PHOTOREFRACTIVE EFFECTS AND PHOTONICS

Growth, Preparation and Characterization of Visible and Infrared Sensitive $KNbO_3$ Crystals

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For holographic applications the magnitude and speed of the photorefractive effect are critical design parameters. Iron-doped $KNbO_3$ is one of the preferred materials for such applications at visible wavelengths. However, the photorefractive response time of unmodified crystals can limit device performance. At near-infrared wavelengths very many applications are feasible because of the availability of cheap semiconductor diode lasers. Unfortunately, no material exhibits a sufficient photorefractive sensitivity at these wavelengths. Our research addresses two related topics: how to increase the photorefractive sensitivity at visible wavelengths and how to extend its wavelength range into the near infrared.

These aims were pursued by two different methods: chemical treatment of crystals in a reducing atmosphere and intentional doping with a range of impurities. In the post-growth chemical treatment a mixture CO/CO_2 was employed. The ratio of the two gases determines the oxygen partial pressure. Oxygen leaves the surface of the crystal and oxygen vacancies are distributed homogeneously in the bulk. For each oxygen vacancy two electrons are available for filling the lowest impurity states. For near-infrared photorefraction a number of different dopants (Cu, Fe, Ni, Co, Mn, Rh, and Mn-Rh) have been introduced into $KNbO_3$ with the aim of producing photoactive centers with sufficiently small ionization energies. By combining the dopant rhodium with our reduction treatment, crystals with large photorefractive sensitivities at 860 nm have been produced. Optical characterization was done by measuring absorption spectra, effective trap densities (a measure of the absolute and relative concentrations of charge donors to acceptors), and photorefractive response times. The effective trap densities were compared to measurements of the absolute impurity concentrations performed with ICPMS, SIMS, and AASCA. The dark and photoconductivity were also measured to determine the mobility-lifetime product of the photoexcited charges.

Reduction of Fe:$KNbO_3$ increases the speed and magnitude of the photorefractive response. Electrons are the dominant photoexcited species in reduced Fe:$KNbO_3$, whereas holes dominate in the untreated crystals. The photorefractive sensitivity is increased by one order of magnitude with response times in the millisecond range. Compared to untreated Rh:$KNbO_3$ reduction increases the photorefractive sensitivity at 860 nm by four orders of magnitude to a value comparable to that of untreated Fe:$KNbO_3$ at visible wavelengths.

Sponsor: Swiss National Science Foundation

Photorefractive Materials for the Ultraviolet

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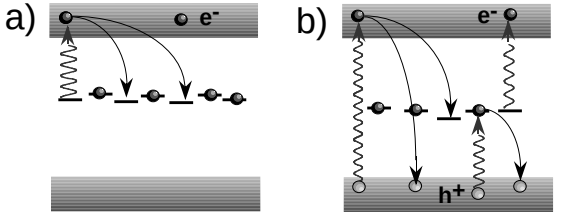
Besides the development of new materials for applications at infrared wavelengths, there is a particular interest for materials showing photorefractive properties in the ultraviolet region of the electromagnetic spectrum. The advantages available by operating at higher photon energies consist, on one hand, in shorter response times, and, on the other hand, in an increased resolution and storage capacity for holographic memory applications due to the shorter wavelengths employed.

Several materials have been investigated like $KNbO_3$, $LiTaO_3$ as well as $LiNbO_3$, and few compounds belonging to the BBO family. The crystals were characterized by two- and four-wave mixing experiments to determine the efficiency and the speed of the photorefractive processes. Measurements on photoconductivity have also been performed to investigate the photovoltaic currents.

