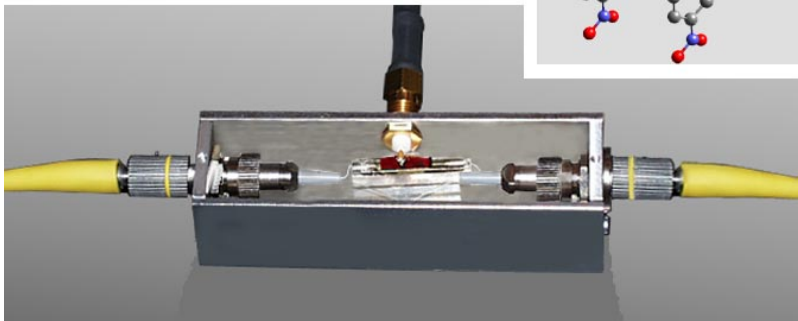
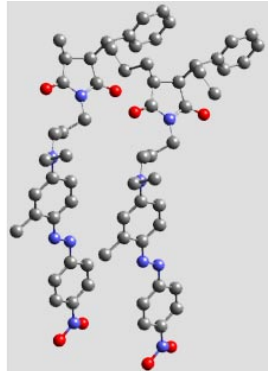
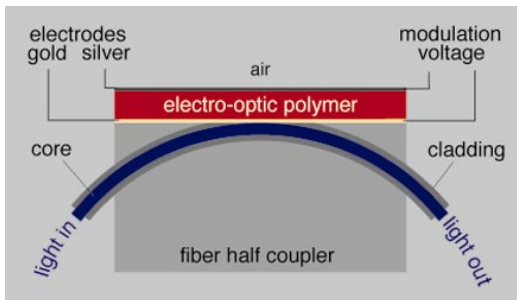


Nonlinear Optics Laboratory



***Previous Page:
Polymer based electro-optic inline fiber modulator.(see page 12)***

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Paola Cereghetti **Dr. Tomas Pliska** *until December*
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SPONSORS

ETH Zürich
Swiss National Science Foundation
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RESEARCH SUMMARY

The main research topics of the Nonlinear Optics Laboratory in 1997 were:

- Molecular Crystals and Polymers
- Photorefractive Effects and Applications in Optical Parallel Processing
- Integrated Optics and Optical Frequency Conversion
- Molecular Beam Epitaxy of Organic Thin Films
- Investigation of Organic Molecules at Interfaces using Nonlinear Optics and Atomic Force Microscopy
- Magnetic Resonance Spectroscopy.

The details of the research projects of these fields are given in this report. In all these topics considerable progress has been made. In this foreword we would like to mention only a few highlights of the results which appear most exciting for us.

The development of infrared, sensitive photorefractive materials which are compatible with laser diodes wavelengths led to sub-second response times in KNbO_3 :Rh crystals and to the development of a new procedure and a better understanding of the influence of the reduction treatment during the growth of the material. The control of the reduction ratio by high energy proton implantation in photorefractive Fe-doped KNbO_3 allows to enhance the performance of KNbO_3 waveguides or thin film crystals for self-pumped phase-conjugation. The build-up time of such self-pumped phase conjugators could be decreased by more than two orders of magnitude in strongly reduced crystals compared to as grown crystals, resulting in a response time of 100 ms at an intensity of 8 W/cm^2 . At the same time the self-pumped phase-conjugate reflectivity in perfectly reduced crystals has been enhanced by a factor of four as compared to the grown crystals and provided reflectivities of 60 percent at a wavelength of 514.5 nm.

Further studies lead to the discovery that the photorefractive two-wave mixing gain can be significantly enhanced by making use of the photoexcitation dichroism present in some specially doped crystals, an effect which had been either ignored or not understood before.

The exciting experiments with the atomic force microscope (AFM) for the manipulation and investigation of ferroelectric domains reported last year were successfully continued. We are now able to measure simultaneously the topography and all three components of the sample polarisation by voltage modulated scanning force microscopy developed in our laboratory. The minimum domain size for writing small "bits" of ferroelectric domains by AFM could be decreased by a factor of four since last year. Circular domains written into barium-titanate crystals by applying voltage pulses to the scanning force microscope tip have now a diameter of only 150 nm. The surfaces of the organic crystals COANP and DAST (4-N,N-dimethyl-

lamino-4'-N'-methylstilbazolium toluene-P-sulfonate) were investigated by atomic force microscopy. A high attractive force (10^{-4} N) is observed between the AFM tip and the sample. High resolution and layer resolution AFM images could be achieved for the first time.

Multilayer organic light emitting devices have been successfully fabricated. In situ AFM, XPS and UPS measurements have been used to study the formation of the metal-Alq interface (Alq: (tris(8-hydroxyquinolate)aluminum (III))). The strong interdiffusion at Mg/Alq interface is avoided by introduction of a thin Ag film (2-3 monolayers).

The organic MBE chamber was extended by an ellipsometer as an optical in situ tool. This instrument has routinely been used for thickness measurements of organic thin films deposited on silicon.

The UHV atomic force microscope has been completed in 1997. In contact mode, atomic resolution has been demonstrated for MoS₂. In addition, the non-contact mode was also realized. It has been used to resolve monoatomic steps and to determine the surface roughness of different organic thin film structures.

Nonlinear optical thin films have been realized on the basis of PVBA (4-[trans-2-(4-pyridyl-vinyl)]benzoic acid), an organic compound developed in our laboratory. This organic material with a high melting temperature of 350° C was deposited onto different substrates at different growth conditions. Optical second harmonic generation experiments of organic films deposited on transparent substrates (glass, quartz and sapphire) reveal molecules lying within the substrate plane with a preferential orientation. This anisotropy is expected to be determined by the oblique incidence of the molecular beam.

In 1997 Dr. M. Ewart, Dr. Th. König and Dr. T. Pliska have completed their PhD thesis and have left our laboratory. Dr. M.S. Wong was elected "senior lecturer" at the Australian National University in Canberra and Dr. L. Eng continues his research at the University of Basel. Dr. Ch. Bosshard completed his habilitation thesis and has received his "venia legendi" for starting his lectures on April 1st, 1998. We thank all these colleagues for their successful work and pleasant cooperation. During 1997 we could welcome Dr. M. Jäger as a new post-doctoral fellow. He joined us after completing his PhD thesis at the "Center for Research on Lasers and Electro-optics (CREOL)" of the "University of Central Florida", where he worked with Prof. G. Stegeman. As a compensation Dr. T. Pliska who graduated with us will start his postdoctoral stay with Prof. Stegeman in February 1998, continuing the successful exchange of scientists between CREOL and the Institute of Quantum Electronics since several years.

Activities of members of the Nonlinear Optics Laboratory in Conference Committees and Editorial Boards

Several members of the nonlinear optics laboratory continued to be active in a series of international committees and as editorial board member of scientific journals in the fields of optics, nonlinear optics, quantum electronics, solid state physics of ferroelectric, organic and polymeric materials. Prof. P. Günter is a member of the editorial board of the following scientific journals:

- "Ferroelectrics"
- "Ferroelectrics Letters"
- "Nonlinear Optics"
- "Optica"
- "Optics Communication"
- "Photonics Science News"

and was/is a member of the advisory or program committees of the following international conferences:

- "Conference on Lasers and Electro-Optics" (CLEO Europe '98)
- "Electrical and Related Properties of Organic Solids" (ERPOS-7)
- "European Conference on Applications of Polar Dielectrics" (ECAPD) (Chairman of the European Steering Committee)
- IEEE Ferroelectrics Committee of the "Ultrasonics, Ferroelectrics and Frequency Control Society"
- "International Conference on Organic Nonlinear Optical Materials" (ICONO-4)
- "International Symposium on Lasers and Nonlinear Optical Materials" (IS-LNOM '97)
- "Materials for Nonlinear Optics" (European Optical Society Topical Meeting 1997)
- "Nonlinear Frequency Conversion: Materials, Devices and Applications" (SPIE 1997 Symposium)
- "Organic Thin Films for Photonic Applications" (1997 Topical Joint Meeting of the Optical Society of America and the Chemical Society of America)
- "Photonics China '98" (SPIE Int. Symp. on Lasers and Optoelectronics)
- "Nonlinear Optics '98"
- OSA Topical Meeting on "Photorefractive Effects and Applications"
- World Ceramics Congress on "Electronic, Magnetic and Optical Ceramics". (CIMTEC'98: 9th International Conference on Modern Materials and Technologies).

Prof. R. Kind is the Secretary General of the Groupement AMPERE.

MOLECULAR CRYSTALS, THIN FILMS, AND POLYMERS**Design, Synthesis, and Crystal Growth of Second-Order Nonlinear Optical Materials**

M. S. Wong, F. Pan, C. Cai and M. Ehrensperger

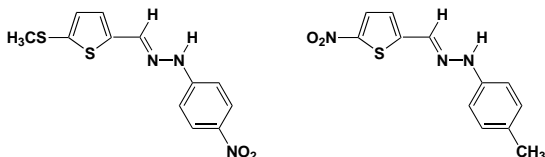
The potential upper limits of macroscopic nonlinearities and long-term orientational stability of molecular crystals are significantly superior to those of polymers and Langmuir-Blodgett thin films. Therefore, these crystals are of great interest for opto-electronic applications, such as ultra-fast (GHz) electro-optic applications, or optical parametric generation. However, the probability of molecules to crystallize noncentrosymmetrically as required for second-order nonlinear optics is very low. Moreover, the crystallinity and crystal properties of the rod-like, highly extended π -conjugation systems are usually poor which is detrimental for practical applications.

With respect to the above problems, we investigate two novel crystal engineering approaches. These are (i) the use of the non-rod-shaped framework-hydrazone skeleton as a π -conjugation core and (ii) the supramolecular synthetic approach based on the short hydrogen bond induced self-assembling of molecular aggregates. In this past year work concentrated on the first approach.

As an example we showed that often hydrogen bond directed lambda-shaped supramolecular assemblies are the key and fundamental basis of hydrazone-based non-centrosymmetric crystal packings.

Thiophene based hydrazone derivatives can considerably enhance the molecular second-order nonlinearities. Two examples of this series with excellent crystal properties are 5-(methylthio)-thiophenecarboxaldehyde-4-nitrophenylhydrazone (MTTNPH) and 5-nitro-2-thiophene-carboxaldehyde-4-methylphenylhydrazone (NTMPH). In the case of NTMPH we showed that re-crystallization of NTMPH in various solvents under different conditions yielded three crystalline phases, red greenish plates, red orange prisms, and black needles (NTMPH-III) in space group $P2_1/n$.

Examples of thiophene based hydrazone derivatives: MTTNPH (left) and NTMPH (right).



References: F. Pan, Ch. Bosshard, M. S. Wong, C. Serbutoviez, K. Schenk, V. Gramlich, and P. Günter, *Chem Mater.* **9**, 1328 (1997)

M. S. Wong, V. Gramlich, Ch. Bosshard, and P. Günter, *J. Mater. Chem.* **7**, 2021 (1997)

Sponsor: Swiss National Science Foundation

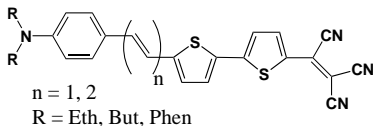
Second-Order Nonlinearities of Conjugated Organic Molecules

I. Liakatas, M. S. Wong, C. Cai and Ch. Bosshard

Organic materials are very interesting for ultra-fast (GHz range) electro-optic applications as their nonlinear optical response depends mainly on the polarizability of the electrons in contrast to inorganic materials where lattice vibrations play a dominant role. Our group is continuously working on the development of conjugated organic molecules with high second-order nonlinearities for crystals or incorporation to polymers. We investigate the second-order nonlinearities of our molecules using electric field induced second harmonic generation (EFISH) and hyper-Rayleigh scattering (HRS).

In organic crystals, molecules need to exhibit high nonlinearity as well as to crystallize in optimized acentric structures. Therefore, a series of multi-donors substituted 4-nitrophenylhydrazones, which show a high preference for noncentrosymmetric packing, have been designed and synthesized, and their first-order hyperpolarizabilities β were investigated and compared. Since these molecules are non-rod-shaped as well as coloured, an HRS experiment at 1542 nm was used for the off-resonant determination of different combinations of β tensor elements of different substituted hydrazone molecules. It was found that the cooperative effect of multiple moderate electron-donors exhibits a donating strength comparable to that of one strong electron-donor enhancing the molecular nonlinearity and the chemical stability. One of the best molecules of this series, 3,4-dihydroxybenzaldehyde-4-nitrophenylhydrazone, 3,4-DHNPH, is used for growing crystals with an optimized structure for electro-optic applications.

In poled polymer applications, such as electro-optic modulators, molecules need to exhibit a large product ($\mu\beta$) of ground-state dipole moment μ and the molecular nonlinearity β as this is the figure of merit for a high degree of orientational order in electric field poled polymeric thin films. Bithiophene based molecules were synthesized and characterized using EFISH at 1907 nm. Our synthesized dibithiophene molecules turned out not to fulfill the expectations by having moderate nonlinearity and rather bad photostability due to their big length. In contrast, a bithiophene in conjugation with a phenyl ring, yielded a $\mu\beta$ product of $20000 \cdot 10^{-69} \text{ m}^5\text{C/V}$ which is one of the largest ever observed for stable chromophores. (see Section 1.7)



Highly nonlinear bithiophenes in conjugation with a phenyl ring. The combination of $n = 2$ and $R = \text{Eth}$ yielded a $\mu\beta$ product of $20'000 \cdot 10^{-69} \text{ m}^5\text{C/V}$.

Reference: I. Liakatas, M.S. Wong, Ch. Bosshard, M. Ehrensperger and P. Günter, *Ferroelectrics*, **202**, 299 (1997).

