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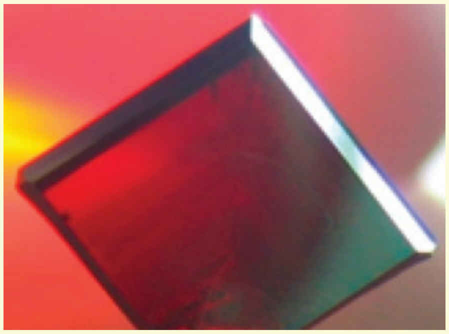
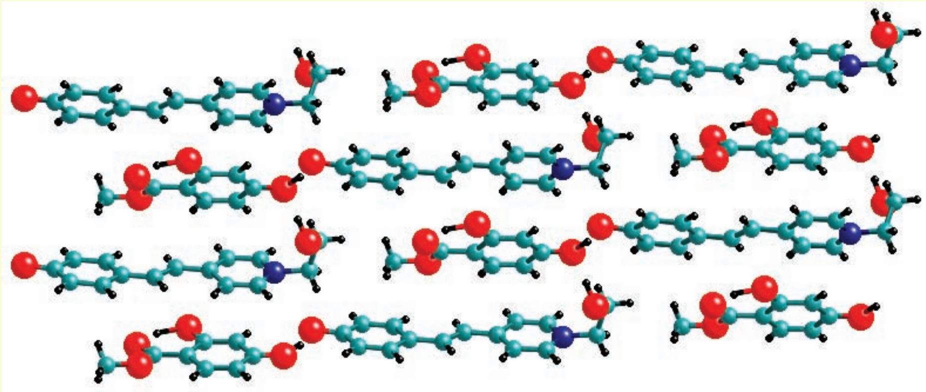
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Institute of Quantum Electronics

Nonlinear Optics Laboratory

NLO Report 1987 - 2009





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Nonlinear Optics Laboratory

1987 - 2009

Prof. Dr. P. Günter
Nonlinear Optics Laboratory
Institute of Quantum Electronics
ETH - Hönggerberg
CH - 8093 Zürich

Phone:	+41 44 633 2295
Fax:	+41 44 633 1056
e-mail:	nlo@phys.ethz.ch
internet:	www.nlo.ethz.ch/

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Chapter 1

Overview of Activities

1.1 Introduction

This report gives an overview of the main research and teaching activities of the Nonlinear Optics Laboratory. This laboratory was founded in 1987 for reinforcing activities in Photonics, Nonlinear Optics and Quantum Electronics. The basis for the decision to form a new center for Quantum Electronics and Photonics at ETH was a report by an international scientific advisory board strongly recommending the ETH president to reinforce activities in quantum electronics, nonlinear optics and lasers. As a first consequence, the Institute of Quantum Electronics was founded in 1980 with the first three laboratories for: Infrared Physics (Prof. F. Kneubühl), Microelectronics and Optoelectronics (Prof. H. Melchior) and Optics (Prof. W. Lukosz). The main mission of the Institute has been defined as: "Research and Teaching of the Fundamentals of (Optical) Information Technology". Our Nonlinear Optics Laboratory joined the Institute in 1987 and the Physical Electronics Laboratory (Prof. H. Baltes) in 1988.
Before giving an overview of the main activities of our laboratory, the most important earlier activities within the Physics Department which were of fundamental importance for nonlinear optics and materials research in our laboratory should be mentioned.

1.1.1 Historical Background

The main mission of the Nonlinear Optics Laboratory was the investigation of the basic physics of novel photonics applications and the research and development of novel materials for these applications. These new materials should have large nonlinear optical and electro-optical effects so that a sufficient response can be obtained at low laser power obtainable with compact, maintenance-free laser sources like laser diodes or at low electric fields respectively. Polar dielectric materials (crystals and polymers) show the highest first order nonlinear susceptibilities and are therefore the prime candidate materials for photonic materials.

"The dwarf sees further than the giant when he has the giants shoulder to mount on" (S.T. Coleridge)

The investigation of polar materials at ETH has a long tradition with a series of "giants" from which all members of the Nonlinear Optics Laboratory could profit. It started with the investigation of polar molecules and their dipole moments" by Peter Debye who was a Professor and Head of the Physics Institute between 1920 and 1927. Even before, in 1916, at the University of Göttingen, he developed together with Paul Scherrer the "Debye-Scherrer" x-ray diffraction method for the investigation of crystal structures. Scherrer, who followed Debye as the head of the Physics Institute, continued research on polar crystals. His PhD student George Busch later developed potassium dihydrogene phosphate (KH_2PO_3) and its derivates and discovered ferroelectricity in this crystal (second known ferroelectric crystal in 1935). After this discovery, the favorable electro-optical properties of KH_2PO_4 have been investigated in 1944 at ETH in the PhD thesis of B. Zwicker "Elastische Untersuchungen an $NH_4H_2PO_4$ und KH_2PO_4 (1946)". The growth of this crystal has now been improved so dramatically that crystals with excellent optical quality and dimensions of more than 1m can be produced. This material is therefore now widely used in high power nonlinear optics e.g. for laser fusion. A first model describing the relation of the electro-optical activity with the dielectric constant and spontaneous polarization was developed by Zwicker and Scherrer. Shortly before 1950, ETH Zurich became one of the leading centers for ferroelectricity research with a series of materials developed and investigated. Some of the "giants" active at that time include B. Mathias ($LiNbO_3$, $KNbO_3$, ...), W.J. Merz ($BaTiO_3$, ...), W. Känzig ($BaTiO_3$, ...) and H. Gränicher ($SrTiO_3$, $LiNbO_3$, $KNbO_3$, ...), Nobel laureates K.A. Müller and J.B. Bednorz ($SrTiO_3$) and others.

Peter Günter started his PhD thesis on the "Electro-optical and photorefractive properties of $KNbO_3$" in 1971 in the so called "crystal group"of Prof. H.W. Gränicher. Most relevant for the research on this topic was the fundamental work of E. Wiesendanger on "Dielectric, mechanical and optical properties of

orthorhombic $KNbO_3$", the fundamental work of R. Hofmann, and the extremely successful growth of pure and doped $KNbO_3$ single crystals by Wilf Huber, Ulrich Flückiger, Hanns Arend and Hermann Wüest.
$KNbO_3$, as one of the most active nonlinear optical, electro-optical and photorefractive material, was the main work horse for many years. Many new effects, e.g. the ultrafast photorefractive effect, photorefractive anisotropic Bragg diffraction could be discovered and new applications, e.g. first all-solid-state blue lasers by direct doubling of laser diodes, optical incoherent to coherent converter, optical image amplification, nonlinear optical one way viewing, self-pumped optical phase conjugation, etc., could be demonstrated.
In 1982, a new activity in the field of organic photonics began. These highly polarizable new organic materials based on polar molecules with extended π-conjugation show even larger and faster optical nonlinearities than the ferroelectric crystals. Until the founding of the Nonlinear Optics Laboratory in 1987, half a dozen new crystals could be grown and very exciting physical properties of the molecules and the crystals could be reported. Since 1987, organic photonics is one of the most important activity in the Nonlinear Optics Laboratory with a wide variety of experimental methods, molecule synthesis and materials preparation techniques having been developed. PD Dr. Christian Bosshard played a very important role in this area as the project leader until 2001. In the following, these and some of the activities in the area are briefly described.

1.1.2 Research Overview

The Nonlinear Optics Laboratory at ETH Zurich has been active in the preparation and application of nonlinear optic materials since the beginning of their activities in 1971 (then at the Solid State Physics Laboratory). The laboratory was active in molecule synthesis, crystal growth and thin film deposition as well as in materials characterization with respect to their linear-, nonlinear-, electro-optical, as well as other properties and their applications in photonics. The laboratory combined in an unique way all steps of modern materials and physics research from the preparation and structuring of new materials (crystals and polymers, nano-structured thin films) to the investigation of their physical properties and its applications. The Nonlinear Optics Group had a series of collaborations with international top laboratories and attracted a high above average number of ETH students to perform diploma (147) and PhD theses (65). This also made it easy to select excellent PhD students and postdocs. This also demonstrates the attractiveness of nonlinear optics research performed also from the students' point of view.

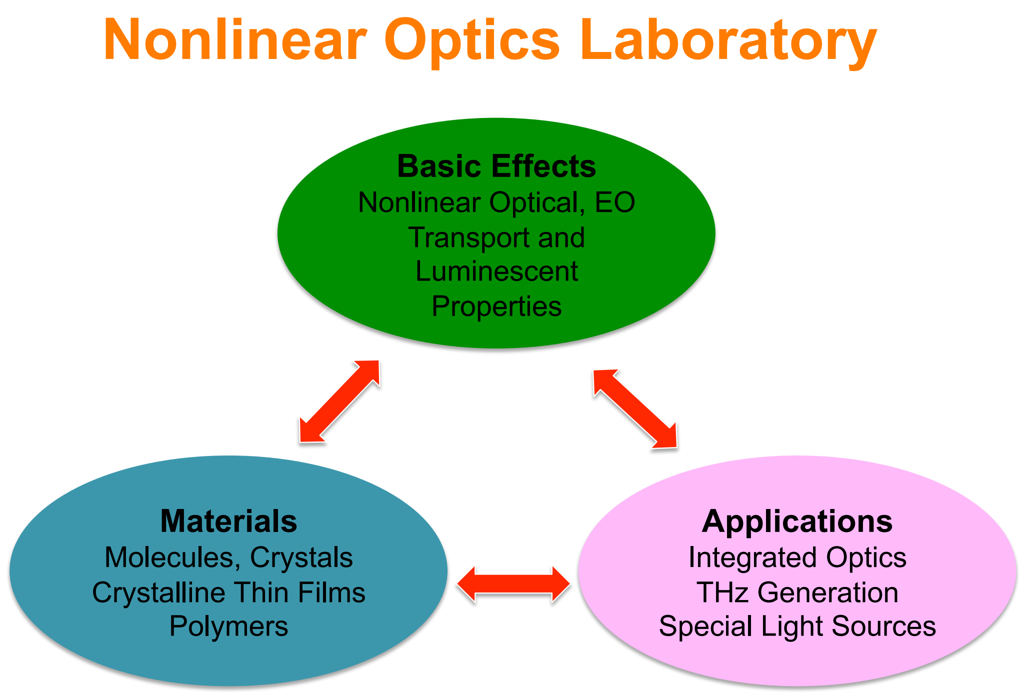


Figure 1.1: Overview of Activities

Some of the main research activities include:

- Nonlinear optics of molecular crystals, molecules and polymers
- Photorefractive effects and applications in optical parallel processing
- Integrated optics and electro-optically active microresonators
- Blue and UV solid state lasers based on optical frequency conversion in waveguides
- Molecular beam epitaxy of organic thin films for organic photonics and electronics

- Photonic material technologies
- Investigation of organic molecules at interfaces using nonlinear optics and atomic force microscopy
- Atomic force microscopy of ferroelectric domains
- Physics and applications of THz waves

In all these activities the interplay of materials science, basic physics and applications as illustrated in Figure 1.1 was most important.

1.1.3 Organic Nonlinear Optics

Various activities at the Nonlinear Optics Laboratory dealt with organic materials, which are of great interest for second- and third-order nonlinear optical applications due to their large optical nonlinearities and their ultrafast, almost purely electronic response. These materials offer many possibilities to tailor-make substances with the desired properties through optimization of the microscopic hyperpolarizabilities (molecular engineering) and the incorporation of the molecules in a crystalline lattice (crystal engineering) and polymers. Some of the highlights include the following topics:

- Several second-order organic nonlinear optical crystals have been developed and/or investigated in our laboratory. Many of these materials present today's best organic nonlinear optical crystals, e.g. DAST, DSTMS, DAPSH, Mero-2-MDB, and OH1*. We optimized the solution growth technique for these crystals and fabricated high optical quality organic crystals with side lengths exceeding 1 cm.
- We have investigated thermal stability of poled electro-optic polymers and have determined fundamental relations between the chromophore relaxation time and the polymer glass transition temperature.
- New groups of organic nonlinear optical polymers had been developed within the group or in collaboration with several external groups, including e.g. highly photostable bithiophenes for electro-optics, photoswitchable photochromic materials for recording reconfigurable optical waveguides, side-chain polyimides based on disperse red, highly nonlinear phenylthiophene molecules.

- We have studied the photochemical stability of organic materials, developed a theoretical background for the photostability measurement and have shown that oxygen is most often the main cause of photodegradation. We have shown that nonlinear optical chromophores may exhibit several orders better photostability in crystalline compared to polymeric environment.

- By Langmuir-Blodgett technique we have produced high-quality ordered thin films and waveguides for a variety of organic molecules. Based of this, we have demonstrated first guided second-harmonic generation in organic nonlinear optical films.

- We have investigated several third-order nonlinear optical organic materials for all-optical applications and have found particularly large nonlinearities in polyenes. We have also found important relations between the molecular structure and resulting third-order nonlinearities.

- Fundamental investigations on the cascaded contributions of the second-order nonlinearities to higher-order nonlinear optical processes have led to a full understanding of all the experiments in organic as well as inorganic crystals. These have allowed developing the first complete theory of degenerate four-wave mixing in noncentrosymmetric materials that includes all second-order contributions and their peculiar dependencies on the geometry of the experimental set-up.

- Several new methods to grow highly nonlinear optical organic single crystalline thin films of organic crystals for integrated optics have been developed. Thin optical quality crystals essential for integrated optics, with a thickness 0.2–5μm have been obtained by solution, melt and vapor growth of different promising compounds.

- High-finesse microring resonators have been fabricated from the inorganic-organic hybrid materials with thermo-optic tuning of the resonance output. Using micro-molding technique, we have also produced polymeric electro-optic microresonators based on a novel coupling approach that additionally allows shaping the output resonance curve, which additionally enhances the electro-optic modulation efficiency.

- We have developed several new methods to microstructure organic crystal for integrated optics, including femtosecond laser ablation, proton ion implantation and electron beam irradiation. This has allowed demonstrating efficient integrated electro-optic modulators in DAST* crystals. Based on a new technique allowing for large-area single crystalline thin films on various substrates, we have also demonstrated guided-wave modulators in OH1* single crystalline wires.

- With a new melt-based growth technique in grooves we have produced single crystalline electro-optic thin films, waveguides and nanowires, and have demonstrated both phase and amplitude integrated optical modulators therein. This technique is particularly attractive since it allows for very high optical quality crystalline organic waveguides with all the structuring done on standard inorganic substrates. Based on this technique, first organic crystalline electro-optic filters and modulators base on the organic crystal COANP* have been demonstrated.

*See Table 3.1 for names and structures of the organic compounds.

Photorefractive Nonlinear Optics

The photorefractive team has been active in the development, characterization and applications of novel infrared sensitive materials, and in the investigation of the photorefractive effect at UV and deep UV wavelengths. We have also developed photorefractive beam combiners and phase conjugators for semiconductor lasers, light induced waveguides and ultrafast optical correlators. Some of the main achievements include:

- We have optimized the growth of $KNbO_3$ and achieved state-of-the-art optical quality of the crystals with various dopants and various reduction levels, which were used to optimize the photorefractive response of these materials at different light wavelengths, as well as for other nonlinear optical applications such as frequency doubling. Along with other materials such as $BaTiO_3$, a full characterization of the linear optical, nonlinear optical, electro-optic, dielectric, piezoelectric, elastic and elasto-optic tensor properties has been performed.

- We have developed first organic photorefractive materials, which have been based on doped organic crystals.

- We have investigated new very promising semiconducting ferroelectric $Sn_2P_2S_6$ crystals. We have determined its basic optical, electro-optic and nonlinear optical properties of this material and have shown its high potential for applications, due to e.g. twenty times higher electro-optic figures of merit compared to the inorganic standard $LiNbO_3$. For photorefractive applications, $Sn_2P_2S_6$ is particularly interesting due to the high sensitivity in the infrared (up to 1550 nm), as well as a very fast photorefractive response (millisecond compared to seconds of the conventional photorefractive ferroelectric oxide materials).

- We have investigated several photorefractive fixing mechanisms. We have developed a detailed theory of the effect of hologram fixing through high-temperature charge compensation or ionic fixing and proposed a new approach of photorefractive grating fixing at room temperature by means of ferroelectric domains (electrical fixing).

- We have performed first experimental and theoretical work on interband photorefractive effects, and have shown that it leads to the increase of the recording speed by two to three orders of magnitude compared to the conventional effect in the same material. Such gratings are also particularly robust against illumination at sub-bandgap photon energies and are therefore interesting for several applications.

