

**NOVEL METHODS AND
APPLICATIONS OF SPATIAL AND
TEMPORAL INVESTIGATION OF
WAVE-MIXING IN
PHOTOREFRACTIVE CRYSTALS**

PhD Theses

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Preliminaries of the research

1. Photorefractive four-wave mixing

Refractive index inhomogeneity may occur in some optical materials due to inhomogeneous illumination. This phenomenon, named photorefraction, was discovered in LiNbO_3 , LiTaO_3 , BaTiO_3 and KTN crystals by A. Ashkin et al. in 1966 [Ashkin, A. 1966]. The hologram storage and reconstruction experiments of F. S. Chen et al. published in 1968 were also based on lithium niobate [Chen, F. S. 1968]. The essence of the process is macroscopic translation of the photoinduced free electrons due to diffusion, drift and/or the photovoltaic effect, followed by electron recapture [Günter, P. 1989]. Hence, the electron density decreases in more illuminated areas, and increases in the less illuminated ones. The changed density results in the build-up of an electric field that modulates the refractive index of the material via the electrooptic effect. Thus, a phase hologram or, if two-wave mixing is used (two monochromatic plane waves incident on the sample in different angles), a diffraction phase grating can be created.

The interference of two non-parallel, coherent, expanded laser beams produces a regular set of stripes. In a photorefractive material the stripes cause Bragg-diffraction for either of the two laser beams. If the parameters are suitably chosen, an energy transfer may occur between the two beams, so the intensity of one beam can be amplified by the other, just like by a pump beam [Günter, P. 1989].

Four-wave mixing can be realized if a two-wave mixing setup is extended by two further coherent waves so that the additional beams are incident in opposite directions to the original ones. Generally three beams are used as input, the fourth beam works as output. Versatility is a great advantage of the setup, which has been used as a light or picture amplifier, multiplexer or phase conjugating mirror. Nevertheless, no studies of any other group of researchers are known dealing with the build-up and change of the modulation distribution inside the crystal as a function of time. One of my collaborating partners, S. Bugaychuk and her co-authors carried out both analytical and computer simulations under the assumption that the photovoltaic effect is negligible in comparison with diffusion, and light absorption may also be neglected. She has shown that the longitudinal distribution of the modulation of the optical dielectric constant (creating the diffraction grating) can be described by the sine-Gordon differential equation and its stationary solution, the soliton distribution, similarly to the case of wave packages propagating in a nonlinear optical medium [Bugaychuk, S. 2000]. She has also shown that the maximum and its location in the crystal, and also the half-width of the

distribution depend on the intensity ratios of the input beams and on the Γ energy transfer gain factor.

2. Thermal fixing of the photorefractive hologram

The photorefractive grating or hologram that is fundamentally photosensitive because of its reversibility can be fixed by thermal treatment in various crystals [Amodei, J. J. 1971; Staebler, D. L. 1972] as e.g. lithium niobate (LiNbO_3). The thermal fixing of the grating (hologram) takes place by the compensation of the electron charge distribution by the electric field of the thermally activated and displaced ions. After this compensation, the inhomogeneity of the electron distribution is decreased by strong incoherent illumination, and the electric field of the ions "frozen" into their new position overcompensates the field of the electrons. In this manner, the field of the ions will be a negative image of the field of the electrons, will not be sensitive to light, and will last long.

In case of congruently grown LiNbO_3 doped with iron many researchers suppose that the ions participating in compensation are protons (e. g. [Bollmann, W. 1977; Vormann, H. 1981; Schmidt, N. 1989; Cabrera, J. M. 1996; Buse, K. 1997]) originating from OH^- ions present in great numbers in all unreduced crystals grown in air. Nevertheless, protons playing an important role in thermal fixing cannot be considered as generally proven for all compositions (congruent, stoichiometric, pure, Fe- or Mn-doped, etc.). Thermal compensation has been shown to occur also in strongly reduced, practically OH^- -free LiNbO_3 crystals, but was shown to be connected with Li^+ ions [Buse, K. 1997; Jundt, D. H. 1992; Yariv, A. 1996].