Sponsors: Swiss National Science Foundation, ETH Zurich

Cascading of Second-Order Nonlinearities

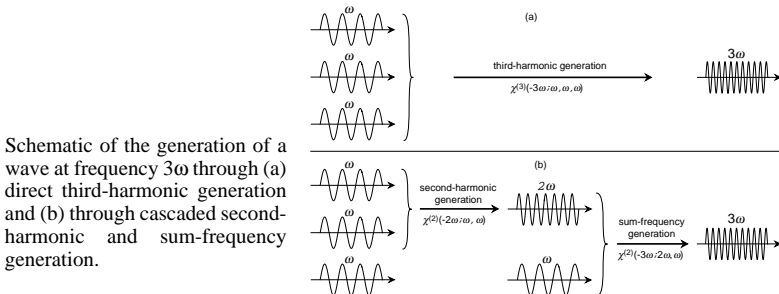
U. Gubler, Ch. Bosshard and I. Biaggio

Cascading is a process where lower-order effects are combined to contribute to a higher-order nonlinear process. Large nonlinear phase shifts due to cascading (second-harmonic generation and difference-frequency mixing as well optical rectification and the linear electro-optic effect) were observed in several materials. Cascading also occurs in third-harmonic generation through the combination of frequency-doubling and sum-frequency generation. Furthermore, cascading through the combination of optical rectification and the linear electro-optic effect can strongly increase the diffraction efficiency in degenerate four-wave mixing.

During this past year we have (i) used cascading in third-harmonic generation at several wavelengths to obtain the dispersion of $\chi^{(3)}$ of KNbO_3 , α -quartz, and fused silica and (ii) degenerate four-wave mixing to determine the most important tensor elements of $\chi^{(3)}$ of the highly nonlinear crystal 4-N,N-dimethylamino-4'-N'-methylstilbazolium toluene-p-sulfonate (DAST).

From theoretical fits of experimental third-harmonic generation curves of KNbO_3 and α -quartz crystals which show cascaded processes we directly obtained the ratio $\chi^{(3)}/(\chi^{(2)})^2$. From the known values of $\chi^{(2)}$, the value of $\chi^{(3)}$ could be determined and a subsequent comparison with the standard material fused silica then yielded $\chi^{(3)}$ of fused silica. We have now completed our work in this research area by the determination of the ratio $\chi^{(3)}/(\chi^{(2)})^2$ at wavelengths between $\lambda=1064\text{nm}$ and 2100nm . Our results showed that a one-oscillator model with an infrared correction term can accurately explain the dispersion of the third-order susceptibility $\chi^{(3)}$.

Third-order nonlinear optical properties of DAST were evaluated with degenerate four-wave mixing using 100ps pulses at $\lambda=1064\text{nm}$ yielding large values of the effective third-order susceptibility of $\chi^{(3)}=4300\pm 500 \times 10^{-22} \text{ m}^2/\text{V}^2$. Our experiments confirmed that degenerate four-wave mixing is a very useful and reliable tool for the determination of tensor elements of the third-order susceptibility $\chi^{(3)}$.



Schematic of the generation of a wave at frequency 3ω through (a) direct third-harmonic generation and (b) through cascaded second-harmonic and sum-frequency generation.

Sponsor: ETH Zurich

Third-Order Susceptibilities of Conjugated Organic Molecules

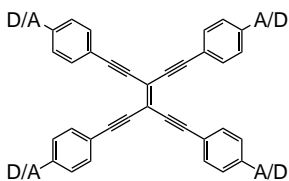
U. Gubler and Ch. Bosshard in collaboration with R. Martin, R. Tykwinski and F. Diederich, Laboratory of Organic Chemistry, ETH Zurich

For electronic and photonic applications, organic molecules with extended conjugation of the π -orbitals and electron-donating or accepting substitution of the end-groups are of particular interest. To improve our knowledge about the influence of molecular structure on the second-order hyperpolarizabilities γ , we focused mainly on two aspects: conjugation length expansion and the symmetry of the substitution pattern.

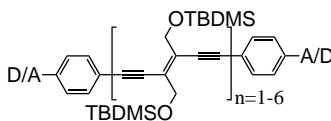
For our one-dimensionally conjugated molecular system polytriacetylene (PTA), we could measure symmetrically substituted oligomers up to a length of six monomer units (about 4.5 nm) using third-harmonic generation. They show higher second-order hyperpolarizabilities γ than the earlier measured unsubstituted samples, but the increase vs conjugation length does not follow a power law anymore.

We extended our investigation on two-dimensionally conjugated molecular scaffolds and changed the substitution pattern deliberately to obtain all possible symmetry configurations. For the first time up to our knowledge for two-dimensionally conjugated molecules in general we were able to explain the series of measured nonlinearities by the symmetry of the electronic wavefunctions.

The synthesis of our PTA- and TEE-molecules was performed in the group of F. Diederich at the Institute of Organic Chemistry of ETH.



substituted, two-dimensionally conjugated molecular scaffold (TEE), where D and A denote electron donating (dimethylamino) and accepting (nitro) groups



substituted, one-dimensionally conjugated molecules (PTA), with conjugation length of up to six monomer units (≈ 4.5 nm) and neutral sidegroups (OTBDMS)

In a cooperation with M. Blanchard (ESN, Paris), we also measured the nonlinear susceptibilities of polyene samples, which show generally the highest second-order hyperpolarizabilities γ . However, lacking thermodynamic and chemical stability will limit the usefulness of these compounds for commercial application.

Reference: R. Martin, U. Gubler, C. Boudon, V. Gramlich, C. Bosshard, J.-P. Gisselbrecht, P. Günter, M. Gross, F. Diederich, *Chem. Eur. J.* **3**, 1505 (1997).

Sponsor: ETH Zurich

Nonlinear Optical Properties of Molecular Crystals

U. Meier, M. Bösch, M. S. Wong, F. Pan and Ch. Bosshard

Within this project, new promising organic nonlinear optical crystals are developed and investigated with special interest on highly extended π -conjugated systems (e.g. donor-acceptor di-substituted stilbene and hydrazone derivatives). A simple model shows that the requirements for the chromophore orientation in the crystal lattice depends on the application: for optical frequency-conversion the active molecules should be oriented with an angle of about 55° with respect to the polar crystal axis for the most interesting point groups, whereas for electro-optics the chromophores should be oriented parallel to each other.

The determination of (i) possible phase-matched parametric interaction as well as of (ii) the relevant figures of merit for nonlinear optical and electro-optic application are based on the values of the refractive indices and the nonlinear optical coefficients. By performing Michelson interferometric and standard Maker Fringe measurements we are investigating these linear and nonlinear optical properties of the most promising organic crystals grown in our laboratory.

This year we focused on the determination of the nonlinear optical coefficients at telecommunication wavelengths and the possibility of efficient phase-matched optical frequency conversion in organic crystals of 4-N,N-dimethylamino-4'-N'-methyl-stilbazolium tosylate (DAST) and 5-(methylthio)-thiophenecarboxaldehyde-4-nitrophenylhydrazone (MTTNPH) developed in our laboratory. We measured the nonlinear optical coefficients at wavelengths of $1.5 \mu\text{m}$ and $1.3 \mu\text{m}$ and determined the resulting phase-matched figures of merit for both organic materials. In both materials phase-matching can be achieved for a wide wavelength range. At $1.3 \mu\text{m}$ the obtained figure of merit d_{eff}^2/n^3 of MTTNPH is 16 times larger than that of KNbO_3 . The figure of merit of DAST which is more than 30 times larger compared to KNbO_3 has been confirmed by measurements of phase-matched second harmonic generation at $1.3 \mu\text{m}$.



Single crystal of 5-(methylthio)-thiophenecarboxaldehyde-4-nitrophenylhydrazone (MTTNPH) with a size of $25 \times 5 \times 5 \text{ mm}^3$.

Reference: F. Pan, M.S. Wong, M. Bösch, Ch. Bosshard, U. Meier and P. Günter, *Appl. Phys. Lett.*, **71** (15), 2064 (1997)

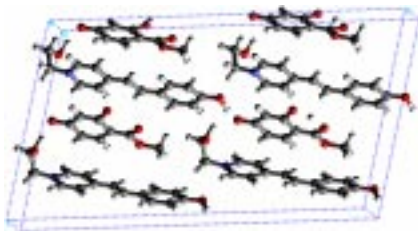
Sponsor: Swiss National Science Foundation

Electro-Optic Properties of Molecular Crystals

R. Spreiter and Ch. Bosshard

The electro-optic effect allows to control a light beam by an electric field, it therefore can be used as an interface between electronics and optics. Important applications are phase or intensity light modulators, electro-optical switches, electric field sensors and generation of THz radiation by optical rectification of fs light pulses.

Organic crystals are especially suited as high frequency, broadband electro-optic materials since the electro-optic effect is almost of pure electronic origin and they have a low dielectric constant. The most important requirements for a electro-optic crystal are a large nonlinearity of the constituent molecules and a preferably parallel, noncentrosymmetric arrangement in the crystal lattice.



Unit-cell of the co-crystal Mero-MDB as determined by x-ray diffraction. The chromophores are perfectly aligned parallel in the crystal lattice as required for electro-optics.

The technique of co-crystallizing a nonlinear chromophore with a guest molecule allows, by the use of short hydrogen bonds, to force crystallization in a arrangement perfect for electro-optics. An example of such a crystal is the highly nonlinear merocyanine chromophore (4-[2-[1-(2-hydroxyethyl)-4-pyridylidene]-ethylidene]-cyclo-hexa-2,5-dien-1-one) co-crystallized with MDB (methyl 2,4-dihydroxybenzoate).

Interestingly there exist a dark-red and a light-red phase of the crystal which have the same lattice parameters and symmetry with different linear, nonlinear and electro-optical properties. The electro-optic response was measured as 65% of the highly nonlinear crystal DAST (4-N,N-dimethylamino-4'-N'-methyl stilbazolium tosylate). This lower than theoretically expected effect is attributed to a distortion of the electron distribution caused by the strong hydrogen bonding between the molecules. High frequency measurements showed that the effect is of pure electronic origin. The wavelength dispersion of the electro-optic effect cannot be explained by a conventional model, usually appropriate for one-dimensional charge transfer molecules. Electro-absorption was shown to have its origin in the shift of the resonance frequency by the applied voltage (Stark effect). In conclusion, co-crystallization is a very promising tool for crystallizing highly nonlinear chromophores in a perfectly aligned way for electro-optics.

Reference: Man Shing Wong, Feng Pan, Martin Bösch, Rolf Spreiter, Christian Bosshard, Peter Günter, and Volker Gramlich, *J. Opt. Soc. Am.*, **15**, 426-431 (1998)

Sponsor: Swiss National Science Foundation

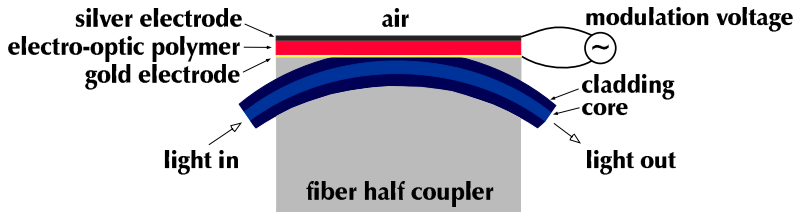
Electro-Optic Polymer Waveguide Modulators

M. Bösch, I. Liakatas, M. Jäger and Ch. Bosshard

Organic polymers have become an important class of nonlinear optical materials for electro-optic modulation. They have been the subject of intense research since they combine the nonlinear optical properties of conjugated π -electron systems with the feasibility of molecular engineering, i.e. creating new materials with appropriate optical, structural, and mechanical properties.

During the recent years the thermal relaxation of the nonlinear optical effect in poled side-chain polymers has been investigated in our group and provided a deeper understanding of the relaxation of the oriented chromophores. In the present project we concentrate on one hand on the application of our polymers as an electro-optic waveguide modulator. On the other hand we work on the development of new materials using chromophores with higher optical nonlinearity (see section 1.2) combined with polymer backbones having higher glass transition temperatures and therefore even better thermal stability.

This year we built a demonstration model of such an electro-optic polymer modulator based on a so called fiber half coupler consisting basically of a curved fiber incorporated into a glass block, polished down close to the core, to form an in-line fiber modulator. Such a modulator is sketched in the figure below. The light passing through the fiber is coupled into the polymer overlay depending on the matching of the effective refractive indices of fiber core and polymer. The refractive index of the polymer can be varied due to the electro-optic effect which can be used to build a very low loss modulator since no pigtailling and waveguiding through the polymer is necessary. The first modulators were tested and amplitude modulation was demonstrated at low frequency (up to 100 kHz).



Electro-optic in-line fiber modulator (principle).

Sponsors: Swiss Priority Program Optique II, ETH Zurich

Photorefractive Effects and Charge Transport Properties in Organic Crystals and Polymers

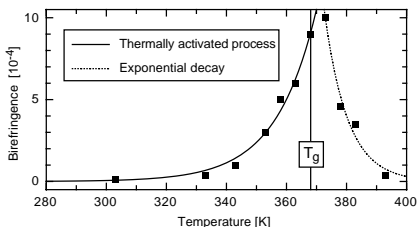
S. Follonier, Ch. Bosshard, I. Biaggio, work on polymers in collaboration with M. Döbler and U. Suter, Institute of Polymers, ETH Zurich

Organic crystals and especially polymers are of prime interest for low cost devices based on light-induced changes of refractive indices. Much progress was achieved in terms of photorefractive sensitivity as well as in the design of new compounds optimized for specific needs, like infrared enhanced photorefractive response. Nevertheless, the limits of organic materials have not been reached yet.

In order to improve the photorefractive sensitivity of organic crystals, the knowledge of charge carrier photogeneration and charge transport properties is of prime importance. Therefore, intrinsic parameters such as mobility and lifetime of charge carriers, quantum efficiency of carrier generation were investigated by DC photoconductivity, two-beam coupling as well as Time-of-Flight experiments in 4-N,N-dimethylamino-4'-N'-methylstilbazolium toluene-p-sulfonate crystals.