- We have demonstrated several prototype photorefractive applications, including light deflection and modulation, associative memory, optical parallel processing, tunable filtering for WDM telecom applications, light-induced reconfigurable optical waveguides, photorefractive waveguides, as well as self-pumped optical phase conjugators and double phase conjugators with various materials and wavelengths.

Integrated Nonlinear Optics and Electro-optics

The Nonlinear Optics Laboratory has been the first laboratory to realize frequency-doubled blue compact all-solid-state lasers, an area which has matured now with commercial products available since years (including products from Rainbow Photonics AG, Zurich, a spin-off from the Nonlinear Optics Laboratory). These lasers have been realized by direct frequency doubling of $Ga_{1-x}Al_xAs$ laser diodes in $KNbO_3$ waveguides produced by He^+ ion implantation and lithographic structuring. More recently, the research efforts in this area have been focused on the development and investigation of all-solid-state continuous-wave UV lasers by optical frequency doubling in beta-barium borate (BBO) waveguides. For this purpose high quality channel waveguides were realized, optimized and their waveguiding and nonlinear optical properties characterized. Up to now we have demonstrated low loss waveguiding and the generation of 266 nm light in our BBO waveguides. In 2001, we started the new research effort in the area of electro-optical microresonators intended for high density electro-optical chips with highly integrated functions such as optical modulation, switching, wavelength multiplexing etc. After an intensive build-up time for this new activity (mainly thin film technology work and planar structuring), the first scientific results have been emerging recently. Highlights:

- Optical waveguides in $KNbO_3$ for the second harmonic generation of blue light in the wavelength region between 430 and 490 nm with a conversion efficiency of up to 23% W^{-1} cm^{-2} were developed.

- For the first time, ridge-type optical waveguides in BBO crystals were fabricated which is an important step towards development of integrated optics devices operating in the UV spectral region (200 - 350 nm). Two alternative techniques were developed for structuring of hygroscopic borate crystals: optical lithography and femto-second laser ablation. The second harmonic generation of UV light at 266 nm was demonstrated using the novel BBO waveguides.

- Development of "smart-guide" technique for fabrication of ion-sliced submicron thin single-crystal $LiNbO_3$ films sandwiched between low refractive-index transparent material (SiO_2 and/or benzocyclobutene - BCB) and metallic electrodes. These films possess optical and electro-optical properties comparable to bulk crystals. Due to a high electro-optical effect and a high refractive-index contrast (~ 0.7), the smart-guide films are suitable as a platform for future highly-integrated electro-optically active photonic devices based on microring resonators and photonic crystal structures.

- Realization of the first electro-optically tunable microring resonators in ion-sliced $LiNbO_3$ thin films and fluorine-implanted $LiNbO_3$ planar waveguides. Microring resonators with a free spectral range of up to 8 nm, a quality factor of 10'000 and a tunability of > 1 pm/V at wavelengths around 1.55 μm have been demonstrated.

- Combining laser lithography patterning and focused ion beam milling we have fabricated also first planar photonic crystal structures in ion-sliced

$LiNbO_3$ slabs. Triangular lattices of holes with a diameter of 240 nm and a separation of 500 nm exhibit a photonic bandgap with an extinction ratio of 15 dB for the TE polarization in the telecom wavelength range.

Organic Photonics and Electronics

Nonlinear optical thin film structures are key elements in future optical devices for realizing high-speed, large-capacity information transmission and processing. Despite the extremely large nonlinear optical and electro-optical effects of several organic materials, the production of high-quality organic thin films has been very difficult due to a large lattice mismatch and the weak interaction between the polar organic materials and the inorganic substrates. Using a novel technique - Organic Molecular Beam Deposition (OMBD) first developed by Prof. Z. Sitar - we have grown organic thin films which utilize sequential self-terminating growth of different organic monomers under ultra-high vacuum to fabricate highly pure, periodic nano-structures with unique homogeneity. The growth conditions (substrate temperature, deposition rates, stoichiometric ratios) are optimized by *in situ* atomic force microscopy, ellipsometry, mass spectrometry, and quartz microbalance as well as *ex situ* characterization including second harmonic generation and infra-red spectroscopy. Besides homoepitaxial growth as a first step, the successful realization of different heterosystems has proven the efficiency of the OMBD technique. The availability of such unidirectional structures is a major breakthrough in optical organic thin films

Physics and Applications of Terahertz Waves

Organic nonlinear optical crystals are used for the efficient generation of broadband, few-cycle terahertz (THz) pulses by optical rectification. In our laboratory, the best crystals (DAST, DSTMS and OH1) for this application have been developed. Using these crystals, a frequency range from 0.3 - 20 THz can be covered. Since these waves go through paper, packing material, plastic etc., they can be used to detect dangerous materials such as explosives, metal weapons, bio-threats even behind such materials. In addition, they also can be used for quality testing these materials. Most of the basic research has been completed at ETH. Within CTI start-up program together with our spin-off company Rainbow Photonics, a new THz instrument will be built up for such applications.

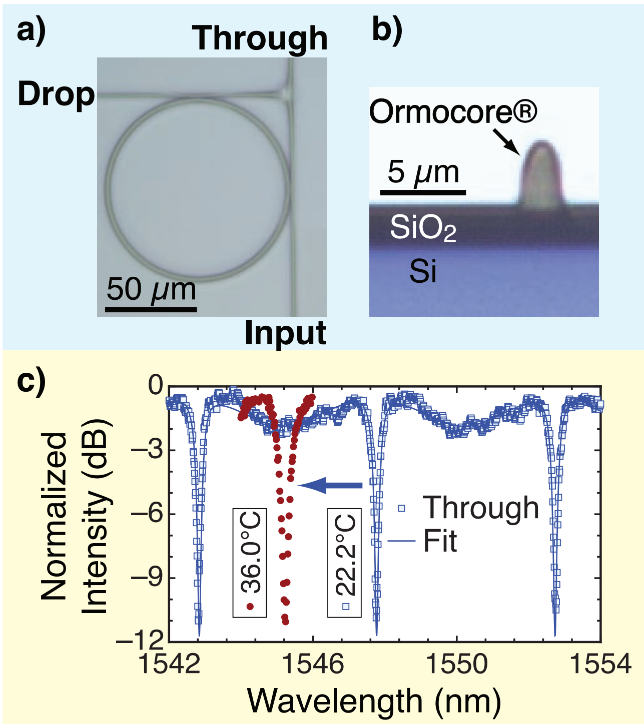


Figure 1.2: The basic design of a microresonator (a, microscope image from the top). Light guided in the input waveguide will be partially coupled into the microring. At resonator frequencies, the intensity of the light in the ring will increase dramatically, and will be coupled out into the drop port waveguide. A result of such a filtering observed by measuring the light intensity through the through port, while scanning the wavelength in the telecommunication range is shown in c). One can see sharp drops of the intensity when the resonance condition is satisfied. We could reduce the ring radius by one order of magnitude compared to typical polymeric rings with a similar filtering performance (finesse of about 20, extinction ratio 12 dB). The resonance could be thermo-optically tuned by 0.2 nm/°C (c).

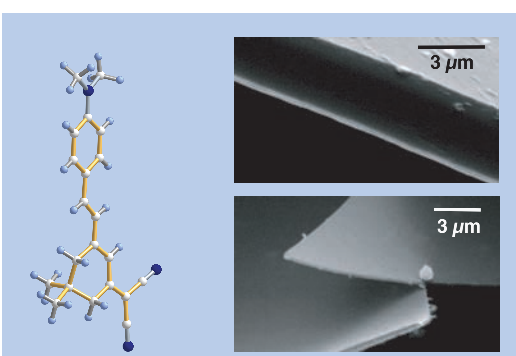


Figure 1.3: Molecular structure and SEM (scanning electron microscope) images of organic nonlinear optical single crystalline thin films of configurationally locked polyene chromophore, DAT2 (2-(3-(2-(4-dimethylaminophenyl)vinyl)-5,5-dimethylcyclohex-2-enylidene), grown by the vapor technique, with area of up to 5x3 mm^2, thickness in the range of 0.2-5μm, and sharp and flat edges.

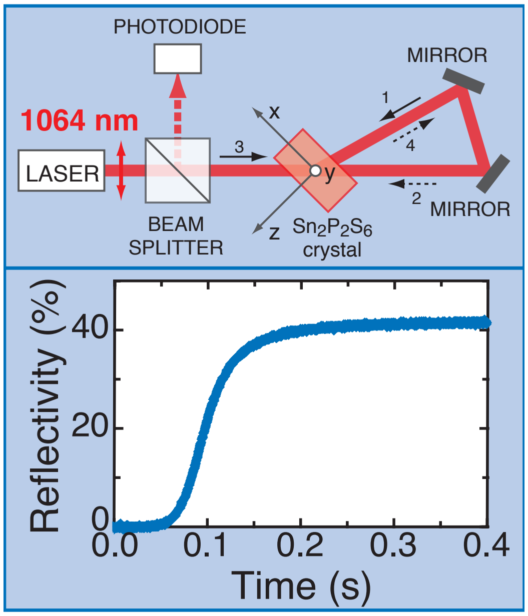


Figure 1.4: Phase conjugate reflectivity at 1064 nm of more than 40% in Te-doped $Sn_2P_2S_6$ with a rise time of below 100 ms at an intensity of 20 W/cm^2. This is more than 100 times faster than any previously reported value in other crystals.

1.1.4 Spin-off Companies

In 1988, the know-how for the crystal growth activities for producing $KNbO_3$ for nonlinear optical applications has been transfered to SANDOZ AG. A new unit called SANDOZ-OPTOELECTRONICS was founded which besides producing $KNbO_3$ crystals was also active (in collaboration with our laboratory) in the development of the first electro-optical polymers for optoelectronic applications. In 1990, VIRGO OPTICS Ltd. was bought by SANDOZ for producing $KNbO_3$ for the US market. Shortly before the merger of SANDOZ with CIBA, the Optoelectronics units were sold to other companies, SANDOZ HUNINGUE to CRYSTAL GmbH in Germany and VIRGO OPTICS to II-VI Inc. (VLOC) in the US. $KNbO_3$ crystals using our technology are still produced by these companies.
In 1997, Rainbow Photonics has been founded as a spin-off of the Nonlinear Optics Laboratory. The first products were direct diode doubled blue lasers using $KNbO_3$ and $KNbO_3$ waveguides. Later products included Brillouin cells for optical phase conjugation of high power solid state lasers, cw tunable Cr:LiSAF lasers and as the latest products, nonlinear optical materials for THz generation and THz instrumentation for security applications (DAST, DSTM, OH1, TeraSys 2000, TeraSys 4000).

OPTOELECTRONICS SANDOZ

Rainbow Photonics

Figure 1.5: Spin-off companies of the Nonlinear Optics Laboratory

Chapter 2

Personnel 1987 - 2009

Figure 2.1: In 1999, Prof. Peter Günter and Dr. Carolina Medrano at the group Christmas party

2.1 Overview

The following leaflet shows the time development of the staff of the Nonlinear Optics Laboratory. Interesting to note is, that the employment rate was roughly constant with a rate of 6.6 person/year, whereas the employment period varied among PhD candidates, postdocs and technicians. The employment rate was mainly determined by the amount of (external) funding and not by the large number of qualified masters students (average number of master students in the Nonlinear Optics Laboratory of 7 per year). Deviations from this rate occured in 1995 and 2005/2006 because of a sabbatical leave of Prof. Peter Gunter (see section 2.2). From 1987 until 2009 20 project leaders (senior assistants), 50 postdocs, 72 PhD students and 147 diploma (masters) students have been active in research in our laboratory. They were supported by 22 technicians and engineers and 7 administrative assistants.

Figure 2.2: In 2006, ECAPD'8 in Metz, France: How do we get there?

Figure 2.3: In 2007, Group party at Prof. Günter's home in Summer: It's always interesting when the boss is talking!

For the supervision of the PhD students and postdocs and the efficient management of the laboratory all the following project leaders (Oberassistenten) have to be acknowledged:

Prof. Dr. Raymond Kind	(- 31.10.2000)
Prof. Dr. Hanns Arend	(- 31.12.1990)
Dr. Dieter Suter	(01.04.1989 - 30.09.1995)
Dr. Carolina Medrano	(01.05.1989 - 31.01.2010)
Dr. Mathias Flörsheimer	(01.10.1989 - 30.04.1993)
Dr. Daniel Fluck	(01.01.1990 - 30.09.2000)
Dr. Jürg Hulliger	(01.01.1991 - 28.02.1993)
Dr. Marko Zgonik	(01.05.1991 - 30.09.1996)
Dr. Zlatko Sitar	(01.01.1992 - 31.08.1995)
Dr. Lukas Eng	(01.04.1993 - 30.06.1997)
Dr. Christian Bosshard	(15.08.1993 - 31.12.2000)
Dr. Ivan Biaggio	(01.01.1996 - 15.09.2002)
Dr. Germano Montemezzani	(01.10.1996 - 31.01.2004)
Dr. Mojca Jazbinsek	(01.12.2001 - 31.01.2010)

Dr. Rizwan Khan	(01.11.2003 - 19.01.2006)
Dr. Gorazd Poberaj	(01.01.2004 - 31.01.2010)
Dr. Arno Schneider	(01.08.2005 - 30.09.2009)
Dr. Blanca Ruiz Brunner	(01.01.2009 - 31.01.2010)

Besides these collaborators a very large number of visiting scientists greatly contributed to the success of our laboratory.

2.2 Sabbaticals

The group activities were slightly reduced during 1995/96 and 2005/06 because of the sabbatical leaves of Prof. Peter Günter. After these sabbaticals, some activities were redirected in new research areas. Particularly in 2000, many new research topics started, e.g. microring resonator research and nano photonics, physics of THz wave research, synthesis, growth and investigation of new ionic nonlinear optical molecular crystals, near infrared photorefractive physics based on a new ferroelectric semiconductor $Sn_2P_2S_6$, etc. These activities will be described in more detail in section 3 - 7.

Figure 2.4: In 2001, On Top of the Mountain!

2.3 Alumni

University Professors:

Prof. I. Biaggio, Lehigh University, USA

Prof. Ch. Cai, University of Houston, USA

Prof. R. Cudney, CICESE, Ensenada, Mexico

Prof. R. Khan, University of Surrey, UK

Prof. J. Hulliger, Universität Bern, Switzerland

Prof. G. Montemezzani, Universite de Metz, France

Prof. I. Poberaj, University of Ljubljana, Slovenia

Prof. L. Eng, Technische Universität Dresden, Germany

Prof. Z. Sitar, North Carolina State University, USA

Prof. D. Suter, Universität Dortmund, Germany

Prof. R. Wong, Hong Kong University

Prof. M. Zgonik, University of Ljubljana, Slovenia

Prof. R. Schlesser, North Carolina State University, USA

Prof. O.P. Kwon, Ajou University, Korea

Prof. Z. Yang, Bejing University of Science and Technology, China

Prof. G. Zhang, Nankai University, Tianjin, China

The activities of other alumni (PhD graduates) are shown in page 223 - 232 together with the titles of their PhD theses.

Figure 2.5: In 2000, At the Conference ICONO'5 (organized by the NLO laboratory), in Davos, Switzerland

Figure 2.6: In 1999, Group Christmas party: Let's dance together!

Chapter 3

Organic Nonlinear Optics

Organic nonlinear optical materials present a relatively new class of functional materials with large and extremely fast nonlinearities compared to their inorganic alternatives. These materials additionally offer a wide variety of design possibilities and therefore a wide spectrum of material properties. The research on organic materials for photonic elements is strongly motivated by the need for the development of high transmission bandwidths and wavelength division multiplexing systems in telecommunication technologies.

We have been involved in several aspects of organic functional materials research. We have designed and synthesized several novel organic nonlinear optical molecules, embedded them in polymers, single crystals or self-assembled films, characterized their microscopic and macroscopic optical and nonlinear optical properties, as well as their thermal and photochemical stability, and finally demonstrated their functionality in several application schemes, including integrated electro-optic amplitude and phase modulation, active microring resonator filtering and modulation, frequency conversion and THz wave generation and detection. We reviewed the research in the field of organic second- and third-order nonlinear optics in several books and invited book chapters. Our main achievements are summarized in the following sections.