In pure $KNbO_3$, four-wave mixing experiments in the near UV range have been performed in order to study the dynamics of the time and spatial evolution of the photorefractive phase gratings induced by direct band-to-band photoexcitation. The dynamics is governed by several characteristic times, the fastest of which is of the order of ~50 μs, whilst the spatial distribution is found to basically depend on the light intensity. In contrast, in $LiTaO_3$ as well as $LiNbO_3$ the charge carriers were photoexcited from levels lying within the energy band-gap of the material. In this conventional regime, among the investigated samples, $LiTaO_3$:Rh crystals show very promising large energy coupling constant (up to 30 cm^{-1}) in two-wave mixing experiments. Beam fanning as well as self-diffraction effects have also been observed. The response times in $LiTaO_3$ are slow, of the order 15 min with 150 mW cm^{-2} intensity at 364 nm wavelength. Compounds of the BBO family have also been investigated. At near UV wavelengths the almost vanishing photon absorption precluded the generation and the detection of a photorefractive phase grating. A proper doping of these crystals might activate these materials as well.



Schematic band model for the conventional (a) and interband (b) photorefractive processes. In (a) transitions between the mid-gap level and only one band are allowed. With interband photoexcitations (b), all the possible transition channels are open.

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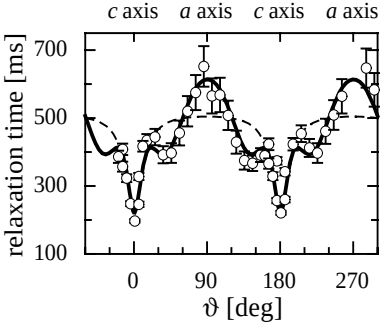
Characterization of Charge-Transport in Electro-Optic Isolators

P. Bernasconi, I. Biaggio and M. Ewart

The understanding of charge transport mechanisms in insulating, photoconductive materials is important both from a fundamental point of view and to develop and predict the performance of devices where these materials are employed. In this project we develop and apply all-optical holographic methods which are particularly well adapted also in the case of low charge carrier densities ($<10^{14}$ cm^{-3}), low mobilities (<1 cm^{2}V^{-1}s^{-1}), and short lifetimes ($\sim$1 ns) where traditional methods, like the time-of-flight method or the Hall effect, are especially difficult or impossible to apply. In our techniques, charge carriers are photoexcited by a sinusoidal interference pattern induced by a short pulse or continuous wave (cw) illumination. When exciting with a short laser pulse, movement of the photoexcited charge distribution takes place in the dark after excitation, and yields information on the carrier mobility and lifetime. When using cw lasers, charge is redistributed during illumination and its dynamics are related to the dielectric relaxation time and therefore to the photoconductivity.

We measured the evolution of the space-charge distribution after illumination with short pulses and obtained information on electron-hole competition and the reduction degree of iron doped $KNbO_3$ crystals. The effects of electron and hole diffusion after photoexcitation in a slightly reduced crystal could be clearly separated. This effect can be used to complete the characterization of the charge carriers in $KNbO_3$ by measuring the band mobility of holes.

Under cw illumination, the directional dependence of the decay time of a space charge distribution was used to determine the anisotropy of the charge carrier mobility in $KNbO_3$ and $BaTiO_3$ single crystals. In *n*- and *p*-type $KNbO_3$ we obtained anisotropies ranging from a factor 1 to 3 along the three different crystallographic directions while in *n*-type $BaTiO_3$ we find a mobility ratio of 20 along the two inequivalent axes.These measurements are also the first direct confirmation of the peculiar way in which acoustic phonons contribute to the dielectric permittivity when the electric field inside the crystal is spatially.



Dielectric relaxation time as a function of the space-charge field orientation. The solid line is a fit taking into account the sinusoidal spatial modulation of the space-charge field. The dashed line corresponds to a spatially homogenous field.

Reference: P. Bernasconi, I. Biaggio, M. Zgonik, P. Günter, *Phys. Rev. Lett.* ***78****, 106 (1997).*

Sponsor: Swiss National Science Foundation

Pulsed 4-Wave Mixing Studies for Fundamental Characterization of Optical Nonlinearities

I. Biaggio, M. Ewart and M. Zgonik

Subpicosecond multi-beam interactions in nonlinear materials are attracting researchers because of their potential for device applications such as fast all-optical switching. Non-centrosymmetric materials with large second order nonlinear optical effects can be very important also for this kind of applications, where a third order nonlinearity is required. We recently demonstrated that in these materials the effective third order nonlinear effects can be amplified significantly by cascading of second order contributions. The second order processes of optical rectification and electro-optic effect contribute significantly to, and can even dominate, nonlinear optical effects such as self-phase-modulation, self-focusing, and degenerate four-wave-mixing.