The project succeeded in determining values of the recombination time of the charge carriers of $\tau = 4 \pm 2$ ns, of the drift mobility of the order of $\mu = 1 \text{ cm}^2/\text{Vs}$ and of a quantum efficiency of less than 10^{-3} at $\lambda = 750$ nm. Thus, the most important factor limiting the photorefractive sensitivity in DAST is the small efficiency of charge carrier photogeneration and appropriate doping of DAST could lead to orders of magnitude improvement of its photorefractive sensitivity.

In the field of polymeric materials, field-induced orientational processes have been accepted as the main contribution to the overall light-induced refractive index changes. Stabilized temperature dependent two-beam coupling experiments were performed on films of polyamides based on DDANS doped with 20% DEH (high glass transition temperature T_g) to determine the ideal working temperature relative to T_g for enhanced field-induced orientational contributions (see figure).



Light-induced birefringence as a function of temperature measured in the Bragg diffraction experiment in a photorefractive polymer.

Sponsors: Swiss National Science Foundation

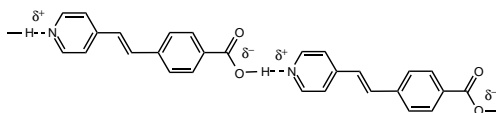
Design, Preparation, and Characterization of Novel Organic Nonlinear Optical Thin Films Grown by Molecular Beam Deposition

C. Cai, Y. Tao, M. Bösch, A. Kündig, B. Müller and Ch. Bosshard

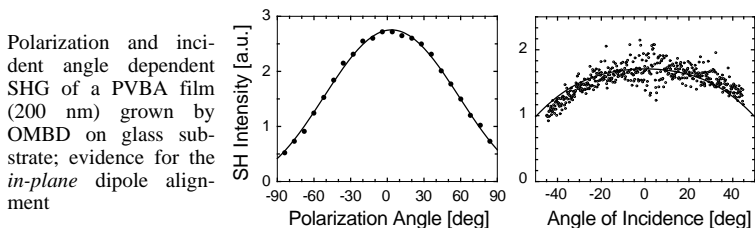
Organic thin films with second order nonlinear optical properties hold promise for applications in fast optical data processing. However, their fabrication has been hampered due to the difficulties in aligning the dipolar molecules non-centrosymmetrically. In addition, their thermal stability is often low.

We have developed novel chromophores which can self-assemble in a head-to-tail fashion via a strong hydrogen bond network. The network serves to align the dipoles and to increase the melting point of the material. PVBA is a typical example of these chromophores. The solid state ^{15}N -NMR spectra of PVBA prove the strong intermolecular hydrogen bonding between the carboxylic H-atom and the adjacent N-atom. As expected, PVBA has a high melting point (350° C).

PVBA {4-[trans-2-(4-pyridyl-vinyl)]benzoic acid} molecules linked via a strong hydrogen bonding



PVBA films with a thickness of 10–2000 nm have been grown on different bare and functionalized substrates (glass, quartz, sapphire, silicon, and indium-tin-oxide) by organic molecular beam deposition (OMBD) under different conditions (deposition rate: 0.01–1 nm/s; substrate temperature: 30–175° C). The films are characterized by a variety of methods. In particular, second harmonic generation (SHG) shows that the films grown on bare glass, quartz, and sapphire have an effective nonlinear optical coefficient of ~ 1 pm/V, stable up to 190° C. Moreover, the average dipole of PVBA on glass is found to be parallel to the projected molecular beam line (incident angle: ca. 20°) on the substrate surface. Therefore, we have demonstrated that *in-plane* dipole alignment on glass substrates can be achieved by OMBD.



Sponsor: Swiss Priority Program "Optics II"

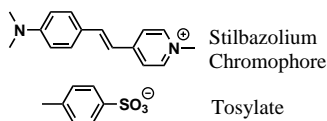
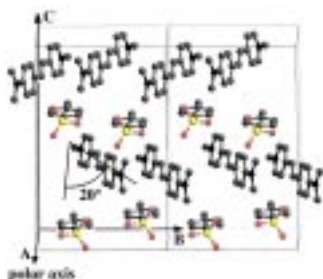
Fluorescence Phenomena in Organic Crystals

U. Meier and Ch. Bosshard

There has been considerable effort during the last years in developing new organic materials with optimized properties for electro-optics, frequency conversion and photorefractive applications. The nonlinear optical properties of these materials originate in the molecular electronic response of the material to applied fields (electrical or optical), modified by the influence of molecular surroundings (crystal structure, solvents). Theoretical descriptions of the optical properties are therefore based on the energy configuration of the electronic states of molecules and crystals. Since some of our newly grown organic crystals show strong fluorescence after excitation with IR or UV, the aim of this project is to investigate the intra- or intermolecular physical processes leading to the observed excitation and emission phenomena of these organic materials.

To determine the relevant underlying processes (intramolecular charge transfer, intermolecular excitons (electrons & holes), ...) which lead to the observed one and two photon excited fluorescence phenomena we perform different absorption and luminescence experiments. The dependence of luminescence and absorption upon temperature, excitation intensity, polarization and excitation wavelength are measured on organic crystals and on molecules in solution.

The most interesting material at the moment is 4-N,N-dimethylamino-4'-N'-methyl-stilbazolium tosylate (DAST). Absorption and luminescence bands of DAST are well separated (in solution 150 nm, in the crystal 90 nm). The emission spectra of DAST crystals are split up by ≈ 0.16 eV for one and two photon excitation, leading to a broad emission range. Two photon excitation at $1.064 \mu\text{m}$ has been verified by the quadratic intensity dependence of the emission intensity. The strong luminescence anisotropy can be correlated to the crystalline packing of DAST single crystals, indicating that the stilbazolium part is the active fluorescent moiety of DAST.



Chemical structure and crystal packing of the polar organic salt 4-N,N-dimethylamino-4'-N'-methyl-stilbazolium tosylate (DAST)

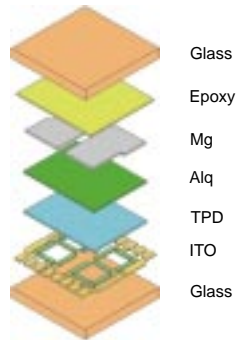
Sponsor: Swiss National Science Foundation

Fabrication and in situ Characterization of Multilayer Organic Light Emitting Devices

Y. Tao, A. Kündig, C. Cai, I. Gamboni and B. Müller

Organic light emitting devices (OLEDs) are promising candidates for the next generation flat-panel displays. The most advanced OLEDs are multilayer devices prepared by vacuum sublimation, with the different layers possessing specialized charge carrier transport and optical properties to provide reduced charge injection barriers, balanced charge injection and better carrier confinement. Due to the charge injection characteristic, the performance of OLEDs is very sensitive to the properties of electrode/organic interfaces, such as the band-bending, barrier height, and Fermi-level position. Therefore it is of great interest to investigate and control the interface between the electrodes and organic layers under well-defined conditions.

We have fabricated double layer OLEDs by organic molecular beam deposition under ultra-high vacuum (UHV) condition. A typical layer sequence for our seven-segment numerical displays is shown at right. In this device, TPD (N,N'-diphenyl-N,N'-bis(3 methylphenyl)-1,1'-biphenyl-4,4'-diamine) was used as hole transporting material and Alq (tris(8-hydroxyquinolate)aluminum (III)) as emissive and electron transporting material. The cathode is made of Mg. Each segment has an emissive area of 20 mm². The turn-on voltage of these diodes ranges from 4 to 12 volts depending on the thickness of organic layers. The emission of the electroluminescence is peaked at 530 nm (yellow green).



Operating at 12 volts, the brightness of these devices is 550-600 cd/mm² at a current density of 20 mA/cm², which is about 5 times as bright as a computer display.

In situ x-ray photoelectron spectroscopy (XPS) and ultra-violet photoelectron spectroscopy (UPS) have been used to investigate the formation of Mg/Alq and Ag/Alq interfaces in UHV. We have discovered that Mg reacts with the nitrogen of Alq molecules during the first 10 nm deposition. Due to the charge transfer between Mg and N, a down-ward band-bending occurred during the first a few monolayers deposition of Mg, and the electron injection barrier is close to zero at this interface. A metallic Mg layer is formed after deposition of more than 16 nm Mg on Alq. This result shows that strong interdiffusion occurs at Mg/Alq interface during cathode fabrication. Furthermore, we have demonstrated that the interdiffusion is avoided by predepositing 2~3 monolayers of Ag on Alq, and thus a sharp interface between the Mg and Ag/Alq can be formed.

Sponsor: ETH Zürich

Tunable Nanosecond and Picosecond Infrared Lasers Based on Optical Parametric Generation in KNbO_3

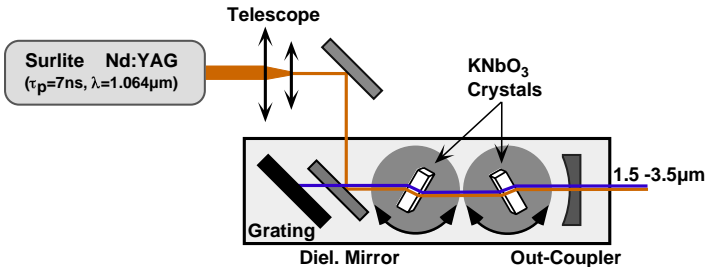
U. Meier, U. Gubler, I. Biaggio and Ch. Bosshard

For the spectroscopic investigations of the optical and nonlinear optical properties of our new materials, a laser source tunable over a wide range of wavelengths in the near infrared is required. The use of optical parametric generation and amplification of light in a nonlinear optical crystal is most attractive to provide such laser radiation.

Within this project we use KNbO_3 as a nonlinear optical material, because of the wide phase matching possibilities, its large effective nonlinear optical coefficients, and its large optical damage threshold. We focus on two different concepts. (i) An optical parametric oscillator (OPO) system for ns pulses, which is directly pumped by a Nd:YAG laser (7 ns) at 1064 nm and uses a two crystal walk off compensated geometry. (ii) An optical parametric generator / amplifier (OPG/OPA) system for ps pulses pumped by an amplified Nd:YLF laser (10 ps) at 1047 nm. Wavelength selection for both systems will be done by angle tuning of the KNbO_3 crystals.

The setup for the ps system was optimized by reducing the number of parts and facilitating the alignment of the beams, concurrent with a reduction in size.

The development of the ns OPO was delayed because the operation threshold couldn't be reached using our 15 mm long KNbO_3 crystals. The determination of the nonlinear optical coefficients for the wavelength range of importance, in consequence, revealed that the nonlinear optical coefficients in KNbO_3 differ significantly from the values estimated by using well-known Miller law (measured values are 40% lower at 1907 nm). These lower values of the nonlinear susceptibilities lead to a much higher threshold for optical parametric oscillation. We hope that the decreased overall losses in our newest crystals and in the optical system will compensate the effect of the lower nonlinear susceptibilities.



KNbO_3 walk-off compensated ns optical parametric oscillator system pumped at 1064 nm.

Sponsors: Swiss Priority Program Optique II

PHOTOREFRACTIVE EFFECTS AND PHOTONICS

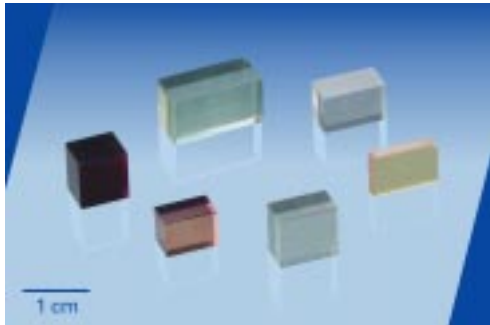
Growth, Preparation and Characterization of Visible and Infrared Sensitive KNbO_3 Crystals

H. Wüest, J. Hajfler, C. Medrano, R. Ryf and M. Ewart

The performance of a photorefractive material for holographic recording is characterized by a figure of merit, the photorefractive sensitivity, which is related to the rise time of the photoinduced refractive index change. Our present material 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 in the range where compact diode lasers are available.

Last year we significantly increased the sensitivity of KNbO_3 doped with various elements by applying a new gas-reduction method to the whole crystal boules immediately after growth. This technique has been perfected further and applied to additional dopants. Crystal quality was characterized by photorefractive techniques, as well as with optical and electrical measurements and chemical analysis.

A thermodynamic point-defect model was now used to analyze the high-temperature gas-reduction treatment, obtaining a good understanding of the reduction process and a good correlation between the experiment and the photorefractive properties predicted from the reduction parameters and the dopant levels. We also experimentally demonstrated the superior spatial homogeneity of the photorefractive



Pure, Rh- and Fe doped KNbO_3 crystals prepared in the nonlinear optics laboratory

properties of gas-reduced samples when compared to earlier reduction techniques. A new batch of reduced Rh doped KNbO_3 samples was produced and their large sensitivity in the near IR was shown to be reproducible. Reduced crystals co-doped with Mn and Rh were found to have similar performance, i.e. a $\sim 7 \text{ cm}^{-1}$ gain coefficient and a $\sim 4 \text{ s}$ response time at 1 W/cm^2 for counterpropagating beams at $\lambda = 860 \text{ nm}$. Reduced rhodium doped crystals were also shown to exhibit self-pumped phase conjugation at 860 nm with a reflectivity exceeding 15%

Reference: M. Ewart, R. Ryf, C. Medrano, H. Wüest, M. Zgonik, P. Günter, *Opt. Lett.* **22**, 781 (1997).