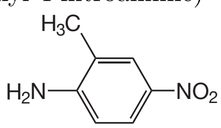
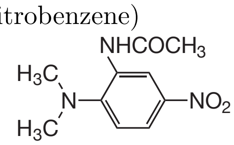
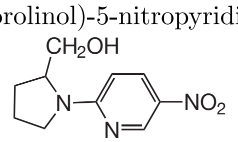
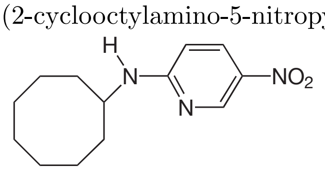
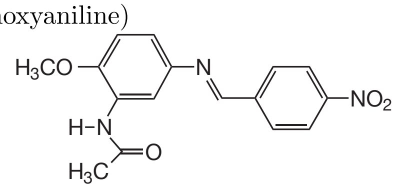
3.1 Second-Order Organic Nonlinear Optical Crystals

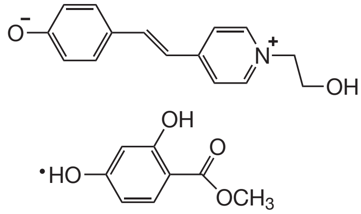
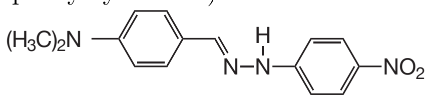
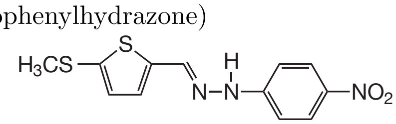
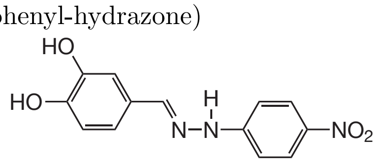
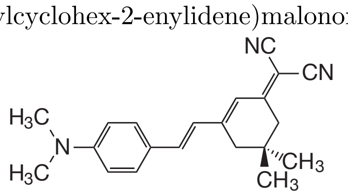
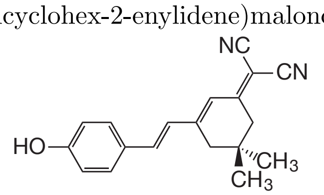
For highspeed second-order nonlinear optical applications, such as electro-optics, second-harmonic generation, optical parametric oscillation, and optical rectification, including THz wave generation, a highly asymmetric electronic response of the material to the external electric field is required. Second-order nonlinear optical organic materials are most often based on π-conjugated molecules (chromophores) with strong electron donor and acceptor groups at the ends of the π-conjugated structure. Such molecules must be ordered in an acentric manner in a material to achieve a macroscopic second-order nonlinear optical response. One possibility to obtain an efficient macroscopic second-order active nonlinear optical organic material is to order the nonlinear optical molecules in an acentric structure by crystallization. The advantages of single crystals compared to other methods are that they allow for a maximum possible chromophore packing density and best thermal and photochemical stability [Rezzonico et al., 2008]. On the other hand, acentric crystallization for these molecules is very challenging, since they are highly polar and therefore tend to aggregate in a centrosymmetric crystalline arrangement. We have been working on four different molecular-design approaches to achieve a noncentrosymmetric crystalline packing of highly nonlinear optical molecules. We have also been investigating and optimizing the processing characteristics, e.g., growth of large size bulk or thin film crystals, which will make such materials useful for applications.

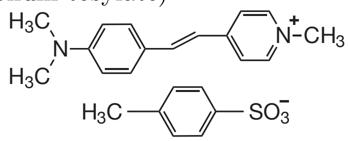
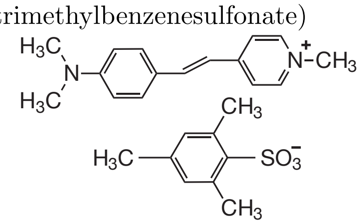
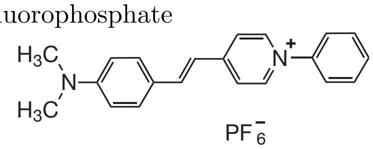
The introduction of molecular asymmetry or chirality. This is a most often employed strategy to achieve non-centrosymmetric packing. The supra molecular packing will be governed by steric interactions and Van der Waals forces. Molecules tend to undergo shape simplification during crystal growth, which gives rise to dimers and then to high-order aggregates in order to adapt to a close-packing in the solid state. The high tendency of achiral molecules to crystallize centrosymmetrically could be due to such a close-packing driving force. Therefore, if the symmetry of the chromophores is reduced, dimerization and subsequent aggregation is no longer of advantage to the close packing and increases the probability of acentric crystallization. This symmetry reduction can be accomplished either by the introduction of molecular (structural) asymmetry or the incorporation of steric (bulky) substituents into the chromophore. Additionally, hydrogen bond functionalities may be included to promote a desired chromophore packing.
In the earlier stages, we were investigating several organic nonlinear optical crystals employing this strategy, including COANP, DAN, PNP, MNA and MNBA (see Table 3.1). These so-called "yellow" materials typically present up to two orders of magnitude higher non-resonant optical nonlinearity compared to the

urea standard which is colorless. In later stages, this strategy was also used for higher-nonlinearity (”red”) chromophores.

Table 3.1: Most important organic nonlinear optical crystals investigated in our laboratory. λ_c is the cut-off wavelength in the bulk, d is the nonlinear optical coefficient, r is the electro-optic coefficient, and T_m is the melting point.

Material	Point group	λ_c(nm)	d, r (pm/V)	T_m (°C)
Cut-off wavelength 450 nm $\leq \lambda_c \leq$ 550 nm				
MNA(2-methyl-4-nitroaniline) 	m	480	d_{11}(1064 nm)= 150 d_{12}(1064 nm)= 23 r_{11}(633 nm)= 67	133
DAN(4-(N,N-dimethylamino)-3-acetamidonitrobenzene) 	2	485	d_{23}(1064 nm)= 38 r_{32}(633 nm)= 13	165.7
PNP(2-(N-prolinol)-5-nitropyridine) 	2	490	d_{21}(1064 nm)= 51 d_{22}(1064 nm)= 16.2 r_{22}(514 nm)= 28.3 r_{22}(633 nm)= 12.8	83
COANP(2-cyclooctylamino-5-nitropyridine) 	$mm2$	490	d_{32}(1064 nm)= 32 r_{33}(514 nm)= 28 r_{33}(633 nm)= 15	72.8
MNBA(4’-nitrobenzylidene-3-acetamino-4-methoxyaniline) 	m	520	d_{11}(1064 nm)= 131 r_{11}(532 nm)= 50 r_{11}(633 nm)= 29	-

Material	Point group	λ_c(nm)	d, r (pm/V)	T_m
Cut-off wavelength $\lambda_c \geq 550$ nm				
Mero-2-MDB(4-2-[1[(2-hydroxyethyl)-4-pyridylidene]-ethylidene-cyclo-hexa-2,5-dien-1-one-methyl-2,4-dihydroxybenzoate) 	m	phase II: 680 phase I: 615	phase II: d_{11}(1318 nm)= 267 r_{11}(1313 nm)= 34 phase I: d_{11}(1318 nm)= 108 r_{11}(1313 nm)= 24	185
DANPH(4-dimethylaminobenzaldehyde-4-nitrophenylhydrazone) 	m	670	d_{12}(1542 nm)= 200 d_{11}(1542 nm)= 150	186
MTTNPH(5-(methylthio)-thiophenecarboxaldehyde-4-nitrophenylhydrazone) 	$mm2$	620	d_{32}(1313 nm)= 32	172
3,4-DHNPH(3,4-dihydroxybenzaldehyde-4-nitrophenyl-hydrazone) 	m	730	-	230
DAT2(2-(3-(2-(4-dimethylaminophenyl)vinyl)-5,5-dimethylcyclohex-2-enylidene)malononitrile) 	2	700	r_{22}(1550 nm) $\approx$ 10	235
OH1(2-(3-(4-hydroxystyryl)-5,5-dimethylcyclohex-2-enylidene)malononitrile) 	$mm2$	650	d_{33}(1907 nm) = 120 r_{33}(1319 nm) = 50 r_{33}(1064 nm) = 56 r_{33}(785 nm) = 75 r_{33}(633 nm) = 109	212

Material	Point group	λ_c(nm)	d, r (pm/V)	T_m
DAST(4-N, N-dimethylamino-4'-N'-methyl-stilbazolium tosylate) 	m	700	d_{11}(1318 nm)= 1010 d_{11}(1542 nm)= 290 d_{11}(1907 nm)= 210 d_{26}(1542 nm)= 39 d_{26}(1907 nm)= 25 r_{11}(720 nm)= 92 r_{11}(1313 nm)= 53 r_{11}(1535 nm)= 47	256
DSTMS(4-N, N-dimethylamino-4'-N'-methyl- stilbazolium 2,4,6-trimethylbenzenesulfonate) 	m	700	d_{11}(1907 nm)= 214 d_{12}(1907 nm)= 31 d_{26}(1907 nm)= 35	256
DAPSH(trans-4'-(dimethylamino)-N-phenyl-4- stilbazolium hexafluorophosphate 	m	750	d_{11}(1907 nm)= 290	-

To obtain a large molecular second-order nonlinearity, long conjugated polyene chromophores provide one of the most effective pathways for efficient charge delocalization between donor and acceptor groups. We investigated several nonlinear optical chromophores based on configurationally locked polyene (CLP) chromophores that consist of a π-conjugationed hexatriene bridge between various electron donors and a dicyanomethylidene electron acceptor; two promising examples are shown in Fig. 3.1 and 3.2.

Use of strong Coulomb interactions. Coulomb intermolecular forces are used for charged chromophores in combination with counter-ions that tend to override the weaker dipole-dipole interactions and promote a non-centrosymmetric packing. Our research in this field was based on stilbazolium chromophore crystals. Molecular engineering of stilbazolium salts is one of the most effective approaches for the development of highly efficient second-order nonlinear optical organic crystalline materials. DAST (4-N,N-dimethylamino-4'-N'-methyl-

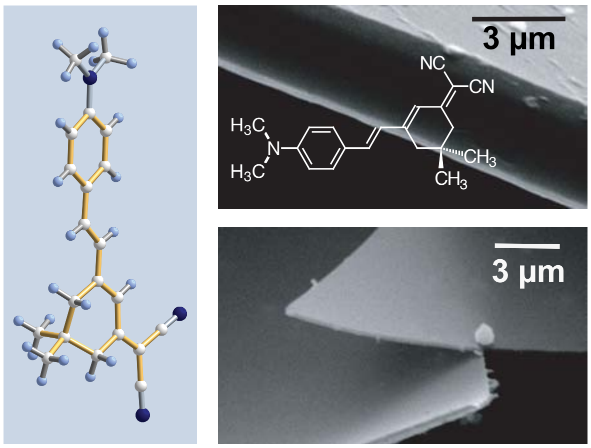


Figure 3.1: CLP chromophore DAT2 (2-{3-[2-(4-dimethylaminophenyl)-vinyl]-5,5-dimethylcyclohex-2-enylidene}malononitrile) has very favorable thin-film crystallization properties from solution, melt and vapor phase. The pictures on the right present scanning electron microscope images of free-standing DAT2 crystals grown by the vapor technique [Choubey et al., 2007], which have an area of up to 5x3 mm^2, thickness in the range of 0.2–5 μm, and sharp and flat edges.

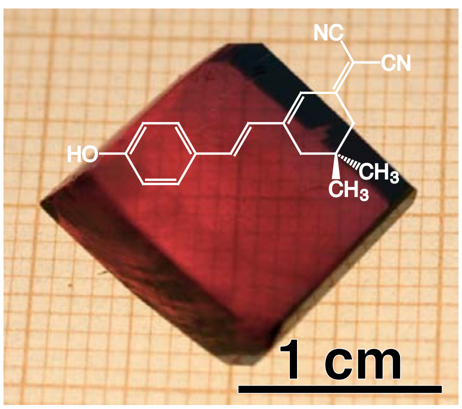


Figure 3.2: CLP crystals OH1 (2-(3-(4-hydroxystyryl)-5,5-dimethylcyclohex-2-enylidene)malononitrile) contain a phenolic electron donor, which also acts as a hydrogen bond donor. The OH1 crystals exhibit large second-order nonlinear optical figures of merit, high thermal stability and very favorable crystal growth characteristics. Higher solubility in methanol compared to analogous CLP crystals is of advantage for solution crystal growth, yielding acentric bulk OH1 crystals of large sizes with side lengths of more than 1 cm with excellent optical quality [Kwon et al., 2008]. A very high potential of OH1 crystals for electro-optic modulators and THz wave emitters has been demonstrated.

stilbazolium tosylate), with a powder second-harmonic-generation efficiency of three orders of magnitude larger than that of the urea standard at 1907 nm, is the best known organic nonlinear optical crystal (see review paper [Jazbinsek et al., 2008]). Several research projects in our group were based on DAST, including the growth of high-quality bulk and thin film crystals (see Fig. 3.3), characterization of DAST properties relevant for nonlinear optics and electro-optics, development of integrated optical modulators, as well as THz wave generation and detection (see chapter 4). We have also designed, synthesized, grown and characterized several DAST derivatives in order to further enhance the nonlinear optical figures of merit and simultaneously achieve superior growth characteristics; examples are shown in Fig. 3.4 and 3.5.

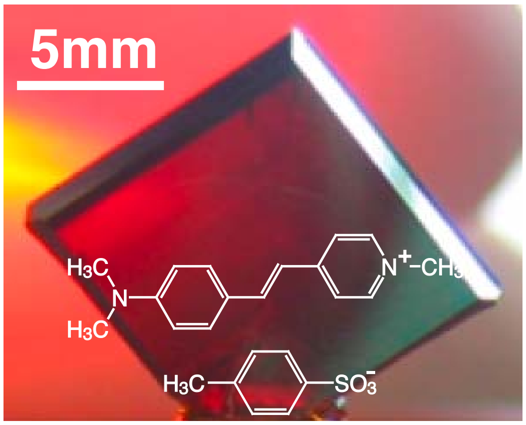


Figure 3.3: We developed an optimized method to nucleate and grow high-quality DAST crystals from methanol solution by low-temperature solution growth. We could achieve a constant growth rate based on a complete thermodynamic characterization of the crystal-solution system. High optical quality DAST crystals up to $30 \times 30 \times 10$ mm^3 size were grown in our laboratory [Ruiz et al., 2008].

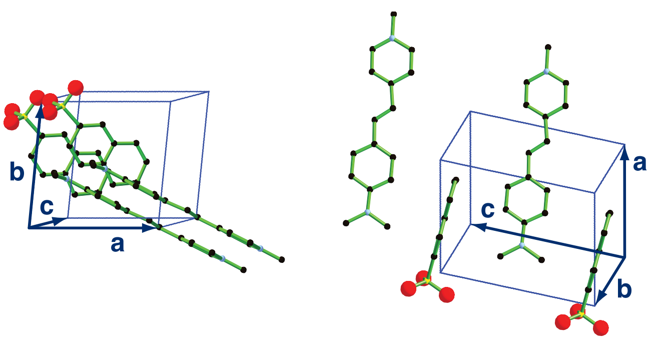


Figure 3.4: Stilbazolium derivative DSNS (4-N, N-dimethylamino-4'-N'-methyl-stilbazolium 2-naphthalenesulfonate) with record-high nonresonant second-harmonic-generation efficiency, 1.5 times as that of DAST at 1.9 μm, was developed. The stilbazolium chromophores in DSNS are aligned perfectly parallel, yielding maximum possible projection of the microscopic to the macroscopic optical nonlinearities [Ruiz et al., 2006].

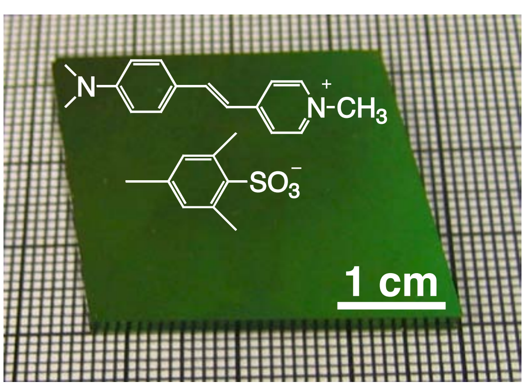


Figure 3.5: Stilbazolium derivative DSTMS (4-N, N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate) shows the best crystal growth characteristics among the investigated salts. The growth of high quality crystals from methanol solution is relatively easy and faster compared to DAST [Yang et al., 2007]. Measurements of the linear and nonlinear optical properties of DSTMS have shown that it is a very attractive alternative to DAST, since its second-order optical nonlinearities are similar or even higher [Mutter et al., 2007]. DSTMS also shows an improved THz generation spectrum with a larger bandwidth and smaller absorption in the THz range compared to DAST [Stillhart et al., 2008].