In our institute stoichiometric and other off-congruent crystals of very high quality can be grown which opens the possibility for investigating thermal fixing more widely, and to identify the compensating ions in case of different compositions. By infrared spectroscopic studies, my colleagues László Kovács and Krisztián Lengyel determined the thermal relaxation properties of the OH^- vibration bands, in particular the temperature dependence of their relaxation time constant [Kovács, L. 2001; Lengyel, K. 2004]. The dependence followed Arrhenius's law with the various bands yielding similar activation energies.

3. Holographic light scattering

Refractive index holograms (phase holograms) can be produced not only by inhomogeneous illumination. In LiNbO_3 unwanted holograms causing light scattering may arise also as a result of intense coherent homogeneous illumination in a process similar to self-oscillation in an amplifier with positive feedback. It is enough to have any small refractive index inhomogeneity, surface reflection, surface or bulk crystal defect (scratch, inclusion, dust, etc.).

for starting the self-amplifying process. This scattering is called holographic (light) scattering or (light) scattering of parasitic holograms, light induced scattering or beam fanning. (There is no unanimously accepted nomenclature yet). This seemingly undesirable phenomenon still has been used by some researchers to serve science and technics [e. g. *Rupp, R. A. 1986; Van Olfen, U. 1989; Fally, M. 2000; Ellabban, M. A. 2001*].

Objectives

1. Objectives in photorefractive four-wave mixing

One of the aims of my work was to check experimentally the soliton-like spatial stationary distribution that is expected in the modulation of the dielectric constant during four-wave mixing, and the properties of the theoretically deduced connection between beam intensities and the position of the maximum of the soliton. My further object was to study the temporal build-up of the stationary distribution.

The importance of the task is partly to prove an interesting theoretical physical phenomenon, namely the existence of a new kind of soliton. This type of soliton cannot be ranged into any of the known groups: there is no direct relationship either with moving wave packages that do not change their shape, or with self-focusing laser beams with diameters of a few microns.

On the other hand, it gives a chance to practical applications in all-optical signal and data processing, optical switches and optical logical networks. One of the most obvious potential applications is in spatial photomultiplexing. The periodic oscillation can be used e.g. in interferometric devices.

2. Objectives in photorefractive hologram fixing

The aim of my search in the field of thermal hologram fixing was to identify those ions that can be made mobile thermally and are responsible for thermal fixing, and to study the temporal behavior of the fixing process. Available results of infrared spectroscopic studies on nearly stoichiometric lithium niobate [*Kovács, L. 2001*] provided a suitable basis for the identification, since the activation energy values obtained by us definitely correspond to the activation energy of the protons of OH^- ions.

The kind of ions taking part in the thermal hologram fixing is an important aspect in the description of this process, as well as the question about the crystallographic centers they are connected with. Having this

information it becomes possible to create an exact model. For this reason I considered the field of hologram fixing to be an important and timely subject.

3. Objectives in holographic light scattering

One of the important characteristics of the materials suitable for thermal hologram fixing is the activation energy of the ions taking part in the compensation of the field of the electrons. This energy mainly depends on the ion itself, but its value can be altered by the bonding status and the crystallographic environment of the ion. The determination of the activation energy is always needed if a new photorefractive material is considered for application, or in the case of searching for a material with optimized composition for a definite purpose. For such a task I suggest a novel experimental method that applies the holographic light scattering phenomenon. The method sets much less requirements for the experimental setup, compared with the standard one based on two-wave mixing. It requires neither a vibration damped and isolated optical table, nor a laser with large coherent length, nor extremely rigid mirrors, filters, sample holders, etc. Between its few disadvantages, one has to note the somewhat reduced accuracy and sensitivity, and the resulting more limited applicability range.

The aim of the study was to develop the method, and to test it using some materials with different composition, comparing the results with data received with other methods. These methods were the standard two-wave mixing method, and the DC electric conductivity measurement.

Using the novel method may facilitate the task of engineers and researchers working on production and development of photorefractive materials.