Sponsor: Swiss National Science Foundation

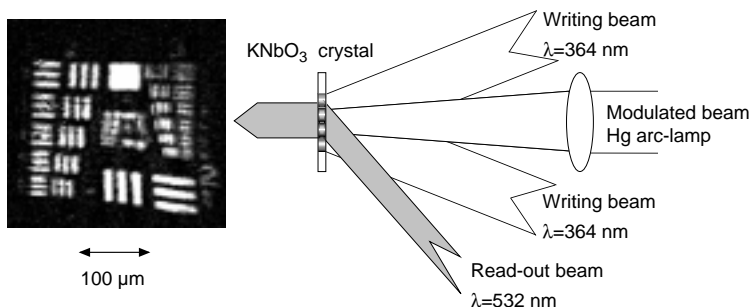
Ultraviolet Photorefraction and its Applications

P. Bernasconi

The recent thorough studies of photorefractive gratings induced by interband photoexcitation showed that strong phase gratings with short response times can be recorded in undoped KNbO_3 . By combining them with the higher resolution inherent to ultraviolet (UV) light, these excellent qualities have been used to set-up an Interband Photorefractive Incoherent to Coherent Optical Converter (IPICOC) operating at short wavelengths and compatible with video rates. In parallel to these quasi-thin grating, the investigations on photorefractive effect in UV transparent materials have been continued with the aim of obtaining sensitive media for phase conjugation and beam manipulation. LiTaO_3 crystals grown with different stoichiometry have been studied and found to give promising results.

The transfer of images from an incoherent to a coherent light beam occurs within a thin KNbO_3 pure crystal ($47\ \mu\text{m}$) where an interband diffraction grating is previously written. The image carried by the incoherent beam partially erases the hologram which is read-out by a coherent beam in real time. The diffracted wave is contrast inverted with respect to the original incoherent image.

In an anisotropic Bragg diffraction configuration, we obtained a resolution in the crystal as high as $120\ \text{lp/mm}$ with a response time of $\sim 30\ \mu\text{s}$ for a UV intensity of the order of $100\ \text{mWcm}^{-2}$, results well beyond the required performances. The diffraction efficiency reached 0.4%. Besides one of the highest resolutions, these results give an optical switching energy per bit of $\sim 1\ \text{pJ}$, a value which competes well with the best optically addressed spatial light modulators available. The diffraction efficiency can be improved by applying an electric field in an isotropic configuration, however in this case resolution and speed are slightly decreased.



Set-up and resolution of an Interband Photorefractive Incoherent to Coherent Optical Converter (IPICOC) and typical resolution achieved in these experiments

Reference: Y. Furukawa, K. Kitamura, Y. Ji, P. Bernasconi, G. Montemezzani, P. Günter, CLEO, Baltimore, May 18-23, 1997, Technical Digest, pp. 120-121.

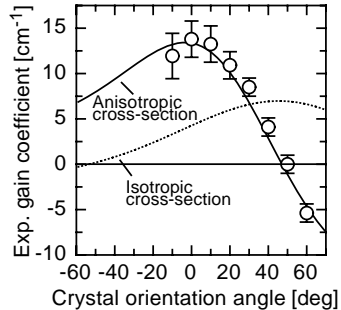
Fundamental Characterization of Photoconductivity by Photorefractive Holography

G. Montemezzani, P. Bernasconi, I. Biaggio and C. Medrano

The understanding of photoexcitation and 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 based on these materials. In this project we develop and apply all-optical holographic methods to characterize fundamental physical properties related with each individual step involved in the photorefractive redistribution of charges, i.e. charge carrier photoexcitation, carrier transport, and carrier recombination.

Our investigation methods are based on a combination of continuous wave (cw) and pulsed real time holography. Two-wave mixing experiments with cw lasers give information on the characteristics of the charge photoexcitation process. The holographic time-of-flight (HTOF) method, based on short pulse excitation, allows the measurement of charge carrier mobilities even when the free carrier lifetime is only of the order of 1 ns. Finally, interband photorefractive experiments induced by ultraviolet light can be used to compare the trap-limited mobilities of electrons and holes and to characterize charge recombination in oxide crystals.

By cw-experiments we could demonstrate that the anisotropy of the photoexcitation cross section with respect to light polarization has dramatic consequences on photorefractive two wave mixing (see Figure) confirming a new model developed during this year. This effect can be used to optimize the exponential amplification gain coefficients. Observing the grating evolution during several decades after the 70 ps recording pulses in HTOF experiments allowed us to estimate the still unknown hole drift mobility in KNbO_3 to be $\mu_h = (0.006 \pm 0.003) \text{ cm}^2/\text{Vs}$. Interband cw-photorefractive experiments showed that in an undoped KNbO_3 sample the trap limited hole mobility exceeds the trap limited electron mobility by 30%, resp. 100% for drift directions along the c resp. b crystallographic axis.



Influence of the anisotropy of the photoexcitation cross-section on two-wave mixing gain

Reference: G. Montemezzani, C. Medrano, M. Zgonik, P. Günter, *Phys. Rev. Lett.* **79**, 3403 (1997).

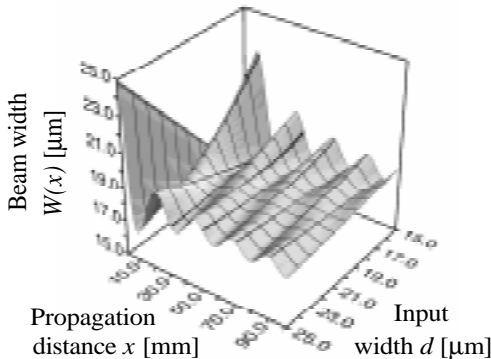
Sponsor: Swiss National Science Foundation

One- and Two-Dimensional Self-Focusing and Spatial Solitons in Photorefractive KNbO₃

R. Ryf and G. Montemezzani

Nonlinear optical interaction can lead to beam reshaping and self-focusing in appropriate nonlinear materials. If the self-focusing effects compensates the natural diffraction of a light beam, light induced waveguides and spatial solitons can be generated. These processes are particularly interesting in photorefractive materials because of the low optical power required for this kind of optical nonlinearity. Reconfigurable waveguides and optical elements such as X- and Y-junctions may be created by the effect of the propagating beams themselves.

Our investigations of self focusing effects and spatial solitons in photorefractive media were pursued along three intercorrelated lines: (1) systematic experimental characterization of spatial solitons and laser beams self-focused in one or two transverse dimensions in KNbO₃ crystals; (2) Theoretical description of the soliton profile and its dependence on the important experimental parameters, i.e. applied electric field and beam intensity; (3) Numerical simulation of nonlinear beam propagation on a supercomputer in order to describe the spatial convergence from the input to the soliton profile and the stability of the latter.



1-dimensional soliton formation for different Gaussian input beam widths

In 12 mm long KNbO₃ samples, non-diffracting beams could be observed for waves confined in one or both dimensions transverse to the propagation direction. For the one-dimensional case, the observed widths of bright spatial solitons are found to be in good agreement with new analytic expressions for the soliton profile based on a power-series-like development. The comparison of experiments with numerical simulations allows the determination of optimum launching conditions and

permits to conclude that, even by starting with a Gaussian beam profile, the beam width can be kept within 3% from the asymptotic soliton width for all propagation distances. The numerically predicted strongly astigmatic nature of 2D photorefractive bright solitons could be confirmed experimentally.

Reference: G. Montemezzani and P. Günter, *Opt. Lett.* 22, 451 (1997).

Third-Order and Second Order Nonlinear Optical Processes in Degenerate Four-Wave Mixing

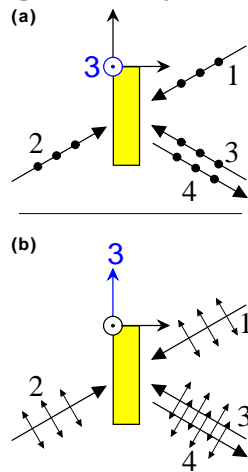
I. Biaggio

Nonlinear optical effects are described by a material polarization that depends on the 2nd and higher powers of the electric field. Examples of 2nd order processes are second harmonic generation and the electro-optic effect. Some third order processes are third harmonic generation, self- and cross-phase modulation, and degenerate four-wave mixing (DFWM). They can mediate a very fast interaction between optical waves, with interesting applications for fast all-optical switching.

New material research directions are given by the fact that 2nd order effects can contribute greatly to the third order polarization by a two-step (cascading) process. We demonstrated that cascading of optical rectification and the electro-optic effect often overshadows the direct third-order contribution in DFWM, which is a standard technique for determining the third order susceptibility. It is important to note that its use in non-centrosymmetric materials always leads to incorrect results if cascading contributions are not properly taken into account. This is because the cascaded contribution is a non-local process that depends on geometrical boundary conditions (direct third order effects are well described as a local effect).

The figure shows two ways to measure the coefficient χ_{3333} of the third order susceptibility tensor of BaTiO_3 (point group $\text{mm}4$). For configuration (a) the induced 2nd order nonlinear optical polarization has zero divergence and non-zero curl, and the cascaded contribution is 2 times larger than in (b), where the polarization has non-zero divergence. (a) has a larger effective third order susceptibility despite the fact that for both configurations all beam polarizations are the same in the sample reference frame. By comparing the four-wave mixing signal for the two configurations shown in the figure and using the calculated values of the cascaded contributions, we were able to determine absolute third-order susceptibility values in BaTiO_3 , without having to rely on any other reference material. We are currently investigating the same technique in KNbO_3 .

Sponsor: Swiss National Science Foundation



The two DFWM configurations for measuring χ_{3333} . For clarity, the angles between the beams are exaggerated. One configuration goes over into the other one by rotating the crystal and all beam polarizations by 90° . Cascaded contributions can differ by a factor 2 between (a) and (b).

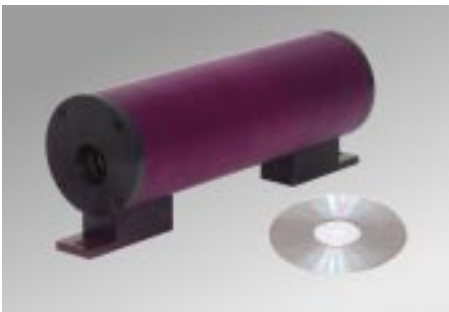
Stimulated Brillouin Scattering and Phase Conjugation at 2.1 μm

C. Medrano, M. Ehrensperger and H. Wüest

High power lasers at a wavelength of 2.1 μm are very interesting for many applications because they operate in the eye safe spectral region. Potential applications range from medicine to remote sensing, range finding, and spaceborne lidar systems. Because of the high energy pulses of these laser systems, a phase conjugating mirror has to be used to compensate for beam distortion induced by the amplifying medium.

The main aims of this research are (1) to optimize a cleaning and purifying technique for producing stimulated Brillouin scattering cells to be used as phase conjugate mirrors, and (2) to characterize the phase conjugate mirror for stability and long time operation, as well as for its performance. The long time operation stability strongly depends on the breakdown threshold of the Brillouin active liquid, as well as on its chemical purity and the size and number of microparticles it contains. The phase conjugation performance was investigated using a 2.1 μm Q-switched Cr,Tm,Ho:YAG laser built in our laboratory. It typically delivers 80 ns long pulses at an energy of 90 mJ and a repetition rate of 2 Hz. SnCl_4 was selected as active medium because of its high resistance to optical breakdowns and its chemical stability. We developed a special technique that allowed us to produce SBS-cells with less than 500 particles/cm³ (estimated size < 50 μm). An appropriate holder to protect the SBS cell from mechanical shocks (see figure) was also designed and tested.

The performance of different cells was found to be similar with a maximum phase conjugation threshold of 30 mJ and an average phase conjugate reflectivity of 35% at a pulse energy of 50 mJ. Thermal stability was tested at 100 °C during at least 150 hours, and the number of microparticles after 1000 breakdowns was less than 10000 per cm³. After these tests, no changes were observed in the reflectivity or in the energy threshold for phase conjugation.



Stimulated Brillouin Scattering cell for phase conjugation at 2.1 μm using SnCl_4 .

Sponsor: European Space Agency, ESTEC

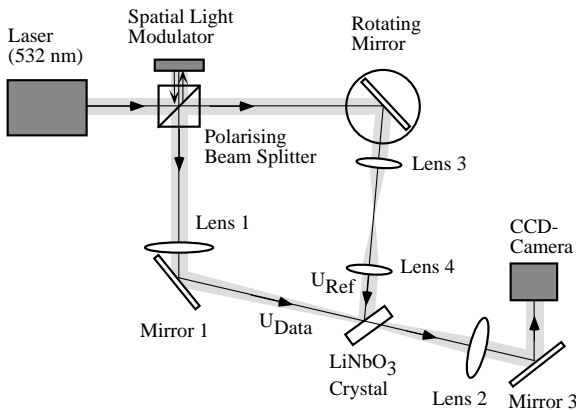
Holographic Optical Processing and Memories

R. Ryf

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. Combining the advantages of holographic memories with optical parallel processing techniques it is possible to obtain dedicated computing elements which are much faster than traditional electrical ones. The aim of this work is to build a fast readout holographic storage system. The system produces a high frame-rate image sequence, which will be used in a second step, to feed an optical processor (for example a high speed correlator).

In our approach we are using a Q-switched laser with a wavelength of 532 nm and a pulse length of 70 ns as laser source for holographic recording. The stored data are modulated on the object beam by means of a spatial light modulator. LiNbO_3 is used as long term storage material.

The optical part of the holographic memory system according to the set-up shown below has been constructed. Currently we are using a ferroelectric liquid crystal spatial light modulator with 256×256 pixels. After synchronisation of its electronic with the mechanical rotation of the galvano-mirror we expect to be able to demonstrate read out of a 100 image memory in 0.1 seconds.



Set-up for holographic fast readout frame-rate memory

Sponsor: Swiss National Science Foundation

POLAR ORGANIC AND FERROELECTRIC SURFACES

Atomic Force Microscopy on Organic Crystals

Ch. Seuret

Organic crystals used in nonlinear optics were investigated by atomic force microscopy (AFM). In comparison with optical techniques employed to characterize the particularities and the quality of the crystals, the scanning techniques offer the advantages to observe local effects.