Use of a relatively flexible π-conjugated core. In contrast to donor-acceptor disubstituted stilbene derivatives, hydrazone derivatives generally adopt a bent, non-rod-shaped conformation in the solid state because of the non-rigid nitrogen-nitrogen single bond (–CH=N–NH–). We have found that donor-substituted (hetero)-aromatic aldehyde-4-nitrophenylhydrazones show an overwhelmingly high propensity for a non-centrosymmetric packing. Of particular importance is that the majority of these acentric crystals exhibit very strong second-harmonic signals that are at least two orders of magnitudes greater than that of the urea standard. Furthermore, most of the hydrazone crystals we developed show very good crystallinity and high thermal stability [Wong et al., 1996]. The best example in this class is 4-dimethylaminobenzaldehyde-4-nitrophenylhydrazone, DANPH (Fig. 3.6), which exhibits a very strong second-harmonic signal that is comparable to that of DAST.

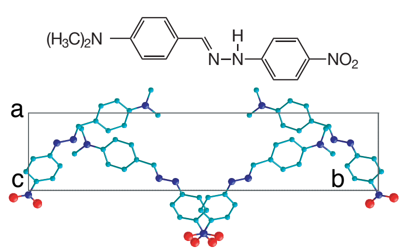


Figure 3.6: Hydrazone nonlinear optical crystal DANPH (4-dimethylaminobenzaldehyde-4-nitrophenyl-hydrazone) has a crystalline packing optimal for phase-matched frequency conversion, for which large off-diagonal susceptibility components are desired. The second-order susceptibility element $\chi^{(2)}_{122}(-2\omega,\omega,\omega)$ reaches 380 pm/V at 1.54 μm, which is about 70% of the largest diagonal coefficient of DAST, but almost five times larger than the largest off-diagonal component of DAST. An effective nonlinear-optical coefficient $\chi^{(2)}_{\text{eff}}(-2\omega,\omega,\omega) = 210$ pm/V at 1.54 μm for phase-matched frequency doubling with DANPH is the largest reported phase-matchable coefficient [Follonier et al., 1997].

Supramolecular synthetic approach. Using this approach, molecular or ionic aggregates or assemblies, most commonly composed of two complementary molecules, are designed to favor a desirable crystal packing. This approach offers more design flexibility, as one or both molecules can be tailor-made or modified to fit one another to acquire the desirable molecular properties in the solid state. Furthermore, the physical properties like melting point and solubility as well as the crystal properties like crystallinity and ease of crystal growth of the co-crystals can usually be improved compared to those of their starting components.

We have found that the co-crystals formed from the merocyanine dyes (Mero-1 and Mero-2) and the class I phenolic derivatives (see Fig. 3.7), in which the electron acceptor is *para*-related to the phenolic functionality together with a substituent either in the *ortho*- or *meta*-position show the highest tendency of forming acentric co-crystals. In addition, a relatively large fraction of acentric co-crystals (25%) based on Mero-2 and the class I phenolic derivatives exhibit strong second-harmonic signals that are at least two orders of magnitudes larger than that of urea. Two most interesting examples of merocyanine co-crystals are shown in Fig. 3.8 and 3.9.

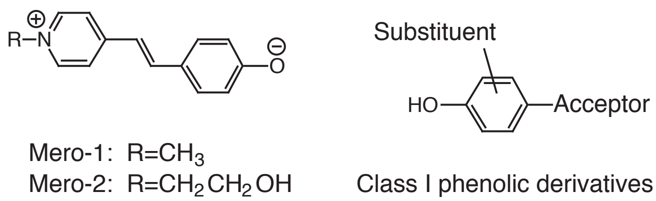


Figure 3.7: Chemical structures of Mero-1, Mero-2 and Class I phenolic derivatives.

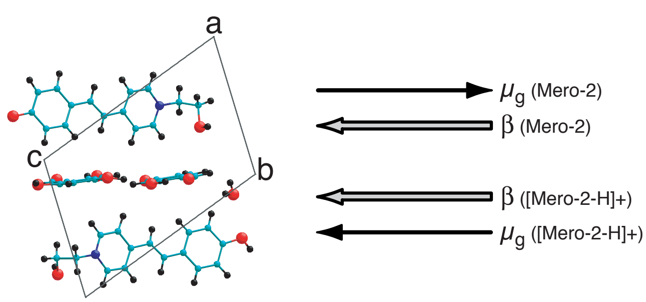


Figure 3.8: An interesting example of merocyanine co-crystals is the hydrated co-crystal Mero-2-DBA (DBA = 2,4-dihydroxy-benzaldehyde). The anionic assembly is constructed by the co-aggregation of two DBA molecules, in which one of the molecules gives up a proton and bonds to another by a hydrogen bond. Mero-2 acquires the proton and co-aggregates in anti-parallel fashion with another Mero-2 by a short hydrogen bond constituting a cationic assembly. Although the net dipole moment almost vanishes in this arrangement, the Mero-2-DBA co-crystal exhibits a large second-harmonic signal in the powder test. This can be explained by the asymmetric position of the hydrogen bonded proton between the two Mero-2 dyes, which results in a positive reinforcement of molecular hyperpolarizabilities within the cationic assembly [Pan et al., 1996].

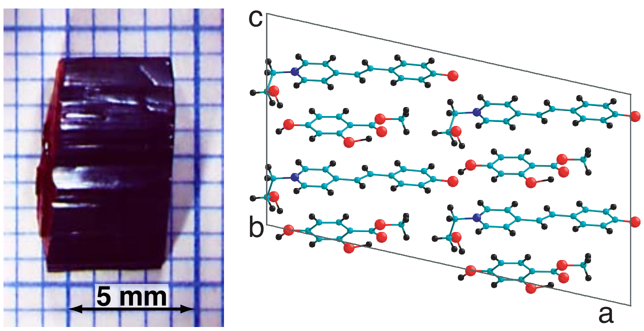


Figure 3.9: Merocyanine co-crystal Mero-2-MDB exhibits an almost parallel alignment of the chromophores, resulting in a high nonlinear optical susceptibility $\chi^{(2)}_{111}(-2\omega, \omega, \omega) = 540$ pm/V at 1.3 μm [Wong et al., 1998]. The molecular aggregate is assembled through short hydrogen bonds. The rod-like aggregates connect laterally by hydrogen bonds resulting in a staircase-like polar chain. These polar chains align in a parallel fashion constituting a two-dimensional acentric layer, which we found also in other highly non-centrosymmetric co-crystals of this class [Wong et al., 1997].

3.2 Electro-optic Polymers and Thin Films

Since the crystallization of nonlinear optical chromophores in a desired orientation is very challenging, a more common approach is based on their integration into a polymer matrix, in which applying a strong electric field above the glass transition temperature may induce a non-centrosymmetric arrangement of the polar molecules. However, this will result in a limited thermal stability of chromophore orientation. We developed several electro-optic polymer systems and investigated their thermal orientational stability and photochemical stability. We also investigated a Langmuir-Blodgett-film approach based on depositing acentric molecular monolayers on substrates from solution.

Langmuir-Blodgett nonlinear optical thin films. The optical and structural properties of ultrathin organic molecular films are essential for the potential applications in optical waveguide structures and filters. One method, the so-called Langmuir-Blodgett (LB) technique, allows the 2-dimensional aggregation of LB molecules on both the solid and the water surface. The most outstanding advantage of this technique is the direct control of molecular assembly processes on the molecular level. We therefore set up optical techniques for both the in-situ and ex-situ investigation of mono- and multilayer LB films. Linear optical (polarization) and non-linear optical (second-harmonic) microscopy were used to complement the scanning probe techniques, which are sensitive to the molecular assembly at the sample surface. In addition, a scanning force microscope (SFM) has been developed that enabled investigating even the liquid/air interface in order to analyze a floating LB monolayer. Optical and structural analysis has been applied to a variety of new LB molecules that have been synthesized to yield a high value for the nonlinear optical susceptibility $\chi^{(2)}$. We could also demonstrate guided-wave second-harmonic generation in nonlinear optical LB films for the first time by Cerenkov-type phase matching (Fig. 3.10) and by mode conversion (Fig. 3.11).

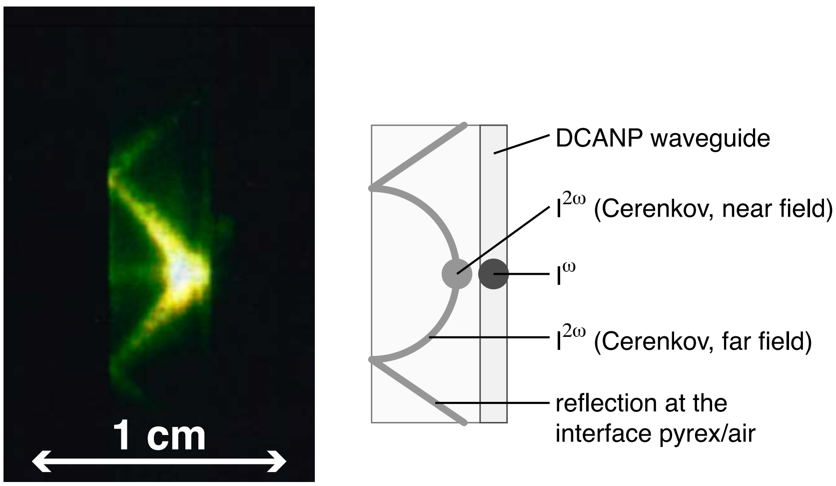


Figure 3.10: Photograph of phase-matched Cerenkov radiation from a LB film of DCANP (2-docosylamino-5-nitropyridine) on a pyrex glass substrate and experimental configuration (fundamental beam at 1064 nm and frequency-doubled at 532 nm). The frequency-doubled wave leaves the substrate through its edge [Bosshard et al., 1991].

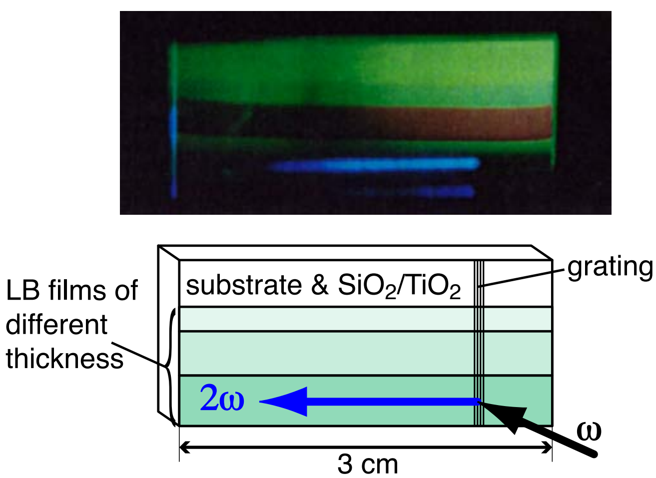


Figure 3.11: Photograph of the guided second-harmonic wave and experimental configuration (top view). The fused silica substrate with the SiO_2/TiO_2 (main guiding layer) is coated with DCANP LB films of different thickness stepwise (films of different thickness can be distinguished by their different interference colors). Only the LB film with the appropriate thickness for the mode matching (199 nm in this case) leads to an efficient conversion into blue as seen in the photograph (the fundamental light is at 926 nm and therefore not visible) [Flörsheimer et al., 1992].

Electro-optic polymers. Polymers present an important class of nonlinear optical materials as they combine the nonlinear optical properties of conjugated π-electron systems with the feasibility of creating new materials with appropriate optical and structural properties. The incorporation of nonlinear optical molecules in polymers is relatively easy and can be done in different ways. The simplest one is the mixing of the active molecules in a polymer matrix forming a guest-host system. To increase the chromophore density and orientational stability without phase separation, the chromophores can be covalently linked to a polymer backbone in the form of a side-chain, main-chain or cross-linked between two polymer chains.

Polyimide side-chain electro-optic polymers based on disperse red (DR1) chromophores (see Fig. 3.12) were developed in the early nineties through our collaboration with Sandoz Optoelectronics Research [Prêtre et al., 1994]. The

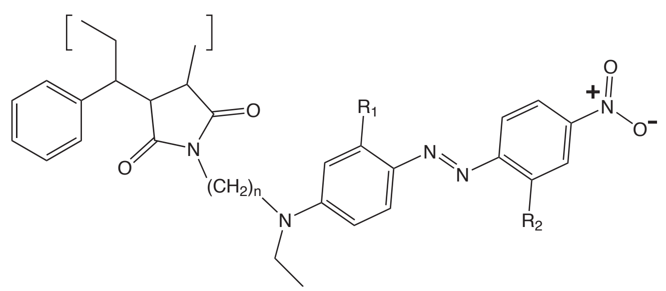


Figure 3.12: Molecular structure of electro-optical side-chain polyimides based on disperse red.

alkyl-amino-functionalized nonlinear optical azo chromophores with various substituents, see e.g. Table 3.2, were attached via a two- or three-carbon spacer linkage to an alternating styrene-maleic-anhydride copolymer.

	n	R_1	R_2	T_g (°C)
A-095.11	3	CH_3	H	137
A-097.07	3	CH_3	Cl	149
A-148.02	2	H	H	172

Table 3.2: Azo dye substitution patterns and glass transition temperatures T_g of selected modified polyimide polymers.

The glass transition temperature and the nonlinear optical properties of nonlinear polymers depend to a large extent on the chromophore concentration and also on the poling field parameters. For example, polymer A-095.11 with a chromophore concentration of 56 wt.% resulted in $T_g = 137°C$, a nonlinear optical susceptibility $\chi^{(2)}_{33}(-2\omega, \omega, \omega) = 68$ pm/V at $\lambda = 1.54$ μm, and an electro-optic coefficient of $r_{33} = 20$ pm/V at $\lambda = 1.313$ μm [Prêtre et al., 1994].

Thermal relaxation of chromophore orientation in poled polymers. Understanding relaxational processes in nonlinear optical polymeric materials is of critical importance in order to evaluate their long-term stability for potential electro-optic applications. An essential requirement for stabilizing polymeric nonlinear optical materials is the formation of a glassy state at relatively high glass-transition temperatures T_g.

Relaxation processes in nonlinear optically active modified polyimide polymers with side-chain azo chromophores presented above, having glass transition temperatures in the range of $140°C < T_g < 170°C$ have been studied by differential scanning calorimetry, dielectric relaxation, and second-harmonic generation experiments. These experiments revealed important information on stability. We have shown that it is possible to model the orientational relaxation behavior of nonlinear optical chromophores both above and below the glass transition over more than 15 orders of magnitude in time using the Tool-Narayanaswamy procedure incorporating the appropriate Williams–Landel–Ferry (WLF) parameters for the nonlinear optical polymers [Kaatz et al., 1996]. This led to a scaling prediction for relaxation times in the glassy state with the scaling parameter $(T_g - T)/T$.

The relaxation time for a certain material type therefore only depends on one parameter: T_g. The underlying theory allowed to define the requirements of T_g for the required stability. As an example, for a polymer with $T_g = 172°C$ the relaxation time at an operating temperature of 80°C is about 100 years. With a T_g of 200°C, this relaxation time increases dramatically (see Fig. 3.13). Annealing of polymers at elevated temperatures further increases the orientational stability [Prêtre et al., 1998].

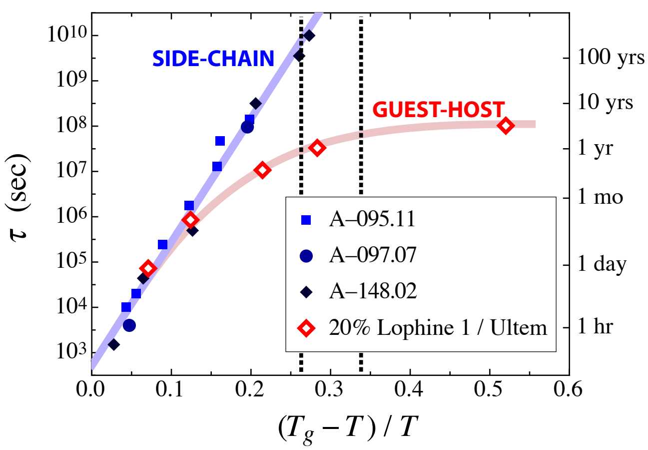


Figure 3.13: Temperature scaling of normalized second-harmonic generation relaxation times with respect to the scaling variable $(T_g - T)/T$ of three disperse-red-based side-chain polyimides and one guest-host polymer (20% Lophine 1/Ultem). It is clearly seen that the nonlinear optical moiety has to be coupled to the polymer chain for increased stability. The two vertical lines are related to examples discussed in the text.

Photochemical stability. Photochemical stability of organic nonlinear optical chromophores is one of the most important issues for applications, especially in cases when interaction with resonant light is involved, but also at telecommunication wavelengths if relatively high light intensities are employed, e.g. in waveguiding structures.

