congruent LN the beam distortion became significant only at intensities significantly larger than 1 kW/cm^2, in case of the undoped and Fe-doped samples, this was observed even for the 1 W/cm^2 intensity as well. I compared the photorefractive sensitivities of the photorefractive LN samples by examination of the temporal evolution of the on-axis intensity in the far field. From the time constants of the transient and from the steady-state normalised transmittance values, I concluded that the undoped stoichiometric sample was more sensitive than the undoped, and even more sensitive than the Fe-doped congruent LN. Furthermore it was

diagnosed, that the photorefractive sensitivity of the Fe-doped samples is increasing with the Fe concentration. [3]

3. From the peak-valley configuration of the *Z*-scan traces, from the circular symmetric beam variations, and from the identical results of the scanning with different velocities it was concluded, that in case of the 0.67 and 5.0 mol% Mg doped stoichiometric, and in case of the 6.1 mol% Mg doped congruent LN photorefraction was negligible compared to other effects. These samples showed the decrease of the transmittance in prefocal, and the increase in postfocal positions on the ms time-scale. From the isotropic distortions, from the positive sign of the n_2 nonlinear refraction determined from the *Z*-scan fittings and from the result of the theoretical calculation of the τ time constant of the transient I concluded to the thermo-optical origin of the effect. By complex measurements I showed that for the congruent composition the photorefractive threshold was between 5.0 and 6.1 mol% Mg concentration in contrast with the 4.6 mol% value given in the references. For the stoichiometric composition the threshold was below 0.67 mol%. [2][3]

4. In order to clarify the origin of the thermo-optical effect for the non-photorefractive samples I performed measurements at different powers. It was shown from the relation of the beam-power, the n_2 nonlinear refraction, the β nonlinear absorption coefficient, that in the thermo-optical effect the nonlinear absorption has dominant role. I supported it by the theoretical calculation of n_2 as well. The adequate fitting of the traces also refer to this. For further improvements I performed scannings at a given power, applying polarisation to be parallel, and perpendicular to the crystal-optic axis of the

given sample. For the extraordinary polarisation the n_2 value obtained from the fitting was found to be about three times larger than the n_2 which belongs to the ordinary polarisation. The ratio of the β values belonging to the different polarisation was found to be identical with the n_2 ratio. These measurements strongly proved the significance of the nonlinear absorption in the formation of the nonlinear refraction. I examined the nonlinear absorption by open-aperture measurements as well, whose results were harmonious with the closed-aperture measurements. These measurements also demonstrated that the nonlinear absorption in LN was anisotrope. [2][3]

5. I showed, that if the light-induced change of refraction had thermal origin, and the absorption was purely nonlinear, the standard *Z*-scan theories were valid to determine the n_2 and β material parameters. But if the absorption is purely linear, or both the linear and nonlinear is presented, the thermo-optical nonlinear refraction depends on the actual beam size as well. This makes the standard theory impossible for the fitting. A model based on numerical calculations was elaborated which is suitable to determine the coefficients of linear and nonlinear absorption for thermal effects.

IV. USAGE OF THE RESULTS

The long-term aim of my measurements is to find the most perspective LN composition for periodic poling, and for the application in optical frequency converters. An adjacent aim – which has already been achieved – was to clarify the origin of the light induced change of refraction in LN, and to characterise it quantitatively in the possible cases. As the result of my complete measurements samples above threshold (absent of any photorefraction) are found to be the most perspective, since during the measurements – performed under the same circumstances – these samples showed the lowest optical damage, furthermore the coercitive force is lower than the congruent LN. The sensitivity of the stoichiometric crystal is increasing by the Mg dopant concentration, however the incorporated amount of dopant may lead to the change of the physical properties of the material. Among the samples I used, the 5.0 mol% of Mg doped stoichiometric LN was found to be the most perspective.

In my 5th thesis point I deal with the extension of the *Z*-scan theory to the thermo-optical effect. Since there is no references related to this thesis, my distinct aim is to publish these results in the near-future.

V. SCIENTIFIC PUBLICATIONS CONCERNING TO THE TOPIC OF THE DISSERTATION

In international refereed journals

1. **L. Pálfalvi**, G. Almási, J. Hebling, Á. Péter, and K. Polgár:
Measurement of laser-induced refractive index changes of Mg-doped congruent and stoichiometric $LiNbO_3$
Appl. Phys. Lett. **80** 2245 (2002)

2. **L. Pálfalvi**, J. Hebling, G. Almási, Á. Péter, K. Polgár:
Refractive index changes in Mg doped $LiNbO_3$ caused by photorefraction and thermal effect
Journal of Optics A (2003, közlésre elfogadva)

3. **L. Pálfalvi**, J. Hebling, G. Almási, Á. Péter, K. Polgár, K. Lengyel, R. Szipőcs:
Nonlinear refraction and absorption of Mg doped stoichiometric and congruent $LiNbO_3$
Journal of Appl. Phys. (beküldve, 2003)

4. X. Zhang, J. Hebling, J. Kuhl, W.W. Rühle, **L. Pálfalvi**, H. Giessen:
Femtosecond near-IR optical parametric oscillator with efficient intracavity generation of visible light
J. Opt. Soc. Am. B **19**, 2479 (2002)

Conference performances

5. **L. Pálfalvi**, G. Almási, J. Hebling, Á. Péter, and K. Polgár:
Quantitative determination of photorefraction in Mg-doped congruent and stoichiometric $LiNbO_3$
CLEO 2002, Long Beach, California, USA, May 19-24, 2002, CThD1, 447

6. **L. Pálfalvi**, G. Almási, J. Hebling, Á. Péter, and K. Polgár:
Quantitative determination of photorefraction in Mg-doped congruent and stoichiometric $LiNbO_3$
19^{th} Congress ICO, Optics for the Quality of Life, Florance, Italy, August 25-30, 2002, 907

Poster presentations

7. **L. Pálfalvi**, J. Hebling, Á. Péter, K. Polgár:
New method for the characterisation of photorefractive damage in $LiNbO_3$ crystals
Fourth Annual Meeting of the COST Action P2 Applications of Nonlinear of Nonlinear Optical Phenomena and Workshop in $LiNbO_3$ (2001)

8. **Pálfalvi L**., Péter Á., Polgár K., Hebling J.:
Examination of photorefraction of $LiNbO_3$ *crystals*
IV. Szimpózium a Hazai Kvantumelektronikai Kutatásokról. Kvantumelektronika (2000)

9. **Pálfalvi L**., Hebling J:
Femtosecond PPLN optical parametric oscillator
IV. Szimpózium a Hazai Kvantumelektronikai Kutatásokról. Kvantumelektronika (2000)

Further publications

10. **Pálfalvi László**:
An unconventional way of measuring moments of inertia
Fizikai Szemle, 2003/4 143-145

11. **Pálfalvi László**:
1994/2 FGY.2787, 1994/5 FN.2758, 1994/10 FF.2798, 1996/8 FF.3017, 2002/3 P.3516, 2002/4 P.3526, 2002/9 P.3574, 2003/1 P.3585, 2003/2 P.3595, 2003/3 P.3604, 2003/4 P.3620, 2003/5 P.3623
Mathematical and Physical Journals for Secondary Schools (publication of problems and solutions)