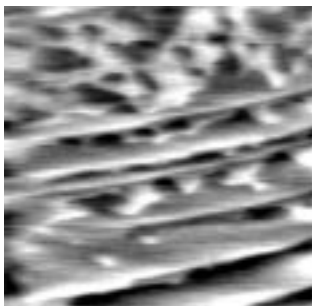
Two organic crystals were investigated: COANP (2-cyclooctylamino-5-nitropyridine) which presents a single layer structure and DAST (4-N,N-dimethylamino-4'-N'-methyl-stilbazolium tosylate) which possesses a double layer structure. The interlayer forces are mainly of van der Waals type in COANP crystals, and ionic in DAST.

The results of the investigations show mainly that for both crystal types a high attractive force (10^{-4} N) interacts between the AFM tip and the sample. This effect comes from the Coulomb interaction between localised charges in the dielectric crystals and the conductive tip.

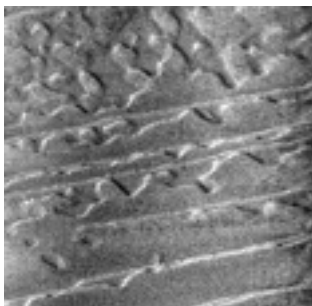
High resolution and layer resolution AFM images were achieved, from which the unit cell parameters could be deduced. The figures below show the double layer structure of DAST; the layer of tosylate disappears after 1-2 hours in contact with the air humidity.

The AFM and the force spectroscopy techniques allow the observation of an ionic compensation effect on the organic crystals surfaces.

AFM topography image



AFM lateral force image



Freshly cleaved surface of a DAST crystal. Both monomolecular layers are observed; the tosylate ions evaporate (bended contours), while the stilbazolium molecules (straight contours) remain stable.

Sponsor: Swiss National Science Foundation

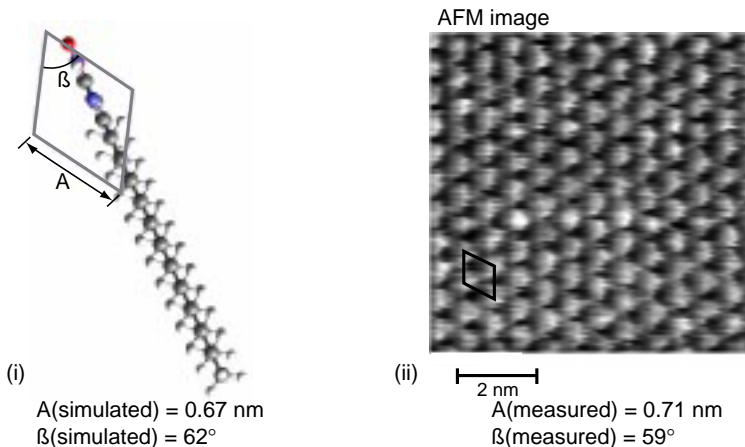
Hyperpolarizability Measurements and Calculations of Langmuir-Blodgett Molecules and Force Fields Simulations of Langmuir Films

Ch. Seuret

Hyperpolarizabilities of Langmuir Blodgett molecules were determined experimentally by EFISH (electric field induced second harmonic generation) and a theoretical prediction was performed by semi-empirical quantum chemistry calculations (MOPAC).

The results show that the hyperpolarizabilities of C218 (4-(N-octadecylamino)-4'-nitrostilbene) at 1907 nm is $230 \cdot 10^{-40} \text{ m}^4/\text{V}$, what is 10 times larger than that of DCANP (2-docosylamino-5-nitropyridine); C218 produces homogeneous monolayers. The predicted hyperpolarizabilities are lower than those measured by about 5% for small chromophores and up to 40% for molecules with two carbon-rings.

The Langmuir monolayer structures were simulated theoretically with a force field technique. The configurations of the LB molecules, MOPAC optimized, are arranged in a 2-dimensional crystal cell and minimized with the Dreiding force fields. The parameters of the simulated LB-films are in good agreement with the measured values; the unit cell size and the chain deflection angle of minimum calculated states and of the empirical models are comparable.



The 2-dimensional unit cell of the DCANP film simulated by the force fields technique (i) and measured by AFM at the air-solid interface (ii).

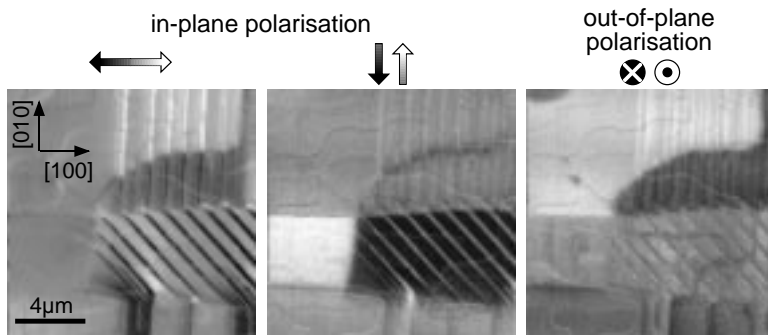
Sponsor: Swiss National Science Foundation

Imaging of Ferroelectric Domains with a Scanning Force Microscope

M. Abplanalp

The features of ferroelectric materials are determined to a major extent by the static and dynamic properties of ferroelectric domains. A non-destructive method sensitive only to the polarisation at the surface is needed to study the nucleation, growth and stability of domains at the surfaces of ferroelectric crystals. Also the method should offer a high lateral resolution, be sensitive to polarisations of any direction and permit to image the surface morphology.

All these requirements are fulfilled by voltage modulated scanning force microscopy. The polarisation is measured via the inverse piezoelectric effect: an ac-voltage applied to the conductive scanning force microscope tip leads to a local distortion of the crystal. A polarisation perpendicular to the surface (out-of-plane) results in periodic contraction and expansion normal to the surface while polarisations parallel to the surface (in-plane) cause a periodic lateral shearing of the crystal. Since the tip is in contact with the surface, the tip will follow these movements bending the cantilever. By measuring the amplitudes of the cantilever vibrations normal and parallel to the surface by phase sensitive amplifiers, three voltages are generated, each representing one component of the polarisation vector. The image below shows these three voltages for a barium-titanate crystal.



Measured components of polarisation for a barium-titanate crystal. The crystal is oriented in such a way that the polarisation vector is always parallel or antiparallel to one of the measured components resulting in a black or white contrast. The other two components then have to vanish at that spot, showing as a grey contrast.

Sponsor: Swiss National Science Foundation, ETH Zürich

Characterization of Ferroelectric Domain Switching on the Nanometer Scale

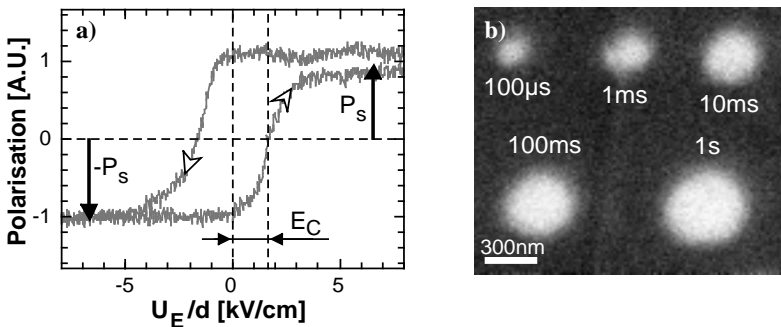
M. Abplanalp and L. Eng

Hysteresis loops of ferroelectric crystals are generally measured by applying a cyclic voltage to macroscopic electrodes situated on the opposite faces of the crystal and by recording the displacement current. The gained polarisation, however, represents a spatial average over the crystal.

In order to investigate the involved processes, the domain reversal upon switching of nanoscaled ferroelectric domains has to be characterized. Namely, one of the macroscopic electrodes is replaced by the conductive tip of the scanning force microscope. Since the displacement current measures as low as 1 fA due to the very small area exposed to domain reversal, the polarisation below the tip was measured by voltage modulated scanning force microscopy.

Figure (a) presents the result of nanoscale domain switching as recorded on ferroelectric bulk single crystals of barium-titanate. The coercive field measures $E_C = 1700$ V/cm which is more than twice as large as for macroscopic switching using extended electrodes. This may be attributed to the energy required to initiate nucleation of ferroelectric domains at the sample surface.

Once the domain has nucleated, it grows laterally. This effect can be demonstrated by applying an electric field of $1.5 \cdot E_C$ to the crystal for various lengths of time. As can be seen in figure (b) the domains grow bigger the longer the electric field has been applied. The minimum domain diameter up to now is 150 nm.



(a) Hysteresis loop of a barium-titanate crystal of thickness $d = 125 \mu\text{m}$ measured with a scanning force microscope tip as a top electrode while cycling the voltage U_E on the counter-electrode; (b) domain pattern produced by applying voltage pulses of various durations to the counter electrode with the tip placed at different positions.

Sponsor: National Research Program NFP 36 “Nanowissenschaften”

Ultra-High Vacuum Atomic Force Microscopy of Surfaces Prepared by Molecular Beam Deposition

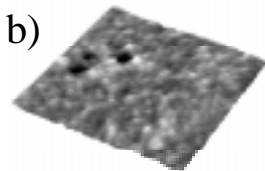
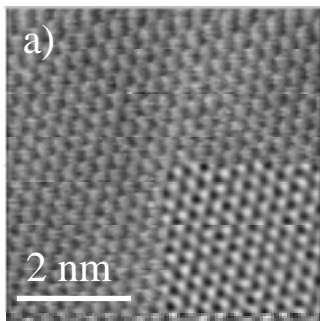
A. Kündig, I. Gamboni, Y. Tao, C. Cai and B. Müller

Atomic force microscopy (AFM) is a powerful tool to investigate growth processes and to characterize the surface morphology of thin films grown by molecular beam deposition in ultra-high vacuum (UHV). It allows *in situ* measurements of surface roughness, island shapes, island sizes, and island densities of the deposit.

Organic thin films usually cannot be characterized in contact mode, because the forces applied to the organic films are too high resulting in film scratching. The interaction between the tip and sample is drastically reduced in non-contact mode. Here, the friction forces are completely eliminated. In this imaging mode, the cantilever is vibrated at its mechanical resonance frequency. The interaction between the tip and sample adds an external force gradient to the intrinsic force constant and thus changes the cantilever's eigenfrequency. With a frequency detector the resonance shift can be measured. This signal is used to regulate the sampling distance in order to record a surface map of constant force gradient.

Our unique UHV-AFM has been improved in contact mode, demonstrated by atomic resolution on MoS₂ (Fig.a). For measurements on organic thin films, we upgraded our system with a frequency demodulator in order to carry out measurements in non-contact mode. The performance of our instrument is shown by resolution of atomic steps on graphite and MoS₂.

In addition, we have studied the surface roughness of indium-tin-oxide (ITO) substrates for organic light emitting devices (OLEDs) for different surface treatments with the aim to improve the lifetime of OLEDs. The active organic layer (tris(8-hydroxyquinolate)aluminium (III)(Alq)) is found to be very smooth. The RMS roughness corresponds to 0.3 nm (Fig.b).



a) Atomic lattice resolution on MoS₂ in constant height mode (6x6nm²), the lower right-hand quarter of the image is shown after filtering
b) Morphology of the active layer (Alq) of an organic light emitting device (8x8 μm²)

Sponsor: Swiss Priority Program Optique II

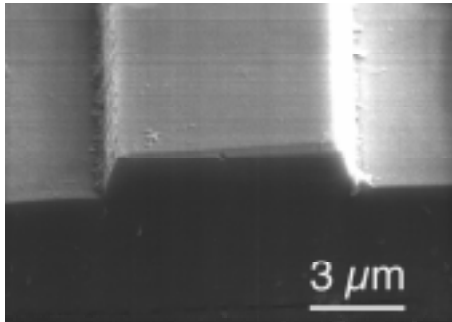
INTEGRATED OPTICS AND OPTICAL FREQUENCY CONVERSION

Second-Harmonic Generation in KNbO₃ Waveguides

T. Pliska and D. Fluck, in collaboration with Ch. Buchal and Lutz Beckers, Forschungszentrum Jülich, Germany, and H. Melchior and E. Gini, ETH Zürich

Blue light generation by nonlinear optical interactions, such as second-harmonic generation (SHG) with diode lasers (e.g. AlGaAs and InGaAs), is an attractive approach to realize a compact all solid-state blue laser. Potassium niobate (KNbO₃) is a particularly suited material for nonlinear frequency conversion.

The use of channel waveguides avoids the problem of walk-off for any of the possible phase-matching configurations because both waves are inherently guided along the same direction. We have developed a fabrication process on the basis of ion implantation and ion sputtering to prepare high-quality ridge waveguides in KNbO₃. The channel waveguides with typical cross sections of 5 x 5 μm fabricated by this process guide modes of both TE- and TM-polarizations which is required for phase-matched SHG by natural birefringence. We have demonstrated phase-matched SHG in waveguides between 870 and 980 nm. The figure below shows the polished front-face of a KNbO₃ ridge waveguide.



Scanning electron microscope image of a KNbO₃ ridge waveguide. The ridge width is $w = 7.6 \mu\text{m}$, and the ridge height $h = 1.3 \mu\text{m}$.

Noncritically phase-matched SHG in a 7 mm long waveguide yielded 14 mW of blue power at 438 nm with a fundamental power of 340 mW coupled into the waveguide with a launch efficiency of about 80 %. This is the highest conversion efficiency reported for birefringence phase-matched SHG in a waveguide demonstrating the suitability of these KNbO₃ waveguides for efficient SHG to blue wavelengths. We estimate that in optimized waveguides more than 30 mW of blue light can be generated by SHG of 300 mW near-infrared diode lasers.

Reference: T. Pliska, et al., *J. Appl. Phys.* **81**, 1099 (1997).