Photochemical degradation of chromophores is a complex process that may include several intermediate products, several degradation pathways, and even partially reversible processes, e.g., trans-cis isomerization. The chromophore lifetime is proportional to the photostability figure of merit $B/\sigma(\omega)$, where $1/B$

is the probability for degradation of a photoexcited molecule, and $\sigma(\omega)$ the absorption cross section of the chromophore. In most previous experiments, the figure of merit B/σ was evaluated assuming a single dominant charge-transfer state with a single decay channel, monitoring only the initial evolution of the photodegradation process. However, the full analysis of the photodegradation kinetics that we developed[Rezzonico et al., 2007] shows that these simplified models may result in overestimations of photostability lifetimes by up to a factor of two. Additionally, our model gives additional important information on the photodegradation processes, including the existence of single and double degradation pathways, as well as direct and indirect degradation processes, which is of importance when considering the development of more photochemically robust chromophores for use in nonlinear optical devices.
We have also directly compared the photochemical stability of nonlinear optical chromophores in polymeric and single-crystalline environments. Under resonant light excitations, DAT2 chromophores in crystalline environment exhibit three orders of magnitude better photostability as compared to guest-host polymer composites, as shown in Fig. 3.14.

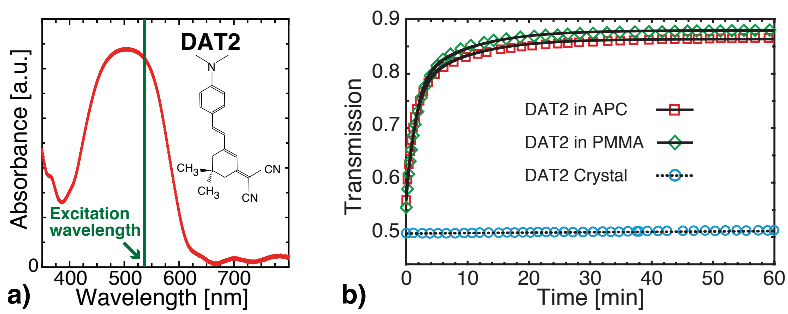


Figure 3.14: a) Absorption spectrum of DAT2 in the amorphous polycarbonate (APC) polymer. b) Photostability measurement for DAT2 guest-host polymers (APC and PMMA) and for DAT2 single crystal illuminated with 532-nm light, showing that the photochemical stability in crystalline environment is much superior to polymeric environments [Rezzonico et al., 2008].

To better understand the mechanisms involved in photodegradation, we performed measurements in different ambient conditions, i.e. in oxygen, air, nitrogen and vacuum. For the bithiophene chromophore CC172 [Cai et al., 1999] in poly(methyl methacrylate) (PMMA) for example, the change from air to pure nitrogen increases the stability by one order of magnitude over the whole wavelength range. This already indicates the importance of photo-oxidative processes in photodegradation. We gained an additional order of magnitude when changing from nitrogen to vacuum. This suggests that even in nitrogen reactive processes lead to photodegradation, i.e., that the degradation processes are mainly triggered by the surrounding atmosphere. Based on these results, the photochemical stability of electro-optical polymers with a novel cover layer has been investigated. This cover layer with a thickness of only 50 nm is impermeable for oxygen, nitrogen, water vapor and other gases and has been fabricated by plasma deposition. With this cover layer we have demonstrated a more than two orders of magnitude improved photochemical stability compared to the uncoated polymers. This increase is attributed to the protection of the gas-barrier layer against photo-oxidation and reactions with nitrogen.

3.3 Third-Order Organic Nonlinear Optical Materials

Third-order nonlinear optical effects present the dominant nonlinearity of centrosymmetric materials and manifest themselves in various frequency mixing phenomena, as well as the change of the refractive index induced by the light beam itself (optical Kerr effect). With such interactions, light beams can be switched or steered by purely optical means. The all-optical switching speed is as fast as the origin of the nonlinearity, which is in the case of electronic nonlinearities in the femtosecond range. Therefore large switching rates can be reached, interesting for applications in optical fiber networks.

We mainly investigated three different molecular systems: polyenes, polytriacetylenes, and tetraethynylethenes. All of them have an extended conjugation of delocalized electrons and the possibility to substitute the endgroups of the conjugation by an electron donating or accepting functional group. In polyenes, we obtained particularly large nonlinearities. For asymmetrically electron donor-acceptor substituted polyenes with elongated conjugation, second-order hyperpolarizabilities of up to $\gamma = 300 \cdot 10^{-48}$ m^5/V^2 ($21400 \cdot 10^{-36}$ esu) were measured in the two-photon resonance. For all-optical applications, resonantly enhanced nonlinearities are not interesting due to the loss mechanism connected to any resonance. We reached values for such nonlinearities of up to $\gamma = 100 \cdot 10^{-48}$ m^5/V^2 ($7400 \cdot 10^{-36}$ esu) for polyenes in degenerate four-wave mixing experiments.

We also investigated the relations between the molecular structure and the third-order nonlinear optical properties, which is a prerequisite to develop optimization guidelines on the way to applications. In polytriacetylenes, a power law scaling of the nonlinearity vs. the elongation of the molecular backbone was observed with an exponent $a = 2.5 \pm 0.1$ [Gubler et al., 1999]. The power law saturates for 60 carbon-carbon bonds to an only linear increase of the nonlinearity vs. conjugation length, as shown in Fig. 3.15. This behavior of the nonlinearity is generally observed for conjugated polymers outside resonance with the same exponent, and the saturation length is independent of the exact backbone structure. We could point out this agreement between the various conjugated backbone systems for the first time. Pure polymer films of polytriacetylene can be produced showing nonlinearities $\chi^{(3)} = 1.5 \cdot 10^{-20}$ m^2/V^2 ($1.1 \cdot 10^{-12}$ esu) comparable with the best polymers measured outside resonance enhancement.

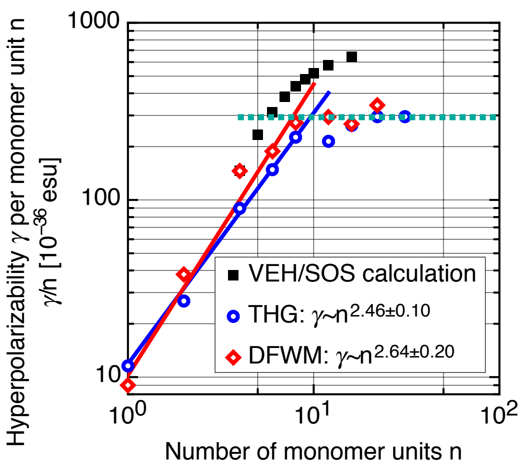


Figure 3.15: The third-order polarizabilities of the polytriacetylene molecules show a power law increase for short oligomeric lengths with a smooth saturation around 10 monomer units, for DFWM (degenerate four-wave mixing), THG (third-harmonic generation), and quantum-chemical calculations for n ranging between 5 and 16. In the saturation regime, the nonlinearity increases linearly upon backbone elongation [Gubler et al., 1999].

In tetraethynylethenes, the largest nonlinearities were measured for asymmetric substitution patterns with donor and acceptor groups. We could explain this fact in a quantum mechanical perturbation approach by the symmetry of the electronic wavefunctions, which has been demonstrated for two-dimensionally conjugated molecules for the first time [Gubler et al., 1998]. Second-order hyperpolarizabilities of up to $\gamma = 14 \cdot 10^{-48}$ m^5/V^2 ($1000 \cdot 10^{-36}$ esu) were measured in third-harmonic generation experiments.

Cascading of second-order nonlinearities. Large, nonresonant optical Kerr-like nonlinearities are a basic requirement for all-optical switching and related applications. Besides the nonlinearity which is proportional to $\chi^{(3)}$, there are other important contributions that only occur in noncentrosymmetric materials and are illustrated in Fig. 3.16. This contribution induces a nonlinear phase shift in nearly phase-matched second-harmonic generation interactions and other parametric processes. Such effects have a high potential for all-optical switching, since very large nonlinear phase shifts due to cascading can be obtained. We demonstrated that there exist other combined processes of second-order nonlinear optical effects that give rise to a large effective nonlinear refractive index n_2

without the need of phase-matching. In particular, we have shown both theoretically and experimentally that the combined processes of optical rectification and the linear electro-optic effect lead to an effective n_2 in noncentrosymmetric materials [Bosshard et al., 1995]. We also analyzed in detail direct third-order and cascaded second-order contributions to third-harmonic generation in various polar materials; an example is shown in Fig. 3.17.

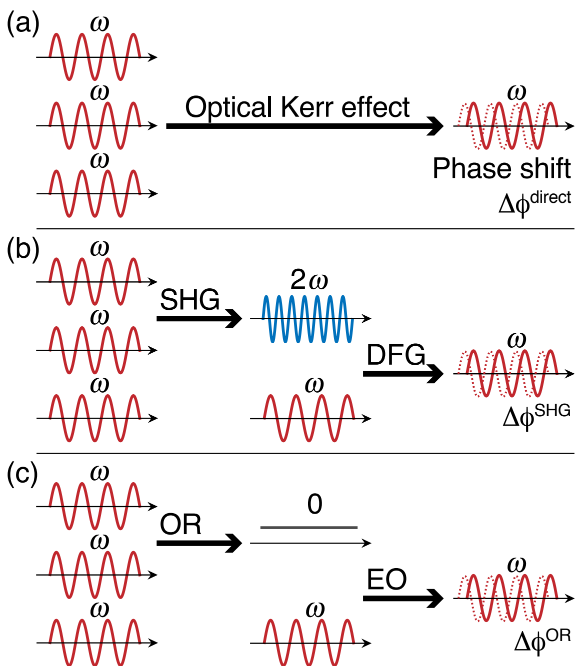


Figure 3.16: Schematic illustration of different possibilities leading to a nonlinear phase shift: (a) a direct optical Kerr effect; (b) cascaded second-harmonic generation (SHG) and difference-frequency gene-ration (DFG); (c) optical rectification (OR) and electro-optic (EO) effect.

3.4 Investigations of Linear Optical, Electro-optic and Nonlinear Optical Properties of Organic Materials

Nonlinear optical characterization of organic molecules. In organic materials, the macroscopically observed nonlinear optical and electro-optic effects can be described as originating from the nonlinear optical properties of their constituent molecules. The basic quantities describing the second(third)-order molecular nonlinearities are called the molecular hyperpolarizabilities $\beta(\gamma)$. Their determination is of fundamental interest for the development of optimized nonlinear optical materials. The hyperpolarizabilities of newly synthesized molecules can be determined with several different experimental methods. The methods to determine the second-order nonlinearities are Electric Field Induced

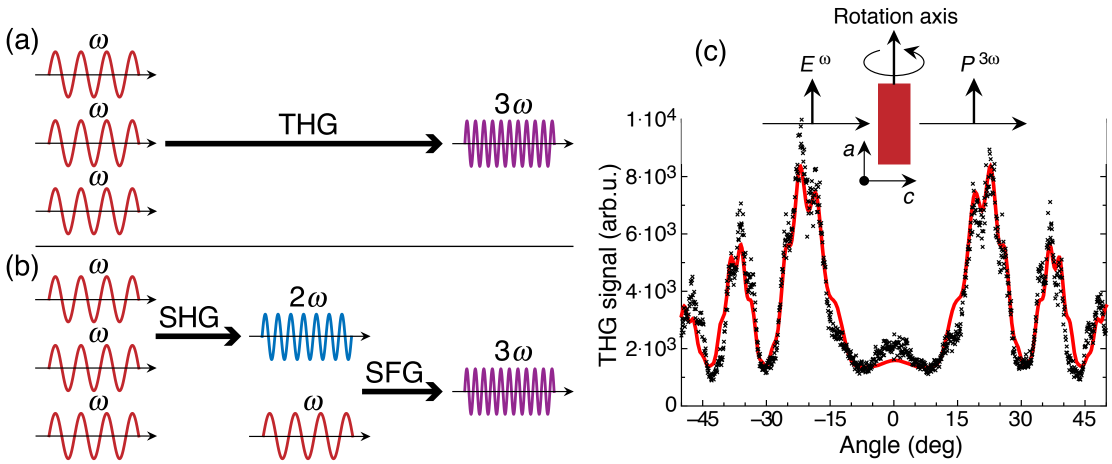


Figure 3.17: (a) Direct third-harmonic generation (THG) and (b) cascaded second-harmonic generation (SHG) and sum-frequency generation (SFG) contributions to the third-harmonic signal in polar crystals. (c) Third-harmonic Maker-fringe curve of a DAST crystal measured at 2100 nm fundamental wavelength for light polarizations along a, where cascaded second-order contributions appear [Bosshard et al., 2000].

Second Harmonic Generation (EFISH) and Hyper-Rayleigh Scattering (HRS). EFISH is the standard method for the determination of molecular hyperpolarizabilities β. With this method, the dissolved nonlinear optical chromophores are oriented by applying a static electric field. This allows the coherent generation of frequency doubled light, whose intensity is measured. With HRS, the scattered light from a solution of the material under investigation is observed. Due to orientational fluctuations, part of the scattered light is frequency doubled. The intensity of this incoherent second-harmonic generation is measured and compared with the intensity obtained from a reference solution. We applied both methods for the investigation of a series of newly synthesized hydrazone derivatives, zwitterionic chromophores, and organic salts; some results are shown in Fig. 3.18. Both techniques gave the same experimental results and revealed the large potential of these materials for nonlinear optical and electro-optic applications.

For the determination of the third-order nonlinearities γ, third-harmonic generation (THG) in solution is most often used. The experimental set-up is quite similar to EFISH except that (i) no external electric field has to be applied and that (ii) the experiments have to be carried out in vacuum in order to get rid of the the contributions to the third-harmonic signal of air. With these measurements, we have shown the importance of the substitution pattern and the functionality (electron donor/acceptor) of the attached end groups for the

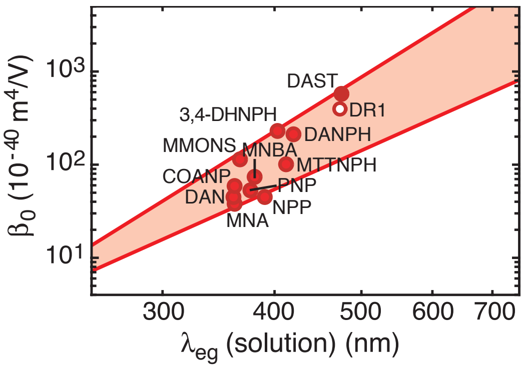


Figure 3.18: First-order hyperpolarizability β_0 extrapolated to infinite wavelengths versus wavelength of maximum absorption for various molecules. The shaded area represents the range of values experimentally determined in various laboratories. Acentric crystals are available for all chromophores included except DR1 (Disperse Red 1).

molecular hyperpolarizability γ.

Powder test. Powder test is a preliminary method used for newly developed organic crystals to determine their potential for nonlinear optical applications. In this method, the second(or third)-harmonic generation efficiency of crystalline powder is measured, usually in comparison to a powder with known nonlinear optical properties. Figure 3.19 shows the second-harmonic powder test efficiency in reflection for several crystalline powders measured in our laboratory using 1.9-μm fundamental light. Also shown are first-principle calculations based on density functional theory to determine the hyperpolarizability β, the oriented-gas model to determine the effective hyperpolarizability in the crystalline state, and averaging over all possible orientations of the powders. A reasonable agreement is obtained, although several effects have been neglected in the calculations, including intermolecular interactions and possible phase matching.

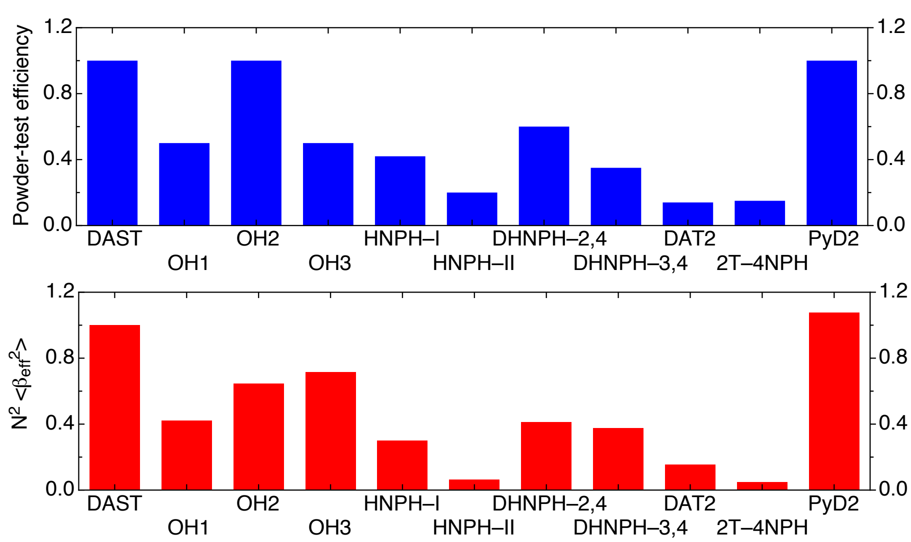


Figure 3.19: Measured powder second-harmonic generation efficiency at 1.9 μm fundamental wavelength (top) and calculated $N^2\langle\beta_{\text{eff}}^2\rangle$ (bottom), where N is the chromophore number density and $\langle\beta_{\text{eff}}^2\rangle$ are squared effective first-hyperpolarizability components (calculated by density functional theory), averaged over all possible orientations. All values are normalized to DAST.