We carried out comprehensive four-wave-mixing experiments where both third-order non-linear optical effects and cascaded second order effects were observed in $BaTiO_3$. By comparing measurements performed with different beam polarizations and wavevector directions we determined the relative magnitudes of second-order contributions and pure third-order effects. The experimental procedure can be summarized as follows. (1) Using the electro-optic and dielectric tensors determined separate experiments the magnitude of the cascaded second order contributions to the degenerate four-wave-mixing signal is calculated. (2) Degenerate four-wave-mixing signals are measured in a variety of geometries where the direct third order contributions and the second-order cascaded contributions have different magnitudes. (3) The absolute values of the third order susceptibility tensor elements are determined from a least-squares fit to the theoretical expression for the effective susceptibility. Absolute measurements of third order susceptibilities normally require an accurate characterization of the beam profile and pulse-shape. This is not the case for the method developed here. Neither the determination of the electro-optic and dielectric properties of the sample, nor the comparison of the various degenerate four-wave-mixing signals require an exact knowledge of beam profile and pulse shapes. This method therefore combines the principal advantages of relative and absolute measurements: (1) It does not require an accurate determination of the laser pulse parameters and beam profile. (2) It does not have to rely on previously determined third order susceptibilities of a reference material, so that it is not affected by any previous errors or calibration problems.

The two main results of this project are the development of an effective new method for the absolute measurement of nonlinear optical susceptibilities, and the demonstration that, in some geometries, cascaded contributions are responsible for more than 90% of the four wave mixing signal in $BaTiO_3$.

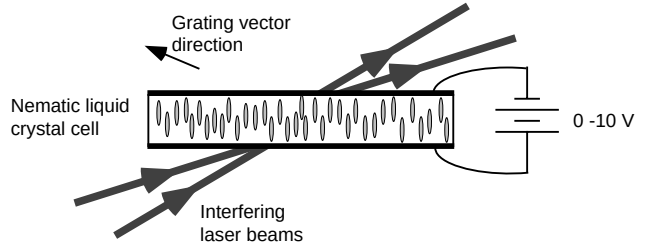
Holographic Gratings in Strongly Anisotropic Materials

G. Montemezzani and M. Zgonik

By means of the photorefractive effect dynamic phase holograms can be produced and interaction between low intensity light waves becomes possible. Worldwide, there is a constant effort to increase the sensitivity and performances of photorefractive materials. One approach towards this aim is to look for materials with a stronger electro-optic response, i.e. a stronger refractive index change in response to an electric field. Highly anisotropic materials such as liquid crystals and low glass temperature polymers are particularly promising in this respect. Field-induced rotation of the strongly birefringent molecules give rise to large quadratic electro-optic effects.

We evaluated the performance of nematic liquid crystal mixtures doped with dye molecules in view of applications in dynamic holography. Samples based on liquid crystals mixtures doped with increasing amounts of Rhodamine 6G (< 0.2 wt.%) were prepared using conventional surface preparation techniques. Dye-photodissociation results in charge separation and formation of a space-charge field in the liquid. In the oblique geometry depicted in the figure this field adds to the external field applied across the sample to produce a phase hologram by a pseudo-linear electro-optic effect related to molecular reorientation.

In 75 μm thick 0.2% R6G-doped E63 samples holographic diffraction efficiencies of the order of 50% could be obtained already with a drive voltage of 4 V using light intensities of 0.15 Wcm^{-2} (λ = 514 nm). The related holographic sensitivity (refractive index change per unit incident energy) is only a factor of 20 below that for reduced $KNbO_3$, a top material for visible holographic applications. To analyze the holographic experiments in these optically highly anisotropic materials a new theoretical treatment of the light diffraction processes was made necessary [1]. Significant modifications with respect to the relationships valid for isotropic materials have been found.