Sponsors: ETH Zürich, Swiss Priority Program Optique II

Control of the Photorefractive Effect in Fe-doped KNbO_3 by MeV Ion-Implantation

S. Brülisauer and D. Fluck, in collaboration with Ch. Buchal and L. Beckers, Forschungszentrum Jülich, Germany

Photorefractive crystals such as KNbO_3 , BaTiO_3 , LiNbO_3 , and $\text{Sr}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$ are very promising for all-optical signal processing. We have demonstrated for the first time, that it is possible to extend the photorefractive response to the telecommunication wavelengths of 1313 and 1550 nm by using high energy (3 MeV) proton irradiation. The major advantage of ion-irradiated ferroelectric oxide waveguides is that charge diffusion is sufficient to build-up very high gains, and therefore, no external electric fields have to be applied in contrast to photorefractive semiconductor devices.

This year we successfully demonstrated the controlled and reproducible adjustment of the photorefractive properties such as gain and response time in Fe-doped KNbO_3 by using the technique of high energy proton implantation. The change of the photorefractive properties is caused by a change of the ratio of $[\text{Fe}^{2+}]/[\text{Fe}^{3+}]$ in the irradiated region due to the production of oxygen vacancies during proton-irradiation. The reduction ratio $R = [\text{Fe}^{2+}]/[\text{Fe}^{3+}]$ can be precisely adjusted over a very large range from $R = 0.01$ up to $R = 40$, unmatched by former reduction methods, by simply choosing the appropriate irradiation dose. For example, we were able to alter the photorefractive gain from 2.5 cm^{-1} to 34 cm^{-1} , the absorption constant from 0.06 cm^{-1} to 2.1 cm^{-1} and the buildup time from 1.6 ms down to $34 \mu\text{s}$ at an intensity of 200 W/cm^2 , corresponding to a power of 2.5 mW in the interaction region, in a Fe-doped KNbO_3 crystal at a wavelength of 514.5 nm. We developed a refined theoretical model for the photorefractive effect in strongly reduced KNbO_3 , and derived a set of basic material parameters, which completely explains all the measurements. This is an important step towards the control of the photorefractive effect, which is of crucial importance for the realization of applications.

Reference: S. Brülisauer, D. Fluck, and P. Günter, "Photorefractive effect in proton-implanted Fe-doped KNbO_3 waveguides at telecommunication wavelengths", *J. Opt. Soc. Am. B* **13**, 2544 (1996).

Sponsors: ETH Zürich

Liquid Phase Epitaxy of $K_{1-y}Na_yTa_{1-x}Nb_xO_3$ (KNTN) ($x=0.45, y=0.10$) Thin Films

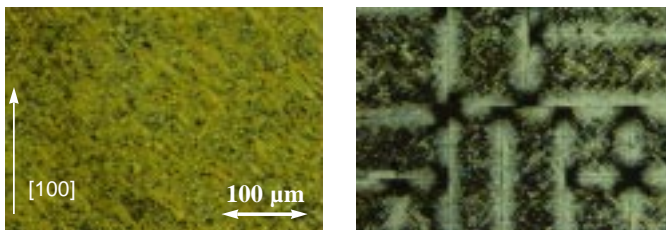
H. Pierhöfer and H. Wüest

Fast electro-optical (EO) waveguide modulators and switches are of considerable interest for all optical signal processing. Ferroelectric thin films such as KNTN are very promising for EO modulation due to their high figure of merit.

Thin films of KNTN are grown by liquid phase epitaxy (LPE). The ratio of Nb to Ta ions varies the transition temperature from the ferroelectric to paraelectric phase from 0 K (only Ta ions) to about 700 K (only Nb ions). For a ratio of $x=0.45$, KNTN shows a phase transition temperature of 333 K (60 °C) and thus high electro-optic and pyroelectric effects at room temperature.

$KTaO_3$ (KT) is used as substrate for LPE of KNTN. A fairly small amount of Na in KNTN (which partially replace the K ions) ensures lattice matching to the substrate without considerably changing the physical properties of the film. KT has been grown by Czochralski's method with addition of Ba in the melt to yield semi-conducting Ba:KT with electrical resistivities as low as 25 Ωm instead of about 10^{11} Ωm of pure KT. Like this it is possible to use the substrate as bottom electrode for poling procedures, electrical measurements or electro-optical applications.

The domain wall pattern in the Figure (a) shows that films which are lattice fitted in the cubic phase have a considerable amount of domains with polarization parallel to the substrate in the [100] and [010] directions. The domain walls are parallel to the [110] direction or perpendicular to it. Poling procedure with an electrical field applied perpendicular to the substrate surface (along the [001] direction) results in additional domains along [001] (Figure b), but at the same time the induced misfit is compensated by cracks in the KNTN film which occur only parallel to the crystalline [100] and [010] axes.



- (a) As-grown KNTN film showing large in-plane polarization.
- (b) Same film after poling with 10 kV/cm shows additional out-of-plane polarization at the induced cracks which only occur in [100] and [010] direction.

Reference: H. Pierhöfer, Z. Sitar, F. Gitmans, H. Wüest, and P. Günter, "New Semiconducting Substrate for Heteroepitaxial Growth of $K_{1-y}Na_yTa_{1-x}Nb_xO_3$ ", *Ferroelectrics* **201**, 269 (1997).

Difference Frequency Generation in KNbO_3

M. Mei and D. Fluck

Difference frequency generation (DFG) is known as a method to generate infrared (IR) light by mixing two incoming beams in a nonlinear material. The beam with the highest frequency is called the pump beam, the other input beam the signal and the generated beam with a frequency which is the difference of the former two the idler beam. DFG offers the possibility to design a tuneable source for the IR wavelength range by tuning one of the incoming beams and either temperature or angle tuning of the nonlinear material.

We use as nonlinear material potassium niobate (KNbO_3) which offers rather high nonlinear coefficients and at the same time a large transparency range from 390-4500 nm. Depending on the wavelength of the idler there exist different phase-matching situations. Generating idler wavelengths of either around 1146 nm or around 1481 nm can be achieved by means of noncritical phase-matching in bulk KNbO_3 where the beams propagate along one of the main dielectric axes thus avoiding walk-off between the energy flow of the three beams.

We generated up to 1.5 mW at 1146 nm by mixing the Argon laser at 488 nm with a tuneable diode laser with center wavelength at 852 nm in a 20.5 mm long crystal. The measured conversion efficiency of 0.27 %/(W cm) is smaller than the theoretical prediction of 0.52 %/(W cm) due to some optical imperfections over the long crystal length.

At 1481 nm we generated up to 0.14 mW by mixing the Argon ion laser line at 458 nm with a tuneable diode laser with center wavelength at 665 nm which is in good agreement with theoretical calculations.

At the larger idler wavelengths in the mid-infrared (2-5 μm) only critical phase-matching is possible (see Reference). The walk-off in a bulk crystal reduces the effective crystal length and thus limits the idler power. As an example we investigated the generation of an idler wave around 4.2 μm by mixing a Ti-Sapphire laser at 850 nm with a Nd:YAG-laser at 1064 nm. A theoretical conversion efficiency of $6 \cdot 10^{-5}$ %/(W cm) is calculated which leads to the conclusion that in order to generate reasonable idler power in the mid-infrared a waveguide configuration with inherently no walk-off should be used.

In conclusion, we generated light at 1146 and 1481 nm by difference frequency mixing and showed that the next step to achieve a tuneable, compact infrared-source is to use KNbO_3 waveguides.

Reference: I. Biaggio, et al., *J. Opt. Soc. Am.* **B9**, 507 (1992).

Sponsors: Swiss Priority Program Optique II

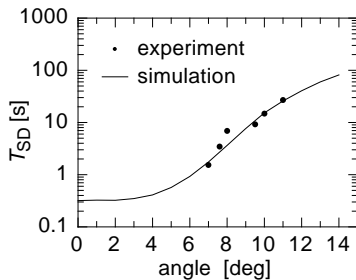
NUCLEAR MAGNETIC RESONANCE

Model Calculations for ^{87}Rb Spin-Diffusion in RDP and D-RADP-50

P. M. Cereghetti and R. Kind

The notion «spin-diffusion» is used in NMR for the heat diffusion in a spin bath. It is due to mutual spin flips of spin pairs via the spin flip-flop term of the dipole-dipole interaction. This mechanism leads to off-diagonal intensity in 2D-NMR exchange patterns which cannot be distinguished from the intensity associated with the motion. For the unambiguous analysis of a 2D-NMR exchange pattern it is therefore needed to know the contribution of spin-diffusion. To clearly explain the nature of the exchange signal observed in the glass phase of the deuterion glass $\text{Rb}_{0.5}(\text{ND}_4)_{0.5}\text{D}_2\text{PO}_4$ (D-RADP-50) the orientation dependence of spin-diffusion was measured in the FE phase of RbD_2PO_4 (RDP) where all motions are frozen-in. Using model calculations the spin diffusion time of D-RADP-50 could be determined with good accuracy. This allows to distinguish unambiguously spin-diffusion from chemical exchange.

The probability for a mutual spin flip is proportional to the secular dipole-dipole interaction as well as to the overlap integral between the corresponding resonance lines. The spin diffusion time T_{SD} depends thus on the density of the ^{87}Rb spins and on the crystal orientation in the external field B_0 . Due to the random distribution of ^{87}Rb and ^{85}Rb atoms in the RDP crystal, T_{SD} has to be calculated with a Monte Carlo simulation. For rotations of the crystal c axis about the a axis



Spin diffusion in RDP.

perpendicular to the external magnetic field B_0 spin diffusion was considered between the two single resonance lines of the ferroelectric RDP 1D spectrum, as well as within the single inhomogeneously broadened lines. 2D-NMR measurements on ^{87}Rb at a temperature of 85 K and different orientations (see Fig.) confirmed the model calculations for the first case. Very precise 2D-NMR measurements were also performed for the second case.

Experimental result and model results for the spin diffusion time T_{SD} were in good agreement, so that the model could safely be applied for the case of D-RADP-50. The observed exchange can now be unambiguously assigned to slow bias fluctuations of the hydrogen-bonds.

Reference: Th. König “Cluster Dynamics in the Solid solution D-RADP-x Investigated by ^{87}Rb NMR”, Diss. ETH No. 12027 (1997)

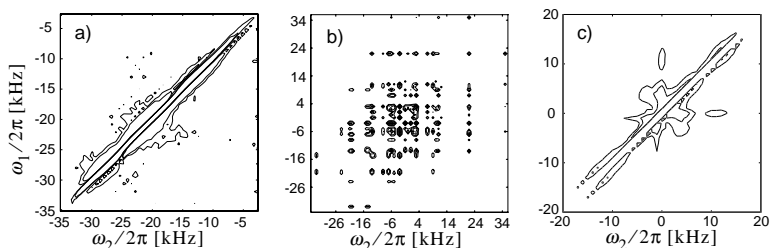
Sponsor: Swiss National Science Foundation

Point-Charge Model Calculations of the Proton Glass Dynamics

Ch. Jeitziner, Th. König and R. Kind

We report on two models simulating the measured ^{87}Rb 2D-NMR exchange difference spectra of the structural glass D-RADP-50 (Fig. a). At low temperatures the deuteron jump rate becomes slower than $1/T_2$ so that the NMR-lines reflect the instantaneous positions and not the time averaged positions of the deuterons. Thus any jump of a deuteron results in a change of the local resonance frequency observable in a 2D-NMR exchange spectrum as off-diagonal intensity. These frequency shifts can be calculated within a point charge model.

The first calculations were done with the eight closest hydrogen bridges to a given Rb. If the deuterons can move independently, all possible deuteron configurations ($2^8 = 256$) can serve with equal probability as starting and ending configuration of a set of motions. However, the frequency jumps in the corresponding calculated 2D-NMR exchange pattern are much larger than the measured ones (Fig. b).



Comparison of measured spectrum (a) with first model (b) and second model (c)

This shows unambiguously that the deuterons do not move independently of each other. The second model is based on the ice rules, i.e. only the energetically favorable Slater configurations are allowed for the PO_4 groups (two deuterons close, two far: D_2PO_4). For this we had to extend the model to 16 nearest deuterons which belong to six neighboring PO_4 -groups. To describe the motions of the deuterons we insert a Takagi group into this lattice (DPO_4 or D_3PO_4) which can move rather freely like a quasi-particle by shifting deuterons from one side of the bond to the other. Accordingly our model was changed to a random walk model for a Takagi group in a Slater lattice. The corresponding ^{87}Rb NMR-frequency changes were calculated, as well as the 2D-NMR exchange pattern (Fig. c). It fits so much better to our measurements that with great probability this random walk is responsible for the low temperature dynamics in D-RADP-50.

Reference: Th. König "Cluster Dynamics in the Solid solution D-RADP-x Investigated by ^{87}Rb NMR", Diss. ETH No. 12027 (1997)

Sponsor: Swiss National Science Foundation

Investigation of Hydrogen Bridge Bias Fluctuations with ^2H -2D-NMR

Ch. Jeitziner and R. Kind

At low temperatures $\text{Rb}_{0.5}(\text{NH}_4)_{0.5}\text{D}_2\text{PO}_4$ (D-RADP-50) forms a structural glass which is characterized by a pseudo-static short range order of the acid deuterons. On each O-D...O bridge the deuteron performs a stochastic motion between the two potential wells. These two positions are characterized by different ^2H -NMR-NQR frequencies ω_1, ω_2 which is essential for 2D-exchange NMR measurements. The deuteron motion is thermally activated and individually biased, i.e., the mean dwell times on either side of the bond generally differ from each other. This leads to a probability distribution $W(p)$ of local polarizations $\bar{p}_j = \tanh(\varepsilon_j/k_B T)$, where ε_j is the bias energy of the j -th bond. In the fast motion regime ($\tau_c^{-1} \gg |\omega_1 - \omega_2|$) the time averaged NMR-frequency $\bar{\omega}_j = (1/2)(\omega_1 + \omega_2) + (p_j/2)(\omega_1 - \omega_2)$ is observed. The inhomogeneous NMR line shape is thus an image of $W(p)$. Slow fluctuations ($\tau_{bias} \gg T_2$) of the local bias ε_j or what is equivalent of the local polarization \bar{p}_j lead to similar fluctuations of $\bar{\omega}_j$ which should be observable in a 2D-exchange NMR experiment.