Maker fringe method. The nonlinear optical coefficients of organic materials are most often measured using the Maker-fringe technique, where a plane parallel sample is rotated around an axis perpendicular to the incoming laser beam. The intensity of the second(third)-harmonic wave $I^{2\omega}(I^{3\omega})$ generated in the sample shows oscillations due to different phase velocities of the fundamental and frequency-doubled (tripled) beams in the material. The envelope of such curves is evaluated in comparison to the Maker-fringes of a reference crystal, quartz in most cases, whose nonlinear optical coefficients are known. An example of the Maker-fringe measurement results is shown in Fig. 3.20. With this method, we determined nonlinear optical coefficients of several organic crystals listed in Table 3.1.

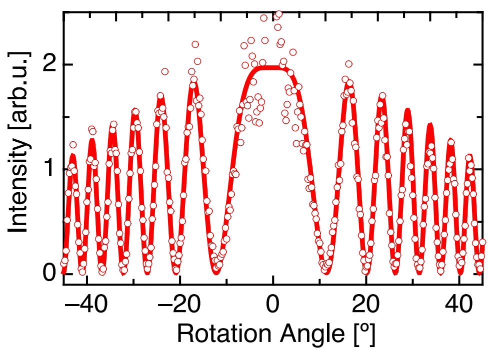


Figure 3.20: Maker-fringe measurement (open circles) and theoretical curve (solid line) used to determine the second-order nonlinear optical coefficients of DSTMS.

Z-scan. With the Z-scan technique, the nonlinear refractive index n_2 can be determined. A single focused (Gaussian) laser beam and a circular aperture placed in the far field behind the sample are used. The sample is translated along the beam axis through the focus and the intensity after the aperture detected. Because of the translation, the incident intensity on a sample is changing, which leads to a variable refractive index change induced by n_2. By analyzing the transmission curve, the sign as well as the magnitude of n_2 can be determined; an example is shown in Fig. 3.21. With this technique, we determined the nonlinear refractive indices and nonlinear absorption coefficients of several third-order nonlinear optical materials, as well as of second-order materials, enabling to quantify cascading second-order contributions to the self-focusing effect in these materials.

Four-wave mixing. In the degenerate four wave mixing (DFWM) experiment, the third-order susceptibility $\chi^{(3)}(-\omega, \omega, -\omega, \omega)$ with degenerate frequencies can be determined. This nonlinear susceptibility is directly proportional to

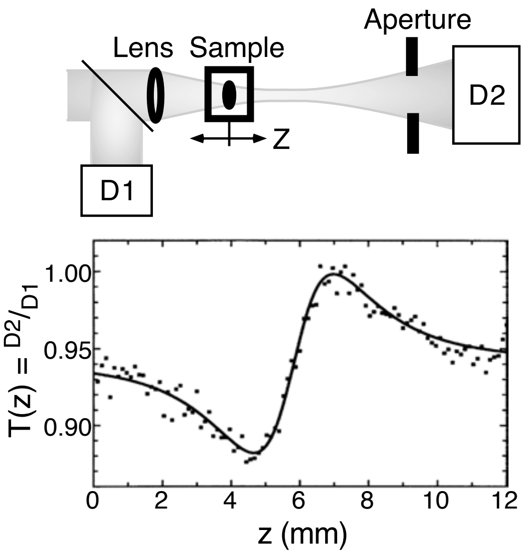


Figure 3.21: Schematic illustration of a Z-scan experiment and experimental Z-scan curve using a $KNbO_3$ crystal, which was used to identify different cascading contributions to the nonlinear refractive index n_2 in this material [Bosshard et al., 1995].

the nonlinear refractive index n_2, which is used to describe optically induced refractive index changes. An advantage of this technique is the possibility to record the temporal shape of the third-order nonlinear optical signal. We have shown that in polar materials, for which cascading contributions may be dominant, the values of $\chi^{(3)}(-\omega, \omega, -\omega, \omega)$ may be different compared to the Z-scan technique, due to the influence of different propagation directions of the interacting beams [Zgonik et al., 1996].

3.5 Applications

Due to the low dispersion of the dielectric constant and the electro-optic coefficient in second-order organic nonlinear optical materials from the DC to the optical frequency range, these materials are very promising for high-speed electro-optic applications and phase-matched THz wave generation. Their nonlinear optical figures of merit at optical frequencies may be several orders of magnitude higher as compared to the best inorganic materials, which is why they are also attractive for optical frequency-conversion applications.

Waveguide electro-optic modulators. Electro-optic modulation is particularly attractive in waveguiding geometries that allow for reducing the required driving voltages by several orders of magnitude compared to bulk materials.

A. Polymer modulators. Since polymers are easily processed in thin films, they are very suitable for integrated optical elements. The relatively stable and efficient side-chain polyimide A-095.11 was used to fabricate waveguide electro-optic phase and Mach-Zehnder modulators by using the photobleaching technique. Amplitude electro-optic modulation with a driving voltage of 50 V and an extinction ratio of 13 dB at 1313 nm has been demonstrated. Effective poling of multilayer structures and optical losses were identified to be the main issues to achieve an improved performance of poled-polymer based devices.

B. Organic single-crystalline modulators. Because of the advantages of crystals compared to polymers, we were also interested in producing optical waveguides in organic nonlinear optical crystals. We developed and investigated several techniques for fabricating planar optical waveguides and sub-10 micrometer thin films, as well as techniques for lateral structuring to produce channel waveguides.

We produced planar nonlinear optical single-crystalline waveguides using the following techniques:

- Thin-film waveguide formation by using the Langmuir-Blodgett method (see Fig. 3.10 and 3.11).
- Growth of free-standing single-crystalline thin films by vapor growth (see Fig. 3.1).
- Solution growth of thin films suitable for waveguiding directly on the substrates. Particularly attractive are the capillary growth method between two amorphous glass substrates [Kwon et al., 2008] and direct solution growth by evaporation-induced local supersaturation by surface interactions (ELSSI) [Kwon et al., 2009]. Thin films of OH1 could be grown on different amorphous substrates (glass, glass with gold electrodes, silicon on insulator) with a thickness between 0.1 and 4 μm and very large single crystalline domains of up to several cm^2.
- Proton ion implantation was used and investigated for the organic crystal DAST to create planar optical waveguides and planar electro-optic phase modulators [Mutter et al., 2007]. This method allows for the precise waveguide thickness control and waveguiding with relatively low losses of 7 dB/cm at 1.55 μm. Phase modulation with a high electro-optic fig-

ure of merit $n^3 r \simeq 400$ pm/V at 1.55 μm has been demonstrated in such waveguides.

For channel waveguide structuring in organic nonlinear optical crystals, the following techniques were developed:

- Melt growth of crystal-cored organic fibers, suitable for materials that are stable above the melting point [Kerkoc et al., 1989].
- Modified photolithography and reactive ion etching, which was used for OH1 thin films to produce electro-optic phase modulators, see Fig. 3.22.

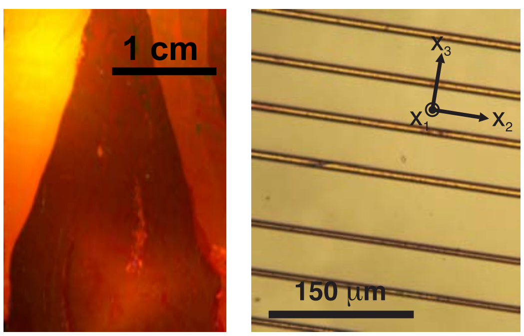


Figure 3.22: Left: Photograph of a large-area OH1 thin film (thickness of ~ 3 μm) on an amorphous glass substrate placed between two crossed polarizers. Regions of the same color indicate single-crystalline domain areas. Right: Microscope image of single crystalline OH1 wire waveguides fabricated in these films [Hunziker et al., 2008].

- Photo-structuring by photochemical refractive-index modification [Mutter et al., 2003] and by femtosecond laser ablation [Dittrich et al., 2003].
- Direct electron-beam structuring is a particularly interesting method for organic salts like DAST, for which photolithography or melt growth are not suitable. By using this method, vertical and horizontal light confinement is achieved in a single processing step. This is a very flexible technique that allowed to fabricate first Mach-Zehnder modulators in DAST crystals [Mutter et al., 2007]. Examples of the fabricated structures are shown in Fig. 3.23.

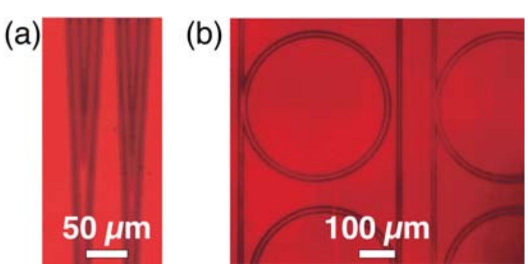


Figure 3.23: Photographs of e-beam irradiated DAST samples in a transmission microscope: (a) a detail of a Mach-Zehnder and (b) microresonator structure.

- Optical waveguides were also grown from the melt between two anodically bonded borosilicate glass wafers, which were structured and equipped with electrodes prior to bonding. The same method was used to fabricate single-crystalline nanostructures inside large-area devices with crystal thicknesses below 30 nm and lengths of above 7 mm, which are shown in Fig. 3.24.

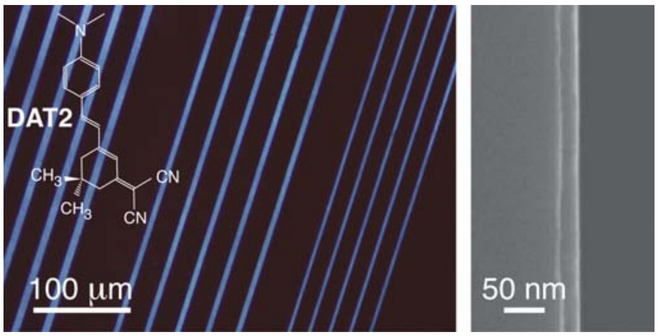


Figure 3.24: Left: Transmission microscope image of approximately 25 nm thick DAT2 crystalline nanowires grown from the melt inside pre-fabricated grooves. Right: SEM image of the corresponding end-facet showing a 25 nm thick electro-optic single crystal [Figi et al., 2008].

Microring-resonator filters and modulators. Mach-Zehnder integrated-optic elements are CMOS compatible and allow for high speed (> 10 GHz) modulation in a traveling-wave configuration. However, present devices are still having a rather large size (lengths in the order of 1 cm) and a relatively high power consumption (several watts). Compact-size and low-power devices are needed for future applications. A promising structure is a microring resonator, in which long electrical and optical interaction lengths are possible on a very small scale (in the order of 10–100 μm), allowing for lower power consumption and very large scale integration (VLSI). An example of a polymer microresonator and performance is shown in Fig. 3.25.

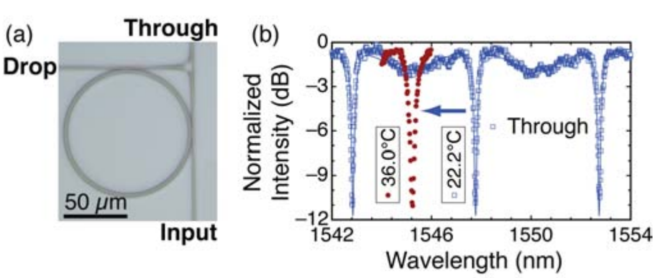


Figure 3.25: Filtering characteristics (through port transmission as a function of the input wavelength) of a polymer add-drop microresonator of 50 μm radius with a high finesse (~ 17) and high extinction ratio of 12 dB. The microring and port waveguides are structured on an oxidized Si substrate in a hybrid polymer Ormocore. Efficient switching of the resonator wavelength by the thermo-optic effect is also shown [Rezzonico et al., 2006].

Microring resonators made of polymers are interesting because of the simple technological prerequisites needed for the fabrication of the devices. Nevertheless, there are still some restrictions in terms of size reduction and dimensions of functional elements made of polymers. Among the major issues associated with electro-optic polymeric ring-like devices, the usually small refractive index contrast between core and cladding makes bending losses non-negligible for curvature radii smaller than 100 μm. Also physicochemical compatibilities, technological features necessary for precise structuring, and successful, stable poling of the active compound in microstructures are generally challenging. To solve these problems, we introduced, designed, modeled, and produced electro-optic polymeric microring modulators coupled to the bus waveguide by means of a so-called Charon coupler (see Fig. 3.26). We showed that this approach offers several advantages over conventional ring resonators, including small polymer-device dimensions (< 50 μm ring radius), enhancement of the coupling acceptance, and increased modulation bandwidth maintaining the large electro-optic sensitivity typical for high-Q resonators.

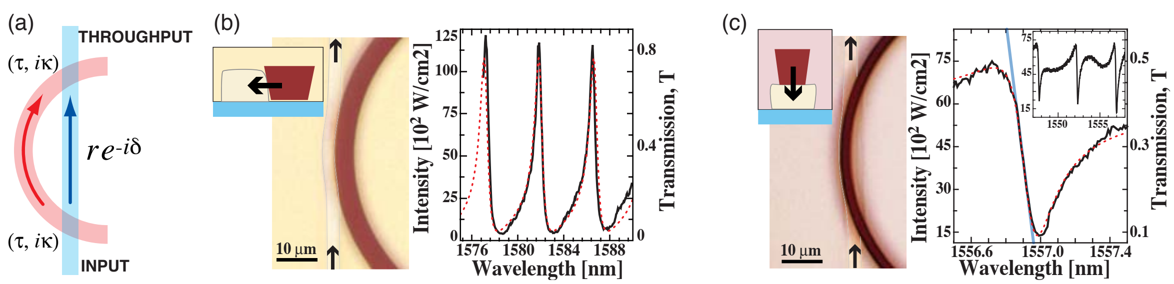
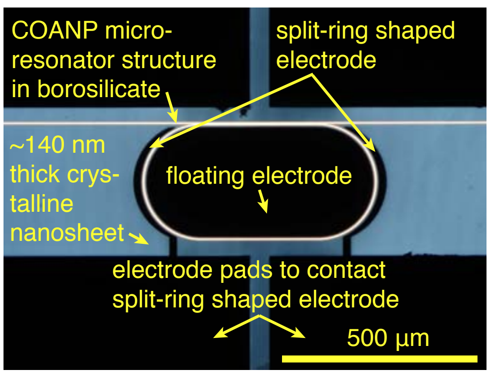


Figure 3.26: a) A simplified scheme of a Charon coupler, (b) laterally and (c) vertically coupled 50 μm-radius electro-optic Charon microresonators produced by micro-embossing technique. (b) The lateral displacement of the bus waveguide is induced by a horizontal shift of the molding stamp. The measured TE transmitted intensity shows high-extinction (11 dB) peak-like resonances. (c) A deformation of the bus waveguide is produced by slightly excessive pressure of the mold. The measured TE transmitted intensity shows strongly asymmetric Fano resonances with approximately linear steepest resonance edge (blue solid line, $|\mathrm{d}T/\mathrm{d}\lambda|_{\max} = 3.4\,\mathrm{nm}^{-1}$). [Rezzonico et al., 2008].

Because of their high orientational and photochemical stability compared to poled polymers, organic crystals are also very attractive for microring-resonator applications. In addition, due to their relatively high refractive index (2.0–2.3 compared to 1.6–1.7 in poled polymers), high index contrast to the cladding material and therefore smaller structures with lower losses are feasible. By melt capillary method, we fabricated first organic single-crystalline microring resonators and demonstrated electro-optic modulation and tuning therein (see Fig. 3.27).



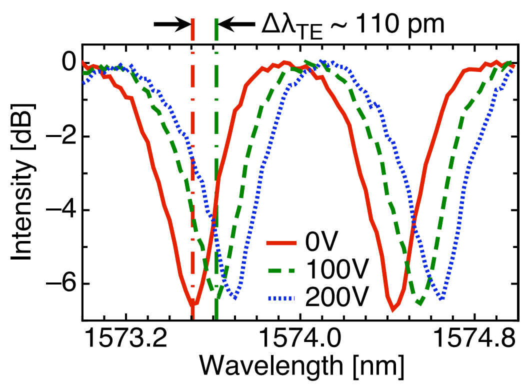


Figure 3.27: *Left:* Top-view microscope image of a racetrack microring resonator in an organic electro-optic crystal COANP between crossed polarizers, showing a single-crystalline COANP microring, port waveguide, and very thin crystalline layer (bright areas) and electrodes (black areas). *Right:* Spectrum and its tuning by using the electro-optic effect. The shift corresponds to a tunability of about 0.13 GHz/V [Figi et al., 2009].