New scientific results

1. I created and developed a topographical system, based on Bragg-diffraction, to investigate the wave-mixing processes in thick crystals. By using the system, I showed experimentally that the soliton-like spatial stationary distribution of the modulation of the dielectric constant, which is a solution of the sine-Gordon differential equation and is expected for four-wave mixing in photorefractive materials with non-local response, is approximately realized in lithium niobate, it really has a local maximum, and its location depends on the input intensity ratios of the recording beams as predicted by the theory. I also showed experimentally, that the shape of the distribution is essentially the same during build-up as later in the stationary state, with no change of its location either. [1, 2, 3, 4, 5]

2. By using activation energy determination, I gave experimental support to the assumption that the ions responsible for the thermal fixing of photorefractive holograms in lithium niobate are protons originating from OH^- ions. In the experiments I measured the temporal behavior of the thermal relaxation of the two-wave mixing gratings at different temperatures in iron and manganese doped nearly stoichiometric and congruent lithium niobate crystals. I observed simple exponential decays with time constants that closely follow Arrhenius' law. The resulting activation energies coincide well with those determined from the thermal kinetics of OH^- absorption bands, described in point 2 of paragraph 'Preliminaries of the research'. [6, 7]
3. I developed and tested a novel method to measure the activation energy of the ions responsible for hologram fixing, valid for all photorefractive materials that require thermal fixing. I found the method to be convenient by showing some advantages in comparison with the two-wave mixing method. The essence of the method is the application of the holographic light scattering phenomenon for the measurement of thermal relaxation. Instead of the Bragg-diffraction on a two-wave mixing grating, we measure the temporal evolution of the intensity loss of a much weaker transmitted beam caused by remaining parasitic holograms, the latter originating from the self-amplification process due to intensive pumping during recording. I found the method to be practicable: the measured activation energies of the thermally mobile ions in nearly stoichiometric iron-doped and in both nearly stoichiometric and congruent manganese-doped lithium niobate crystals coincide both with the activation energy of protons and the activation energy determined from the standard two-wave mixing method. In the case of the iron doped congruent sample larger differences were found that, after excluding other reasons, could be interpreted in terms of excessively deep modulation presumably occurring in two-wave mixing experiments. [8, 9, 10]
4. I measured the angular distribution of the holographic scattering in lithium-niobate, and found two empirical formulae that are easy to integrate and fit both to the experimental data and to the function calculated from the theoretical angular dependence of the gain Γ . The empirical functions depend on spatial frequency in the following manner:

$$I_s(k) = \frac{a}{\left[(k - k_1)^2 + k_2^2\right]^2} - b \cdot \exp(-c_1^2 k^2), \text{ and}$$

$$I_s(k) = a \cdot \exp(-b^2 (k - k_1)^2) - c_1 \cdot \exp(-c_2^2 k^2),$$

where a , b , c_1 , c_2 , k_1 , and k_2 are parameters to fit. I used the formulas as integrating weight functions, and calculated Taylor-approximations of activation energy values calculated by two kinds of time-dependent formulas. For the parameters of our samples, I found that the use of both time-dependent formulas practically eliminates the effect of the problem arising from the fact that there occurs not only one but an infinity of grating periods, and the relaxation time depends on the grating period. Moreover, I have given an explicit inequality relation incorporating the fitting parameters of any of the two weight functions and the material constants of the crystal, which can be used for the decision whether the holographic scattering method can be applied in a given case. [10]

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Scientific publications connected to the theses

1. S. Bugaychuk, L. Kovács, G. Mandula, K. Polgár, R. A. Rupp: "Wave-mixing solitons in ferroelectric crystals", *Radiation Effects and Defects in Solids*, **157** (2002) 995-1001
2. S. Bugaychuk, L. Kovács, G. Mandula, K. Polgár, R. A. Rupp: " Non-uniform dynamic gratings in photorefractive media with non-local response" *Physical Review* **E 67** (2003) 046603/1-8
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The whole PhD dissertation is available at www.szfki.hu/~mandula/phd.html.

Further scientific publications

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13. G. Mandula, R. A. Rupp, M. Balaskó, L. Kovács: "Decay of photorefractive gratings in $\text{LiNbO}_3\text{:Fe}$ by neutron irradiation"; *Applied Physics Letters*, accepted for publication