To check whether such slow fluctuations are present we have performed 2D-exchange difference measurements on a single crystal of D-RADP-50 in a fixed orientation with respect to the magnetic field of 7 T. Several experiments with mixing times between 20 ms and 3.2 s were carried out for various temperatures between 70 K and 130 K where the system is in the fast motion regime.

The 2D-exchange NMR spectra reveal off diagonal intensity bands close to the diagonal (≤ 2 kHz). This means that possible polarizations fluctuations are smaller than 0.4. Only the intensity but not the shape of the cross peaks showed a dependence on the mixing time t_{mix} .

At present we are not sure whether our results can be really assigned to fluctuations of the bias energy during the mixing time or whether we are just dealing with spin diffusion which is the exchange of magnetization by mutual spin flip-flop processes between spin pairs. Because the latter process depends strongly on the frequency difference between the spin pairs, we plan to perform measurements for different orientations with respect to the external magnetic field to change the relative frequency separation of the spins which would lead to a large change in the spin diffusion time constant. In contrast to that the bias fluctuations are only temperature but not orientation dependent so that is should be possible to identify the process responsible for our observations.

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"Photorefractive Effect in LiNbO₃ Crystals Enhanced by Stoichiometry Control"
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- R. Kind, N. Korner, Th. König und Ch. Jeitziner
"Finite Size Effects in Proton Glass"
J. Korean Phys. Soc. **32** (supplement), January 1998
- I. Liakatas, M.S. Wong, Ch. Bosshard, M. Ehrensperger and P. Günter
"Stilbazolium Based Zwitterionic Chromophores for Electro-Optic Polymers"
Ferroelectrics **202** (1-4), 299-306 (1997)
- W. Lukosz, Ch. Stamm, H.R. Moser, R. Ryf and J. Dübendorfer
"Difference Interferometer with New Phase-Measurement Method as Integrated-Optical Refractor, Humidity Sensor and Biosensor"
Sensors and Actuators B **38-39**, 316-323 (1997)
- A. Marini, C. Medrano, I. Poberaj and P. Günter
"Non-Linear Optical Devices for Spaceborn Laser Applications"
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- R.E. Martin, U. Gubler, C. Boudon, V. Gramlich, Ch. Bosshard, J-P. Gisselbrecht, P. Günter, M. Gross and F. Diederich
"Poly(triacetylene) Oligomers: Synthesis, Characterization, and Estimation of the Effective Conjugation Length by Electrochemical UV/Vis, and Nonlinear Optical Methods"
Chem. Eur. J. **3** (9), 1505-1512 (1997)
- G. Montemezzani and M. Zgonik
"Light Diffraction at Mixed Phase and Absorption Gratings in Anisotropic Media for Arbitrary Geometries"
Physical Review E **55** (1), 1035-1047 (1997)
- G. Montemezzani and P. Günter
"Profile of Photorefractive One-Dimensional Bright Spatial Solitons"
Optics Letters **22** (7), 451-453 (1997)
- G. Montemezzani, C. Medrano, P. Günter and M. Zgonik
"Charge Carrier Photoexcitation and Two-Wave Mixing in Dichroic Materials"
Physical Review Letters **79** (18), 3403-3406 (1997)
- B. Müller and M. Henzler
"Comparison of Reflection High-Energy Electron Diffraction and Low-Energy Electron Diffraction Using High-Resolution Instrumentation"
Surface Science **389**, 338-348 (1997)
- B. Müller and V. Zielasek
"Inelastic Scattering in Reflection High-Energy Electron Diffraction from Si(111)"
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- L. Nedelmann, B. Müller, B. Fischer, K. Kern, D. Erdös, J. Wollschläger and M. Henzler
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Chemistry of Materials **9** (6), 1328-1334 (1997)
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"A Highly Efficient Organic Second-Order Nonlinear Optical Crystal Based on a Donor-Acceptor Substituted 4-Nitrophenylhydrazone"
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- H. Pierhöfer, Z. Sitar, F. Gitmans, H. Wüest and P. Günter
"New Semiconducting Substrate for Heteroepitaxial Growth of $K_{1-y}Na_yTa_{1-x}Nb_xO_3$ "
Ferroelectrics **201** (1-4), 269-275 (1997)
- T. Pliska, C. Solcia, D. Fluck, P. Günter, L. Beckers and Ch. Buchal
"Radiation Damage Profiles of the Refractive Indices of He⁺ Ion-Implanted KNbO₃ Waveguides"
J. Appl. Phys. **81** (3), 1099-1102 (1997)
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"Tetraethynylethene Molecular Scaffolding: Nonlinear Optical, Redox, and Amphiphilic Properties of Donor Functionalized Polytriacetylene and Expanded Radialenes"
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"High-Frequency Response and Acoustic Phonon Contribution of the Linear Electro-Optic Effect in DAST"
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- Y. Tao, F. Gitmans, Z. Sitar, H. Pierhöfer, A. Kündig, I. Gamboni and P. Günter
"Dielectric, Pyroelectric and Structural Properties of LiTaO₃ Thin Films Grown on Silicon by a Modified Molecular Beam Epitaxy"
Ferroelectrics **201** (1-4), 245-253 (1997)
- M.S. Wong, F. Pan, V. Gramlich, Ch. Bosshard and P. Günter
"Self-Assembly of an Acentric Co-Crystal of a Highly Hyperpolarizable Merocyanine Dye with Optimized Alignment for Nonlinear Optics"
Advanced Materials **9** (7), 554-557 (1997)
- M.S. Wong, Ch. Bosshard and P. Günter
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- M.S. Wong, V. Gramlich, Ch. Bosshard and P. Günter
"Hydrogen Bonded Lambda-Shaped Packing Motif Based on 4-Nitrophenylhydrazones: A Promising Design Tool for Engineering Acentric Crystals"
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- M.S. Wong, Ch. Bosshard and P. Günter
"Engineering of Polar Molecular Crystals with Optimized Chromophoric Orientation for Nonlinear Optics"
Ferroelectrics **202** (1-4), 51-64 (1997)

Conference Publications

- P. Bernasconi, I. Biaggio, G. Montemezzani and P. Günter
"Anisotropic Charge Mobility in KNbO_3 and BaTiO_3 "
Conference on Lasers and Electro-Optics (CLEO '97), OSA Technical Digest Series (Optical Society of America, Washington DC) **11**, 122 (1997)
- P. Bernasconi, G. Montemezzani, I. Biaggio and P. Günter
"Multiple Photorefractive Gratings Induced by Interband Photoexcitation in KNbO_3 Crystals"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 399-402 (1997)
- I. Biaggio, P. Bernasconi, M. Ewart and P. Günter
"Characterization of Charge Transport in BaTiO_3 and KNbO_3 by Pulsed and Continuous Wave Photorefractive Techniques"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 71-74 (1997)
- Ch. Bosshard, F. Pan, M.S. Wong, M. Bösch, U. Meier and P. Günter
"Thiophene Based Hydrazones: a New Class of Nonlinear Optical Molecular Crystals"
Proceedings of Organic Thin Films for Photonic Applications, Long Beach, CA, USA, Optical Society of America, Technical Digest Series **14**, 101-103 (1997)
- M. Ewart, R. Ryf, C. Medrano, H. Wüest, M. Zgonik, I. Biaggio and P. Günter
"High Photorefractive Sensitivity in the Visible and at 860 nm in Reduced Iron and Rhodium-Doped KNbO_3 "
Conference on Lasers and Electro-Optics (CLEO '97), OSA Technical Digest Series (Optical Society of America, Washington DC) **11**, 333 (1997)
- M. Ewart, I. Biaggio, M. Zgonik and P. Günter
"Third-Order and Cascaded Second-Order Contributions to Pulsed Degenerate Four-Wave Mixing in BaTiO_3 "
Quantum Electronics and Laser Science Conference (QELS '97), OSA Technical Digest Series (Optical Society of America, Washington DC) **11**, 123 (1997)
- M. Ewart, R. Ryf, C. Medrano, H. Wüest, M. Zgonik and P. Günter
"Reduced KNbO_3 for Photorefractive Applications at Visible and Near-Infrared Wavelengths"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 531-534 (1997)
- D. Fluck and P. Günter
"Compact Blue Laser Based on Sum-Frequency Mixing of Laser Diodes in KNbO_3 "
Helv. Phys. Acta **70**, Separanda 1, 11-12 (1997)
- Y. Furukawa, K. Kitamura, Y. Ji, P. Bernasconi, G. Montemezzani and P. Günter
"Photorefractive Properties in Rh:LiTaO_3 in the Ultraviolet and Visible Wavelength Region"
Conference on Lasers and Electro-Optics (CLEO '97), OSA Technical Digest Series (Optical Society of America, Washington DC) **11**, 120-121 (1997)
- Y. Furukawa, K. Kitamura, S. Matsumara, P. Bernasconi, G. Montemezzani and P. Günter
"Photorefractive Effects in Rh:LiTaO_3 at Ultraviolet and Visible Wavelengths"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 153-156 (1997)

- K. Kitamura, Y. Furukawa, Y. Li, M. Zgonik, C. Medrano, G. Montemezzani and P. Günter
"Amplification in Two-Wave Mixing Enhanced by LiNbO₃ Stoichiometry Control"
Conference on Lasers and Electro-Optics (CLEO '97), OSA Technical Digest Series (Optical Society of America, Washington DC) **11**, 121-122 (1997)
- G. Montemezzani, M. Zgonik and P. Günter
"Light Diffraction at Thick Gratings in Anisotropic Media"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 391-394 (1997)
- G. Montemezzani, M. Zgonik, C. Medrano and P. Günter
"Photorefractive Two-Beam Coupling Revisited"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 645-648 (1997)
- R. Ryf, G. Montemezzani, M. Wiki and P. Günter
"Self-Focusing and Spatial Solitons in Photorefractive KNbO₃ Crystals"
Quantum Electronics and Laser Science Conference (QELS '97), OSA Technical Digest Series (Optical Society of America, Washington DC) **11**, 169-170 (1997)
- R. Ryf, A. Lötscher, M. Wiki, G. Montemezzani and P. Günter
"Study of Photorefractive Self-Focusing and Spatial Soliton Generation in KNbO₃ Crystals"
Proceedings of Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan, 395-398 (1997)

Articles in print

- J. Dolinsek, M. Koren and R. Kind
"Kinetic Freezing of the Supercooled Liquid State in 2-Cyclooctylamino-5-Nitropyridine (COANP)"
Phys. Rev. B **57** (2), January 1998
- R. Kind, N. Korner, Th. König and Ch. Jeitziner
"Finite Size Effects in Proton Glass"
J. Korean Phys. Soc. **32** (supplement), January 1998
- U. Meier, M. Bösch, Ch. Bosshard, F. Pan and P. Günter
"Parametric Interactions in the Organic Salt 4-N,N-Dimethylamino-4'-N'-Methyl-Stilbazolium Tosylate at Telecommunication Wavelengths"
J. Appl. Phys. **83** (7), (1998)
- R. Ryf, A. Lötscher, Ch. Bosshard, M. Zgonik and P. Günter
"Z-scan Based Investigations of Photorefractive Self-Focusing in KNbO₃ Crystals"
J. Opt. Soc. Am. B **15** (3), (1998)

ORAL PRESENTATIONS 1997

* = invited talk

M. Abplanalp

"Mapping the Domain Distribution at Ferroelectric Surfaces by Scanning Force Microscopy"

STM '97, Hamburg, D

20.-25.7.97

H. Arend

"Organisch-anorganische Doppelhalogenide und Komplexverbindungen und ihre Bedeutung für die Festkörperwissenschaften"

Seminar Anorganisch-Chemisches Institut der Universität Zürich, Zürich

25.4.97

J.V. Barth, B. Müller, L. Nedelmann, B. Fischer, H. Brune and K. Kern

"Strain-induced Island Ramification in Heteroepitaxy Cu/Ni(100)"

European Conference on Surface Science, Enschede, NL

16.-19.9.97

P. Bernasconi, I Biaggio and P. Günter

"Anisotropic Drift Mobility in KNbO_3 and BaTiO_3 "

Frühjahrstagung der SPG, Neuchâtel

27.2.97

P. Bernasconi, I. Biaggio, G. Montemezzani and P. Günter

"Anisotropic Charge Mobility in KNbO_3 and BaTiO_3 "

CLEO/QELS '97, Baltimore, USA

18.-23.5.97

I. Biaggio, M. Ewart, M. Zgonik and P. Günter

"Third-Order and Cascaded Second-Order Contributions to Pulsed Degenerate Four Wave Mixing in BaTiO_3 "

CLEO/QELS '97, Baltimore, USA

18.-23.5.97

I. Biaggio, P. Bernasconi, M. Ewart and P. Günter

"Characterization of Charge Transport in BaTiO_3 and KNbO_3 by Pulsed and Continuous Wave Photorefractive Techniques"

Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97),

Chiba, Japan

11.-13.6.97

I. Biaggio, P. Bernasconi, G. Montemezzani and P. Günter

"Multiple Photorefractive Gratings Induced by Interband Photoexcitation in KNbO_3 Crystals"

Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97),

Chiba, Japan

11.-13.6.97

M. Bösch, I. Liakatas, Ch. Bosshard, M.S. Wong, M. Ehrensperger and P. Günter
"Stilbazolium Based Zwitterionic Chromophores for Electro-Optic Polymers"
Frühjahrstagung der DPG, Münster, D
17.-21.3.97

* **Ch. Bosshard**

"Noncentrosymmetry in Nonlinear Optics"
Workshop on Nonlinear Optical Properties of Polymers and Related Topics
(OPTIMAS), Bayreuth, D
30.6.-2.7.97

Ch. Bosshard, U. Gubler, P. Kaatz, W. Mazerant and P. Günter
"Optical Third-Harmonic Generation: Cascaded Second-Order Nonlinearities in KNbO₃"
"Materials for Nonlinear Optics", 11th Topical Meeting of the European Optical Society, Capri, I
8.-12.7.97