Chapter 4

Science and Technology of Pulsed Terahertz Radiation

Until few years ago, electromagnetic radiation with frequencies near one terahertz had not attracted a lot of attention. Through recent progress in materials and laser research, very short pulses of terahertz radiation became feasible, enabling a variety of novel applications. The Nonlinear Optics Laboratory has contributed to this progress in two aspects. First is the development of novel nonlinear organic crystals for the generation and detection of terahertz pulses, second are innovations in their coherent detection.

4.1 THz Waves

Humans have always used light to collect information about their environment. With our eyes, we can inspect objects from a distance very quickly and without touching them. Where we are limited by our physiology – visible light contains only a small fraction of the electromagnetic spectrum – we have developed technical utilities such as X-ray or infrared cameras to complement our visual sense.

The terahertz spectral range comprising frequencies between 10^{11} and 10^{13} Hertz (Fig. 4.1) offers intriguing possiblities that may even go beyond those of X-rays. They allow new kinds of insights because the contrasts between transparent and opaque matter are different compared to other spectral ranges. In contrast to X-rays, terahertz radiation is not harmful towards our health because it is not ionizing, and the intensities used are usually very small.

Terahertz radiation had long been very hard to employ in applications since it is

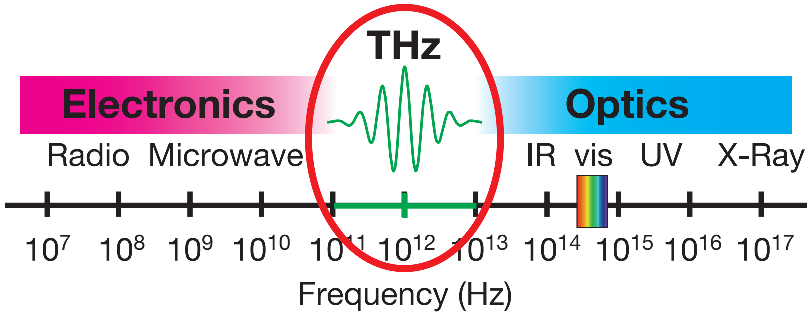


Figure 4.1: THz range of the electromagnetic spectrum

comprised in the blackbody radiation at room temperature. This thermal background often masks the actual signal of continuous terahertz sources. One may overcome this problem by using few-cycle terahertz pulses and detecting them in a coherent manner, e.g. by electro-optic sampling. The main benefit of this technique is that the time-dependent waveform of the pulse may be recorded such that one obtains full amplitude and phase information from a single measurement. This is the key to new applications such as spectroscopic imaging that has a very high potential for security purposes or material inspection.

4.2 Optical Rectification

Few-cycle terahertz pulses are routinely generated through the second-order nonlinear effect of optical rectification (OR). The theory of OR for terahertz generation is similar to that of other well-known nonlinear conversion processes such as second harmonic generation (SHG). Besides the nonlinear optical susceptibility $\chi^{(2)}$, the most relevant parameter for efficient SHG is the coherence length, describing how well the phases of the interacting waves are matched.
Unlike in SHG, the coherence length alone is not sufficient to describe the conversion efficiency of terahertz generation. Due to vibrational excitations of the crystal lattice (phonons), there is usually non-negligible absorption at terahertz frequencies. A second difficulty is the inherent broadband nature of the pulses; dispersion makes it impossible to define a single coherence length for the whole pulse.
Through a theoretical analysis of the THz generation process, we have identified a quantity L_{max} that can be calculated analytically for any given material from experimental data (refractive indices and absorption coefficients). From

a graphical representation of L_{max} as a function of optical and terahertz frequencies, one may easily find the optimum experimental parameters for efficient terahertz generation. One example may be found in Figure 4.2.

4.3 Organic Materials

In the search of materials for efficient OR, we focused on organic crystals for two reasons. On the one hand, the nonlinear optical susceptibilities d of organics often exceeds those of inorganics. On the other hand, their refractive indices at terahertz frequencies are close those of optical waves, leading to large values of the key quantity L_{max}.

The main focus of our research lay on the ionic crystal DAST (4-N,N-dimethylamino-4'-N'-methylstilbazolium tosylate) which has one of the highest known values of d. The calculation of L_{max} revealed that efficient generation of terahertz pulses in DAST is feasible in several configurations. One of these covers frequencies between 1.5 and 4 THz that are well phase-matched with infrared pump pulses at wavelengths between 1300 and 1600 nm (see Figure 4.2). From a technological point of view, this is highly relevant since such lasers are commercially available, namely compact and reliable fiber lasers originally developed for the telecommunications industry. Other laser wavelength ranges where phase-matching is possible employ lasers at 1040-1070 nm or 700-750 nm, respectively.

The only drawback in the use of DAST as a source for terahertz pulses is a phonon resonance at 1.1 THz that is clearly seen as a "valley" in the values of L_{max} in Figure 4.2. In an actual experiment, this leads to a reduction of the emitted spectrum at this frequency which is undesirable for some applications. In order to overcome this problem, we have modified the chemical structure of DAST with the aim of reducing the negative effect of the phonon resonance without comprising its excellent nonlinear optical properties.

We have achieved this goal by developing DSTMS (4-N,N-dimethlyamino-4'-N'-methylstilbazolium 2,4,6-trimethylbenzenesulfonate), a crystal with a similar composition and structure as DAST. In fact, the nonlinear susceptiblities and phase-matching conditions are nearly identical; however, the phonon oscillation strength is reduced, and the "valley" in L_{max} is much less pronounced than in DAST (see Figure 4.2). Hence it is expected that DSTMS will soon replace DAST for terahertz applications.

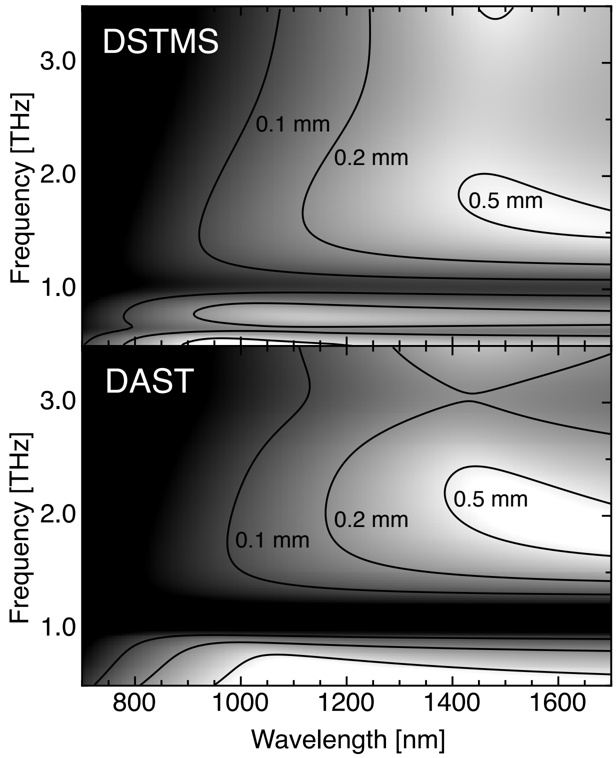


Figure 4.2: L_{max} of DAST and DSTMS

4.4 Coherent Detection of Terahertz Pulses

The standard way to the time-resolved detection of few-cycle terahertz pulses is electro-optic sampling. Through the linear electro-optic effect, the terahertz electric field induces a distortion of the indicatrix, i.e. a change of the refractive indices along the dielectric axes. Under properly chosen experimental conditions, this change effects the polarisation state of a copropagating sub-picosecond optical probe laser pulse which may then be read out by classical optical methods. Unfortunately, this principle is not applicable to birefringent electro-optic materials since such short laser pulses do not have a defined polarisation state after the crystal, unless it is parallel to a dielectric axis. In this case however, the terahertz electric field does not alter this polarisation state and thus cannot be measured. This is a major drawback of electro-optic sampling since the electro-optic coefficient of cubic, i.e. non-birefringent crystals is much lower than that of the birefringent ones.

We have developed an alternative method for the coherent detection of terahertz pulses that also employs the linear electro-optic effect but that has no birefringence restriction. It uses the spatial distribution of the refractive index change that reflects the spatial profile of the terahertz field amplitude. Such an index distribution has the same effect on the probe pulse as a lens, namely an intensity change in the far-field which can again be read out by classical optical methods (terahertz-induced lensing, Fig. 4.3).

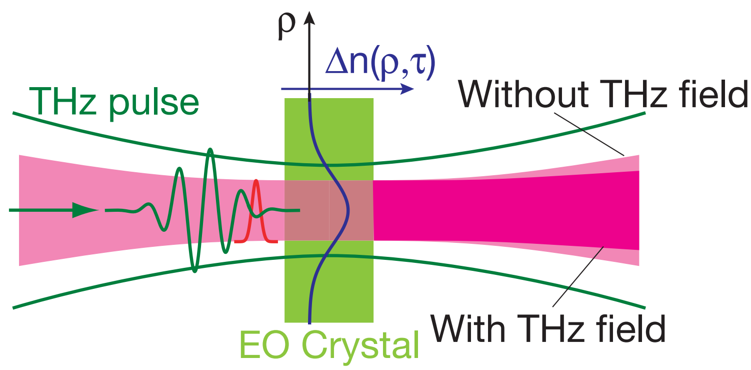


Figure 4.3: Principle of terahertz-induced lensing.

It is important to note that the same quantity L_{max} introduced above is also the key figure for the coherent terahertz detection in electro-optic crystals. A terahertz system combining two DAST or DSTMS crystals – one for the generation and one for the detection – and femtosecond fiber lasers thus promises a performance that is unreachable by any other method. In fact, we have demonstrated a photon conversion efficiency of about 10% with laser pulse energies below 30 μJ. In Figure 4.4, we present such a measurement; the peak probe pulse modulation in terahertz-induced lensing ranges to an unprecedented value of 140%.

4.5 Applications of Terahertz Pulses

4.5.1 Terahertz Time-Domain Spectroscopy

The most straightforward application of coherently measured terahertz pulses is time-domain spectroscopy. The transient terahertz electric field is measured twice: Once after transmission through a sample and once without a sample (see Figure 4.5). The sample causes a temporal shift of the pulse which is equivalent to a phase shift in the Fourier spectrum. From this phase shift, the refractive

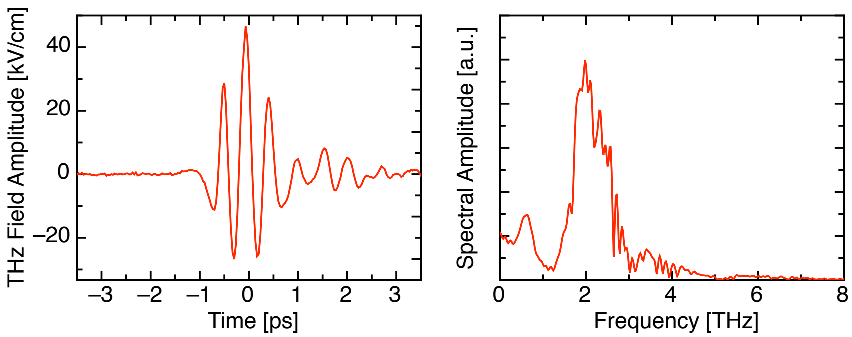


Figure 4.4: Time-domain waveform of a terahertz transient generated and detected in DAST crystals at a laser wavelength of 1500 nm, and its Fourier spectrum.

index may easily be calculated over the whole bandwidth. Similarly, the frequency dependent absorption coefficient may be obtained from the attenuation of the spectral amplitude. Hence terahertz time-domain spectroscopy enables the determination of the linear optical properties of a material in a single measurement over a wide frequency range without the need of a tunable source nor of a monochromator.

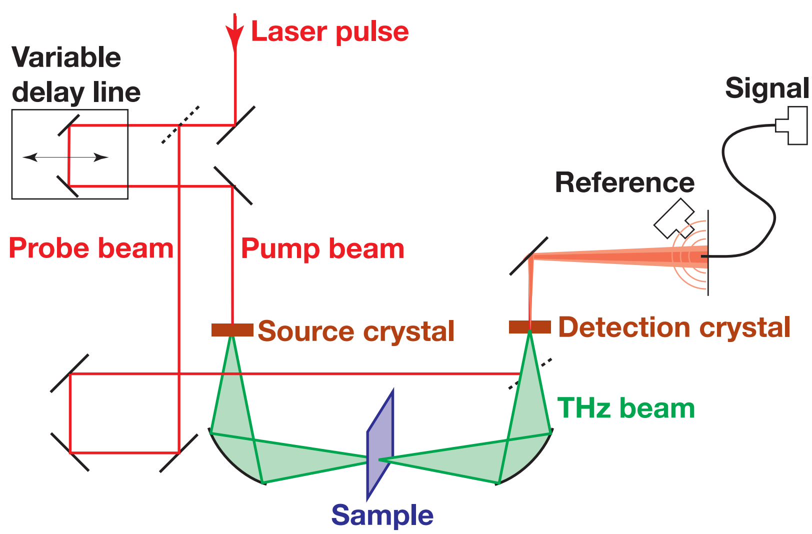


Figure 4.5: Experimental setup for terahertz time-domain spectroscopy

We have applied this technique mainly to organic nonlinear optical crystals in order to find the parameters under which they are ideally used for the generation of terahertz pulses.

4.5.2 Parametric or Spectroscopic Imaging

Most imaging techniques are based on differences in the amplitude that is reflected from, scattered by, or transmitted through the sample under investigation. In parametric terahertz imaging, also the time-retardation is measured that contains information on the sample thickness. Like this, contrasts become visible that are not perceivable with other methods (see Figure 4.6).

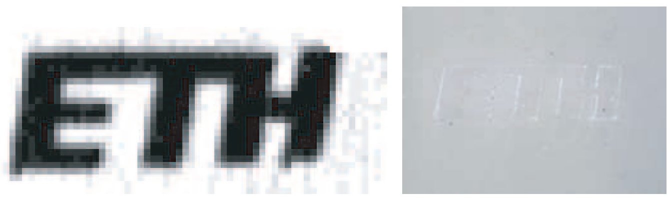


Figure 4.6: Phase resolved terahertz image of a transparent sample. The contrast is due to the different transit times of the terahertz pulse through one or two layers of overhead projector slides.

The full potential of imaging using terahertz pulses is exploited when one measures a complete spectrum for every pixel of the sample (spectroscopic imaging). Despite a rather long acquistion time, this technique enables to identify various kinds of substances even when they are sealed within or hidden behind packaging materials. This is especially useful when it comes to the search for e.g. explosives or drugs in the mail or in the luggage at airports.

Reference: A. Schneider, M. Neis, M. Stillhart, B. Ruiz, R. Khan, and P. Günter, J. Opt. Soc. Am. B **23**, 1822 (2006)

Chapter 5

Ferroelectrics and Atomic Force Microscopy

Ferroelectric materials possess a spontaneous polarization which can be reoriented by an external electric field. The direction of the spontaneous polarization can be used to store information. This is employed in nonvolatile random access memories (FRAM). Ferroelectrics also often exhibit large dielectric permittivities which can be used in thin film capacitors and dynamic random access memories (DRAM). Their piezoelectricity is used in actuators, microelectromechanical systems (MEMS) and pressure sensors, while their pyroelectricity is used for pyroelectric sensors. The electro-optic, nonlinear optic, and photorefractive effects are used for photonic applications such as electro-optic modulators and deflectors, optical frequency converters, and high density optical data storage, real time holography, and optical phase conjugation, respectively.

This chapter is focused on the scanning force microscopy (SFM) method for imaging and writing of domain patterns in ferroelectric crystals. In the NLO Laboratory, this method was studied in detail with the aim to develop high-density ferroelectric nonvolatile memories [Abplanalp et al., 2004]. Most of the studies were performed on triglycine sulfate (TGS) and barium titanate ($BaTiO_3$).