Oblique geometry for holographic recording in nematic liquid crystal cells.

Reference: G. Montemezzani and M. Zgonik, "Light diffraction at mixed phase and absorption gratings in anisotropic media for arbitrary geometries", *Phys. Rev. E* **35**, 1035 (1997).

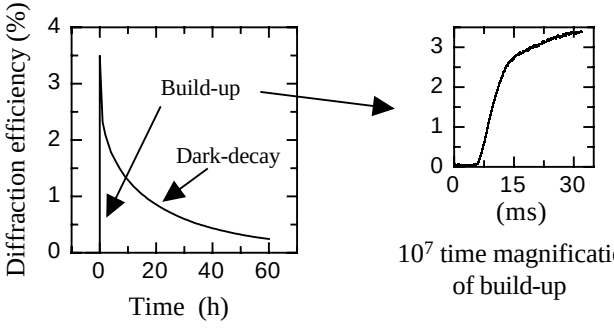
Optical Memories and New Materials for Long-Term Storage

R. Ryf, C. Medrano, G. Montemezzani, in collaboration with K. Kitamura, National Institute for Research in Inorganic Materials, Tsukuba, Japan

Optical memories based on photorefractive volume holography have potentially important advantages over traditional two dimensional storage systems. The storage density is increased by several orders of magnitude because the information is stored in the volume, and a very high read-out throughput can be achieved by use of parallel readout techniques. The main difficulty of holographic storage is that the ideal material which should have a short writing time (ms) and long storage time (hours-years) is yet to be found. The aim of this project is to investigate how the properties of photorefractive materials can be improved using different dopants and different growth conditions.

We have investigated two materials: lithium niobate ($LiNbO_3$) and potassium niobate ($KNbO_3$). $LiNbO_3$ is the mostly used material for optical storage because of long dark storage times (month-year), but as drawback needs a long writing time (sec-min). We investigated the build-up time and the diffraction efficiency of an holographic grating in newly grown stochiometric pure and Fe doped $LiNbO_3$ crystals. In $KNbO_3$ the writing time is much shorter than in $LiNbO_3$, but the storage time is usually also shorter. Possible improvement of the holographic storage time in the dark by doping with inputities (Ni, Rh, Mn and Ce) has been investigated.

In newly grown stoichiometric Fe doped $LiNbO_3$ we obtained a writing time of 500 ms at 1 W/cm^2 light intensity and 488 nm wavelength. This is 3-4 order of magnitude better than in pure $LiNbO_3$. In $KNbO_3$ a maximal storage time of 103 hours was measured in Mn doped crystals. Ce doped $KNbO_3$ crystals are very promising due to the extremely large asymmetry between recording time and dark storage time (see figure). We measured a fast build-up time of 20 ms at 1 W/cm^2 and a storage time of 31 hours.



Buildup and decay of a holographic grating in Ce doped $KNbO_3$

Sponsor: Swiss National Science Foundation

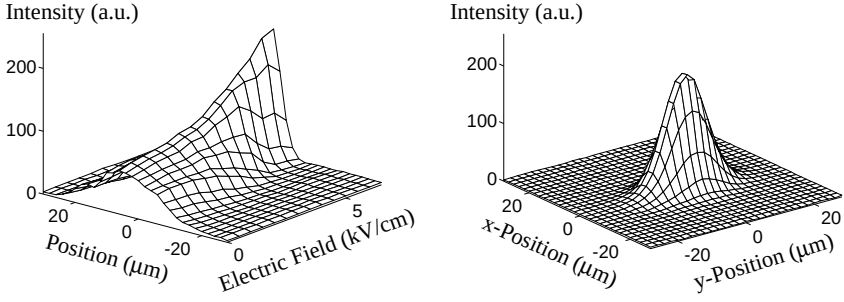
Self-Focusing and Spatial Solitons in Photorefractive $KNbO_3$

R. Ryf, G. Montemezzani and M. Zgonik

Nonlinear optical interaction can lead to beam self-focusing in appropriate nonlinear materials. If the self-focusing effects compensates the natural diffraction of a laser beam, light induced waveguides and spatial solitons can be generated. These processes are particularly interesting in photorefractive materials because there is no need for a high power light source. In this materials the light induced optical elements will persist in the dark due to their long dark decay times. The induced waveguide can be used to guide a beam at a wavelength where the materials shows no photorefractive sensitivity.