* **Ch. Bosshard**

"Molekularkristalle für die Nichtlineare Optik"
Universität Mainz, Mainz, D
14.7.97

Ch. Bosshard, F. Pan, M.S. Wong, M. Bösch, U. Meier and P. Günter
"Thiophene Based Hydrazones: a New Class of Nonlinear Optical Molecular Crystal"
Organic Thin Films for Photonic Applications, Long Beach, CA, USA
15.-17.10.97

* **Ch. Bosshard**

"Molecular Crystals for Nonlinear Optics: Recent Developments"
University at Davis, California, USA
24.10.97

* **Ch. Bosshard**

"Molekularkristalle für nichtlinear optische Effekte zweiter und dritter Ordnung: neue Entwicklungen"
Seminarvortrag Ludwig-Maximilian Universität, München, D
4.12.97

S. Brülisauer

"Infrared Photorefractive Effect up to 1550 nm in Fe-Doped KNbO₃ by Proton-Irradiation"
CLEO/QELS '97, Baltimore, USA
18.-23.5.97

P. Cereghetti, N. Korner and R. Kind

"Analysis of ⁸⁷Rb Spin Diffusion in Mixed Crystals of Rb_{1-x}(ND₄)₄D₂PO₄"
Gordon Conference on Magnetic Resonance, Henniker, NH, USA
22.-27.6.97

* **L. Eng**

"Ferroelectric Domain Switching and 3-Dimensional Electric Field Mapping at Ferroelectric Surfaces by Scanning Force Microscopy"
Seminar on Solid State Physics, ETH Zürich
22.5.97

* **L. Eng**

"Direct Writing of Ferroelectric Domains at Room Temperature by Scanning Force Microscopy"
9th Int. Conference on Scanning Tunneling Microscopy and Related Techniques (STM '97), Hamburg, D
20.-25.7.97

M. Ewart, R. Ryf, C. Medrano, M. Zgonik, H. Wüest and P. Günter
"Reduced KNbO₃ for Photorefractive Applications"
Frühjahrstagung der SPG, Neuchâtel
27.2.97

M. Ewart, C. Medrano, R. Ryf, C. Medrano, H. Wüest, M. Zgonik, I Biaggio and P. Günter
"High Photorefractive Sensitivity in the Visible and at 860 nm in Reduced Iron- and Rhodium-Doped KNbO₃"
CLEO/QELS '97, Baltimore, USA
18.-23.5.97

* **D. Fluck**

"Stable 45 mW Blue Laser by Frequency Doubling in KNbO₃"
Laser '97, München, D
16.-20.6.97

Y. Furukawa, K. Kitamura, P. Günter and M. Sato
"Development of Photorefractive LiTaO₃ Crystals for Ultraviolet and Visible Wavelength Region"
Annual Meeting of Japanese Applied Physics Society, Funabashi, Japan
31.3.97

Y. Furukawa, K. Kitamura, Y. Ji, P. Bernasconi, G. Montemezzani and P. Günter
"Photorefractive Properties in Rh:LiTaO₃ in the Ultraviolet and Visible Wavelength Region"
CLEO/QELS '97, Baltimore, USA
18.-23.5.97

Y. Furukawa, K. Kitamura, S. Matsumara, P. Bernasconi, G. Montemezzani and P. Günter
"Photorefractive Effects in Rh:LiTaO₃ at Ultraviolet and Visible Wavelength Region"
Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan
11.-13.6.97

- U. Gubler**, R.E. Martin, Ch. Bosshard, R.R. Tykwinski, P. Günter and F. Diederich
"Influence of Conjugation Length and Substitution on the Second-Order Hyperpolarizability in Polytriacetylene"
KOPO '97 Workshop - Konjugierte Polymere und Oligomere, Blaubeuren bei Ulm, D
2.-6.7.97
- U. Gubler**, R.E. Martin, Ch. Bosshard, R.R. Tykwinski, P. Günter and F. Diederich
"Influence of Conjugation Length Expansion and Substitution on the Second-Order Hyperpolarizability of γ of Oligotriacetylenes"
"Materials for Nonlinear Optics", 11th Topical Meeting of the European Optical Society, Capri, I
8.-12.7.97
- * **P. Günter**
"Organic and Inorganic Photorefractive Materials"
4th International Conference on Frontiers of Polymers and Advanced Materials, Cairo, Egypt
4.-9.1.97
- * **P. Günter**
"Organic and Inorganic Photorefractive Materials"
International Workshop on Nonlinear Optics and Laser Materials, Bilbao, Spain
28.5.97
- P. Günter**, M. Ewart, R. Ryf, C. Medrano, H. Wüest and M. Zgonik
"Reduced KNbO₃ for Photorefractive Applications at Visible and Near-Infrared Wavelengths"
Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97), Chiba, Japan
11.-13.6.97
- * **P. Günter**
"Novel Organic Nonlinear Optical Materials"
CLEO/Pacific Rim '97, Chiba, Japan
14.-18.7.97
- * **P. Günter**
"KNbO₃ and Blue Lasers"
Int. Symposium on Lasers and Nonlinear Optical Materials (IS-LNOM '97), Singapore
3.-6.11.97
- * **R. Kind**
"Break-Through in the Interpretation of the Low Temperature Dynamics in the Deuteron Glass D-RADP-50"
Institute Jozef Stefan, Ljubljana, SLO
25.3.97

*** R. Kind**

"Finite Size Effects in Proton Glass"

9th International Meeting on Ferroelectricity, Seoul, South Korea
24.-29.8.97

K. Kitamura, Y. Furukawa, Y. Ji, M. Zgonik, C. Medrano, G. Montemezzani and P. Günter

"Amplification in Two Wave Mixing Enhanced by LiNbO₃ Stoichiometry Control"
CLEO/QELS '97, Baltimore, USA
18.-23.5.97

K. Kitamura, Y. Furukawa, G. Montemezzani and P. Günter

"Stoichiometric Lithium Niobate Single Crystals for Holographic Storage Devices"
The 28th National Conference on Crystal Growth 1997, Sapporo, Japan
24.7.97

O. Leifeld, B. Müller, D.A. Grützmacher and K. Kern

"A UHV-STM for in situ Characterization of MBE/CVD Growth on 4-inch Si-Wafers"
9th Int. Conference on Scanning Tunneling Microscopy/Spectroscopy and Related Techniques, Hamburg, D
25.-25.7.97

O. Leifeld, B. Müller, D.A. Grützmacher, R. Hartmann, T. Jung and K. Kern

"Morphologische in situ Charakterisierung von MBE-Schichten auf 4"-Si-Substraten mittels UHV-STM"
MBE-Workshop '97, Bremen, D
22./23.9.97

I. Liakatas, M.S. Wong, Ch. Bosshard and P. Günter

"Nonlinear Optical Chromophores Based on Multi-Donor Substituted 4-Nitrophenyl-hydrazones"
"Materials for Nonlinear Optics", 11th Topical Meeting of the European Optical Society, Capri, I
8.-12.7.97

I. Liakatas, M.S. Wong, Ch. Bosshard, M. Bösch and P. Günter

"Novel Zwitteronic Chromophores for Electro-Optic Polymers"
"Materials for Nonlinear Optics", 11th Topical Meeting of the European Optical Society, Capri, I
8.-12.7.97

R.E. Martin, U. Gubler, R.R. Tykwinski, C. Boudon, Ch. Bosshard,

J.-P. Gisselbrecht, P. Günter, M. Gross and F. Diederich

"Poly(triacetylene) Oligomers: Synthesis and Characterization"

KOPO '97 Workshop - Konjugierte Polymere und Oligomere, Blaubeuren bei Ulm, D
2.-6.7.97

R.E. Martin, U. Gubler, R.R. Tykwinski, C. Boudon, Ch. Bosshard,
J.-P. Gisselbrecht, P. Günter, M. Gross and F. Diederich
"Structure-Property Relationships in Poly-triacetylenes: Effective Conjugation
Length, Influence of Lateral Conjugation, and Donor-Acceptor Substitution"
"Materials for Nonlinear Optics", 11th Topical Meeting of the European Optical
Society, Capri, I
8.-12.7.97

M. Mei, D. Fluck and P. Günter
"Optical Difference Frequency Generation in KNbO_3 "
Herbsttagung der SPG, La Chaux-de-Fonds
10.10.97

U. Meier, Ch. Bosshard and P. Günter
"Two Photon Induced Fluorescence in Nonlinear Optical Organic DAST Crystals"
Herbsttagung der SPG, La Chaux-de-Fonds
10.10.97

- * **G. Montemezzani**
"Advances in Photorefractive Materials and Effects at ETH Zürich"
NIRIM Seminar on Crystal Growth, National Institute for Research in Inorganic
Materials, Tsukuba, Japan
3.3.97
- * **G. Montemezzani**
"Introduction to the Photorefractive Effect"
PIONEER Corporate Research and Development Laboratory,
Tsurugashima, Japan
5.3.97
- * **G. Montemezzani**
"New Developments in Photorefractive Materials"
Frontier Research Program Forum, RIKEN Institute, Wako, Japan
5.3.97
- * **G. Montemezzani**
"L'effetto fotorifrattivo: un'introduzione alle nonlinearità ottiche a basse intensità"
Colloquio del Centro di ricerca in fisica matematica (Cerfim), Locarno
25.4.97
- G. Montemezzani**, R. Ryf, M. Wiki and P. Günter
"Self-Focusing and Spatial Solitons in Photorefractive KNbO_3 Crystals"
CLEO/QELS '97, Baltimore, USA
18.-23.5.97
- G. Montemezzani**, M. Zgonik, C. Medrano and P. Günter
"Photorefractive Two-Wave Mixing Revisited"
Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97),
Chiba, Japan
11.-13.6.97

G. Montemezzani, M. Zgonik and P. Günter

"Light Diffraction at Thick Gratings in Anisotropic Media"

Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97),
Chiba, Japan

11.-13.6.97

G. Montemezzani, R. Ryf, A. Lötscher, M. Wiki and P. Günter

"Study of Photorefractive Self-Focusing and Spatial Soliton Generation in KNbO_3
Crystals"

Topical Meeting on Photorefractive Materials, Effects and Devices (PR '97),
Chiba, Japan

11.-13.6.97

* **B. Müller**

"A Microscopic Insight into Heteroepitaxial Growth of Metals"

Colloquium at Condensed Matter Physics and Chemistry Department,
Risø National Laboratory, Risø, DK

22.1.97

* **B. Müller**

"First Stages of Heteroepitaxy on Metals: Cu/Ni (100)"

Colloquium at Institute of Physics and Astronomy, Center for Atomic-Scale Materials
Physics, University of Aarhus, DK

23.1.97

* **B. Müller** and M. Henzler

"Comparison of Reflection High-Energy Electron Diffraction and Low-Energy
Electron Diffraction Using High-Resolution Instrumentation"

US Japan Seminar on Surface Dynamics and Structures in Epitaxial Growth,
Nagoya, Japan

24.-28.3.97

* **B. Müller**

"Island Shape Transition in Heteroepitaxial Growth on Square Lattices"

US Japan Seminar on Surface Dynamics and Structures in Epitaxial Growth,
Nagoya, Japan

24.-28.3.97

B. Müller

"Growth Kinetics Versus Thermodynamics in Heteroepitaxy"

Science and Technology Colloquium, IBM Research Division, Research Laboratory,
Zürich

10.9.97

B. Müller

"Molekularstrahlepitaxie organischer Materialien für Nichtlineare Optik"

MBE-Workshop '97, Bremen, D

22./23.9.97

B. Müller

"Die Wirkung des Strain bei der Heteroepitaxie - Untersuchungen zu Inselformen im Nano- und Mikrometerbereich"

Institut für Festkörperphysik, Universität Hannover, D

25.9.97

T. Pliska

"Efficient Blue Light Second-Harmonic Generation in Ion-Implanted KNbO₃ Channel Waveguides"

8th European Conference on Integrated Optics (ECIO '97), Stockholm, S

2.4.4.97

T. Pliska, D. Fluck and P. Günter

"Efficient Blue-Light Second-Harmonic and Sum-Frequency Generation in Ion-Implanted KNbO₃ Channel Waveguides"

CLEO/QELS '97, Baltimore, USA

18.-23.5.97

T. Pliska, D. Fluck, P. Günter, L. Beckers and Ch. Buchal

"Efficient Birefringence Phase-Matched Second-Harmonic in KNbO₃ Ridge Waveguides"

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10.10.97

R. Ryf, M. Wiki, G. Montemezzani and P. Günter

"Photorefractive Spatial Solitons in KNbO₃ Crystals"

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27.2.97

Ch. Seuret

"Atomic Force Measurements and Computer Simulations of Langmuir Monolayers"

STM '97, Hamburg, D

20.-25.7.97

U. Suter, M. Döbler, Ch. Weder, P. Neuenschwander, S. Follonier, Ch. Bosshard and P. Günter

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4.-9.1.97

J. Wollschläger, D. Erdös, B. Müller, B. Fischer, L. Nedelmann, J. Barth and K. Kern

"Epitaktisches Wachstum von Cu/Ni(100): vergleichende SPA-LEED und STM Untersuchungen"

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17.-21.3.97

PHD THESES IN PHYSICS 1997

Ewart, Michael

"Reduced KNbO_3 for Photorefractive Applications"

ETH Zürich, Nr. 12484 (Prof. Dr. P. Günter & Prof. Dr. H. Melchior)

König, Thomas

"Cluster Dynamics Processes in the Solid Solution $\text{Rb}_{1-x}(\text{ND}_4)_x\text{D}_2\text{PO}_4$ Investigated by Means of ^{87}Rb NMR Techniques"

ETH Zürich, Nr. 12027 (Prof. Dr. R. Kind, Prof. Dr. P. Günter & Dr. J. Roos)

Pliska, Tomas

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Korreferat:

Gini, Emilio

"Plasma Etched Optical Corner Mirrors and Grating Demultiplexers in GaAsP/ InP"

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Fischer, Stefan

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Messmer, Peter

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