We concentrated the investigations of self-focusing effects to $KNbO_3$ crystals because of the large electro-optic coefficients and the strong photorefractive properties. In order to characterize the material nonlinearity we first investigated light induced lens in thin crystal using the Z-scan technique. In Z-scan the focus of a Gaussian beam is moved along the propagation direction of the light through the investigated nonlinear material. The width of the beam, which depends on the strength of the light induced lens, is determined in the farfield as a function of the position of the focus. The formation of photorefractive spatial soliton was systematically investigated varying the applied electric field, the beam width and the beam intensity and monitoring the profile of the outcoming beam for each condition (see figure).

In $KNbO_3$ strong light induced lens were observed even without applying an electric field. The effect is in this case driven by the photogalvanic effect. We demonstrated that photorefractive Z-scan permits a complete and precise determination of the material parameter relevant for photorefractive soliton observation. For the first time we demonstrated one dimensional photorefractive spatial solitons in $KNbO_3$ with beam width as small as as 15 μm. By focusing in both transversal directions we were able to form a waveguide which was used to guide a second beam at a longer wavelength.

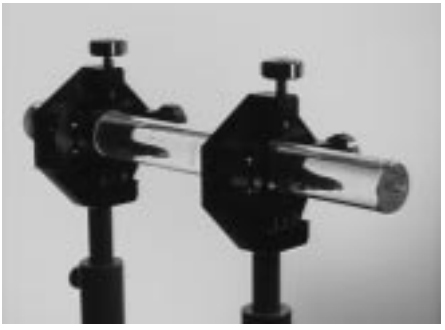


One dimensional (left) and two dimensional (right) photorefractive soliton

Study and Development of Wave Front Reversing Mirrors and Laser Sources at 2.1 μm Wavelength

I. Poberaj and I. Biaggio

Laser systems operating at 2.1 μm are very interesting for many applications since they operate in the eye safe spectral region. Potential applications range from medicine to remote sensing, range finding, and spaceborne lidar systems.
Our research effort is twofold. We first developed a flashbulb pumped Q-switched master oscillator power amplifier (MOPA) system operating at 2.1 μm based on Cr,Tm,Ho:YAG as active material. The power amplifier uses a phase conjugating stimulated Brillouin scattering (SBS) liquid cell (see figure) to compensate for beam distortion induced by the amplifying medium. Secondly, we evaluated the potential of an all solid state laser oscillator system. The use of a nonlinear optical interactions to generate the required wavelength around 2.1 μm was investigated, with the aim of finding the optimal system design and materials.



Liquid Stimulated Brillouin Scattering (SBS) cell for optical phase conjugation

The flash lamp pumped 2.1 μm MOPA system developed in this study serves now as an additional light source in our laboratory. It has typically 90 mJ pulse energy with 80 ns pulse length and 1 Hz repetition rate. Among the different liquids tested for SBS phase conjugation (CCl_3F, $C_2Cl_3F_3$, CS_2, $TiCl_4$ and $SnCl_4$) $SnCl_4$ was found to be the best one because no optical breakdown occurs at higher laser pulse energies. In this liquid we found a phase conjugation threshold of 23 mJ and a phase conjugation reflectivity of 60% at a pulse energy of 55 mJ. This results could be achieved only after development of a special cell cleaning and liquid purification technology. For the second part of the project our initial technical evaluation singled-out promising configurations for farther research. They make use of a diode-pumped oscillator system coupled to an optical parametric oscillator. The potential of systems taking advantage of difference frequency generation is also being investigated.

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