5.1 Piezoresponse Scanning Force Microscopy

The idea of piezoresponse scanning force microscopy (SFM) is to monitor the local electromechanical vibrations of the ferroelectric sample caused by an ac voltage applied between an electrode below the sample and the probe tip. This oscillation is sensed by the tip in the contact mode. By measuring cantilever oscillations (normal or lateral), the direction of the spontaneous polarization (normal or parallel to the surface) can be detected [Abplanalp et al., 1998]. Since the SFM tip is used as a top electrode, the electric field is strongly non-homogeneous. Numerical calculations showed, however, that only the top few nanometers contribute to the surface deformation. Thus, the inhomogeneous electric fields results in a measurement of a thin layer at the surface only. Fig. 5.1 shows the experimental setup used for piezoresponse SFM. The measurement principle is depicted in Fig. 5.2.

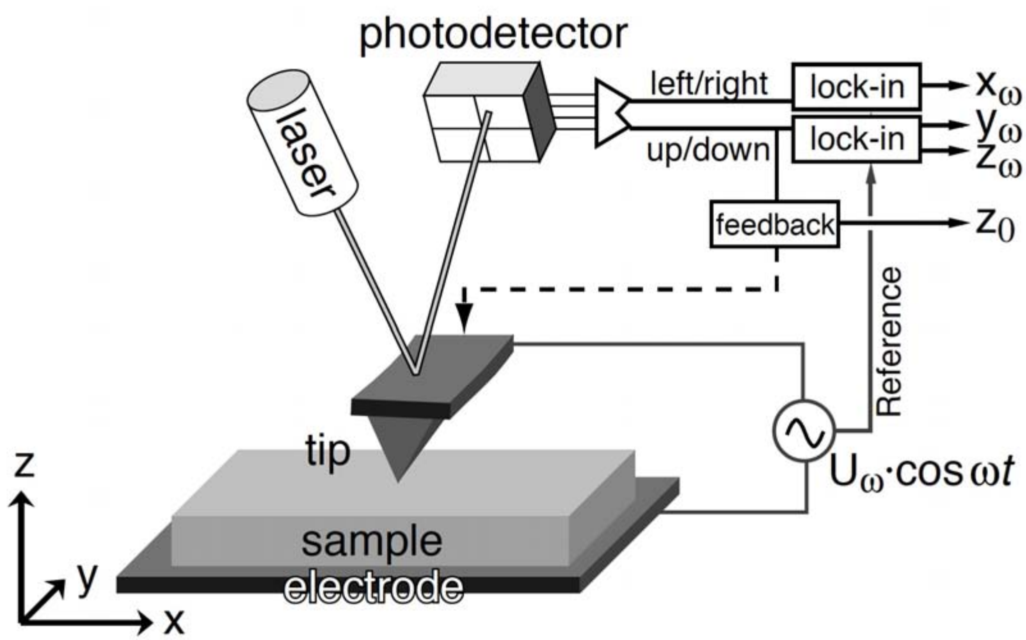


Figure 5.1: Setup used for the piezoresponse SFM. The cantilever deformation is measured by laser deflection onto a four quadrant detector. The up/down deflection is fed into a feedback adjusting the static force. The ac voltage applied between electrode and tip results - depending on the direction of the spontaneous polarization below the tip - in oscillations of the cantilever. These are detected by two lock-in amplifiers.

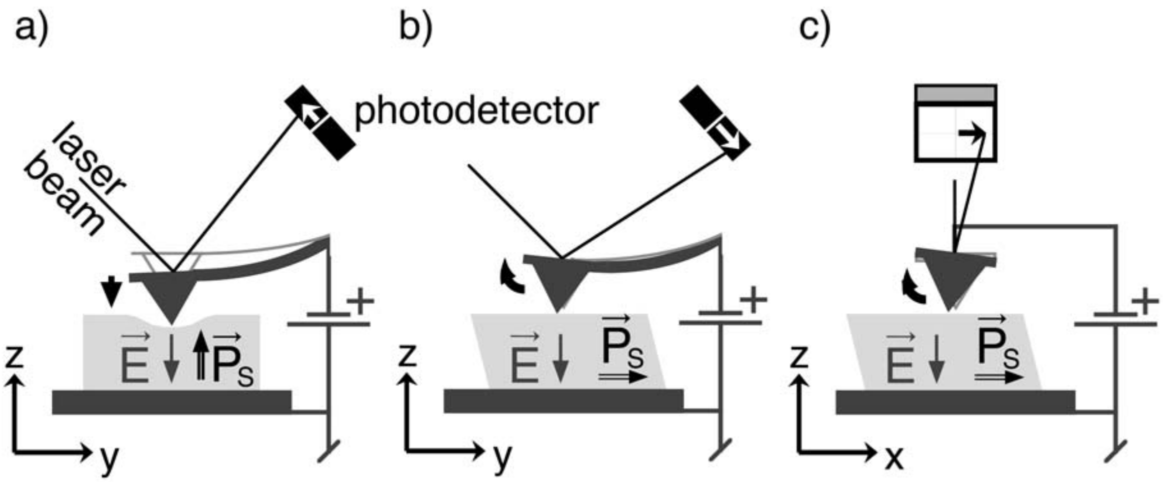


Figure 5.2: Principle of the SEM measuring technique based on the inverse piezoelectric effect. (a) The electric field created by the voltage applied to the tip is antiparallel to the spontaneous polarization leading to a local contraction of the sample. The tip follows this movement and the reflected laser spot on the photodetector moves. (b) and (c) For a spontaneous polarization parallel to the surface, a local lateral shear of the sample results (for clarity, the whole crystal is sheared in the figure). These movements can also be sensed by the deflected laser. (b) is the side view for P_S parallel to y, (c) is the front view for P_S parallel to x.

5.2 Poling of domains and their stability

A systematic study of domain poling was performed by applying voltage pulses of various durations. Fig. 5.3 shows an example where domains were written in a $BaTiO_3$ crystal. As can be seen, a voltage pulse produces a circular domain in $BaTiO_3$ when the tip is located above a larger c-domain. With increasing pulse duration, the domain grows bigger. The dependance of domain size on pulse voltage and duration is shown in Fig. 5.4. As expected, the domain size increases with longer pulses and larger pulse amplitudes. For a given amplitude, there exists a minimum duration needed to create a domain stable enough to be imaged. Similarly, for a fixed duration, a threshold voltage has to be reached.

In the theory discussed in [Abplanalp, 2001], a stable circular domain is predicted and in fact the calculated dependence of the domain diameter as a function of the applied voltage shows some similarities with the experimental results shown in Fig. 5.4. The main difference is that in the experiment the threshold field below which no domain is formed is much larger than in theory. The time dependance can only be discussed qualitatively. After switching the voltage on and off for a short time, the stable state should be the initial monodomain state. The stable domain size found experimentally can only be reached after some finite time

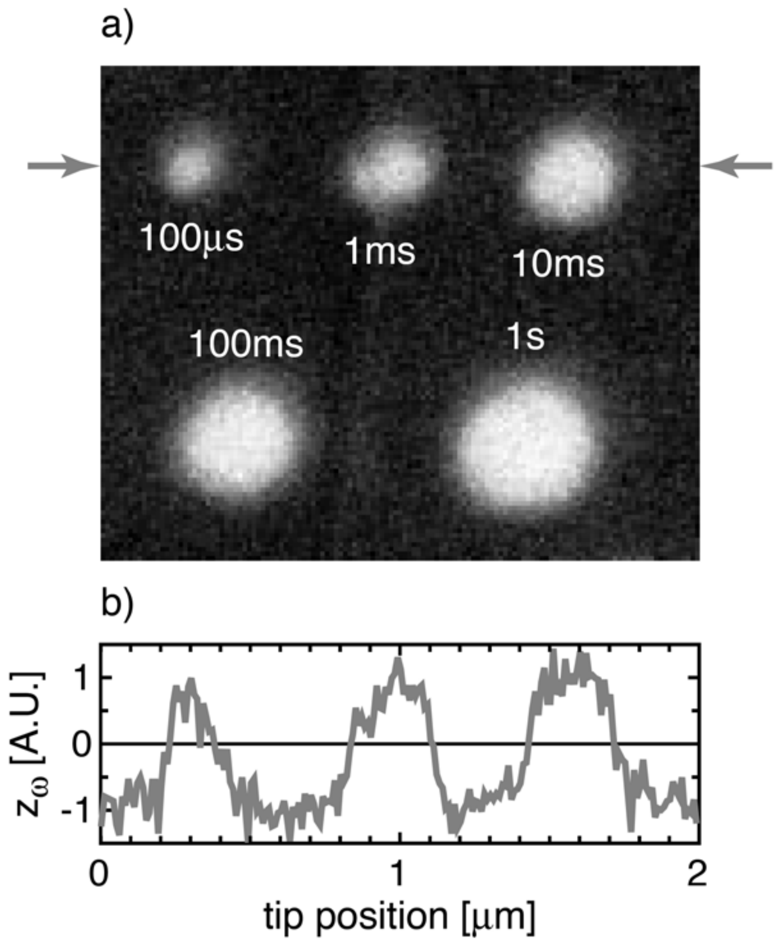


Figure 5.3: (a) Domains written into a $BaTiO_3$ crystal of 125 μm thickness at room temperature by positioning the tip at five positions and applying a voltage pulse of 30V for various durations. (b) Scan line at the position indicated by arrows in (a). Clearly the size of the domains is found to increase with duration.

if the depolarizing energy is compensated by a redistribution of charges while the field is still applied. Then any movement of the domain wall requires extra energy and the domain structure can remain in a metastable state after removing the field. The charge compensation takes some time, however, depending on the concentration of free charges, their mobility and on the dielectric constant, like in the photorefractive effect. Additionally, the surface conductivity also contributes to charge compensation. A relaxation time for screening may explain the minimum time required to observe a domain. For long times, the domain grows larger than the stable domain size found in theory.

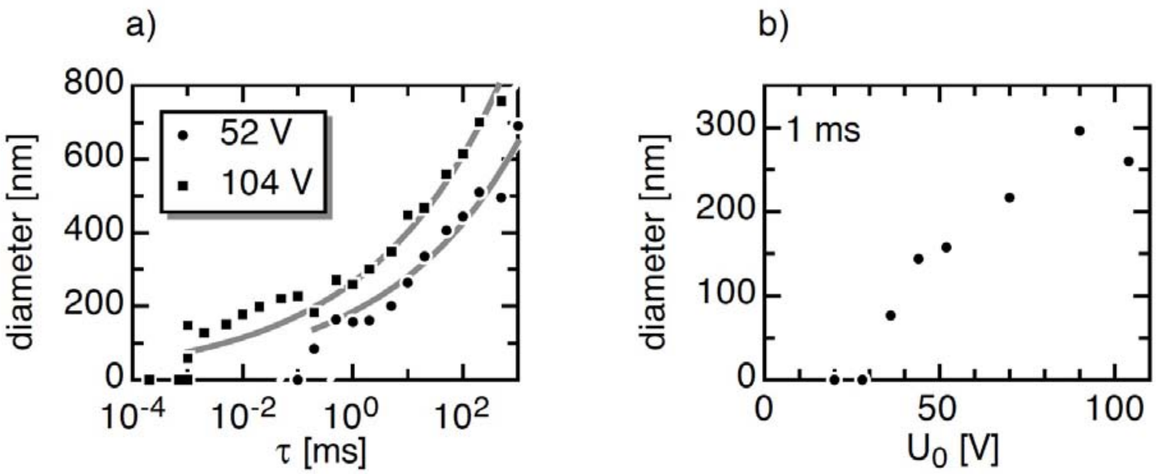


Figure 5.4: (a) Diameter of domains created in a 215 μm thick $BaTiO_3$ crystal by single voltage pulses of 52 V and 104 V, respectively, and various pulse durations. (b) Domain diameter for a fixed duration of 1 ms, but various amplitudes.

At the end, we mention only briefly our achievements in sub-micron precision of domain writing as can be used for information storage purposes. Fig. 5.5 shows an example, where in a previously uniformly poled area domains were nucleated at predefined places. The pattern can be interpreted as binary data as shown in the figure. The scan lines in Fig. 5.5b could be interpreted as the information "11000100111" and "10000100101". The values are clearly detectable at the separation of 360 nm. Note that nucleation of a domain was successful at any desired place within c-domains. The resulting domains are of equal size. Topographic steps which can be seen in Fig. 5.5a as faint lines do not influence the domain formation. Erasure of information is also possible. The tip can be positioned over a single domain which is then deleted by a voltage of opposite sign. Larger areas can be erased by scanning the tip while a voltage is applied.

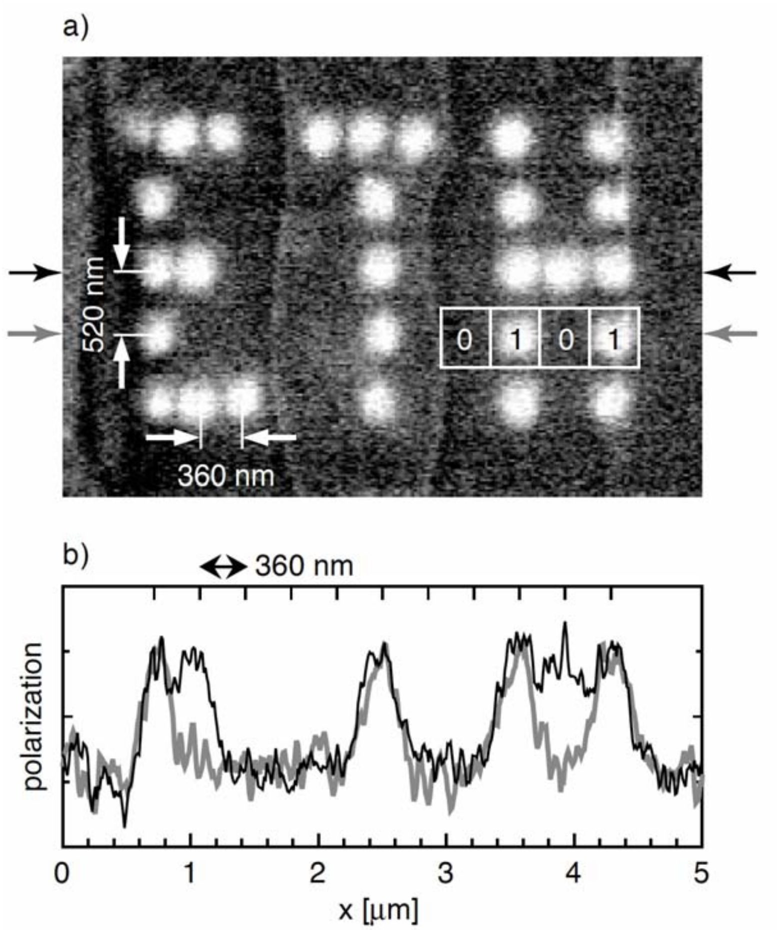


Figure 5.5: (a) Pattern of circular domains written into $BaTiO_3$ by positioning the tip at the desired positions and applying a voltage pulse. In such patterns, binary information can be stored as shown in the lower right part of the figure. (b) Two horizontal scan lines at positions indicated in (a). The difference in the stored information is clearly seen.

Chapter 6

Inorganic Nonlinear Optics and Microresonators

6.1 Potassium Niobate ($KNbO_3$)

$KNbO_3$ is an excellent nonlinear material for optical and electro-optic applications. Second-harmonic generation, sum frequency mixing, and optical parametric oscillation are important processes for converting available laser wavelengths into the blue and the near infrared spectral regions. In addition, $KNbO_3$ is a useful material for photorefractive applications. Because of its high second order nonlinear coefficients and favourable phase-matching properties, $KNbO_3$ is the material of choice for frequency doubling low power laser diodes operating in the near infrared.

The Nonlinear Optics Laboratory possesses a strong expertise in growing of high quality single-domain bulk $KNbO_3$ crystals. Apart from bulk crystal growth, different methods for growing $KNbO_3$ thin films [Schwyn Thöny, 1992] for applications in integrated optics were studied and developed such as radio frequency (rf) sputtering. For the fabrication of optical waveguides in bulk $KNbO_3$ crystals, another technique based on ion implantation and lithographic structuring was developed in our laboratory. This technique will be described later in Section 6.3.

Bulk $KNbO_3$ is grown by top-seeded pulling or by the Kyropoulos technique. The crystalline structure of $KNbO_3$ depends strongly on temperature. When cooling the crystal from melt temperature (T_{melt}=1050°C) to room temperature, it undergoes two structural phase transitions, namely cubic to tetragonal at 427°C and tetragonal to orthorhombic at 223°C. Furthermore, at -50°C a third transition from orthorhombic to rhombohedral can be observed. All these transitions result from the condensation of soft lattice vibrational modes in which almost rigid oxygen octahedra vibrate against potassium and niobium atoms. $KNbO_3$ is an optically biaxial crystal in the orthorhombic phase with the principal axes of the indicatrix being parallel to the orthorhombic coordinate system. The wavelength and temperature dependence of the refractive indices is illustrated in Fig. 6.1.

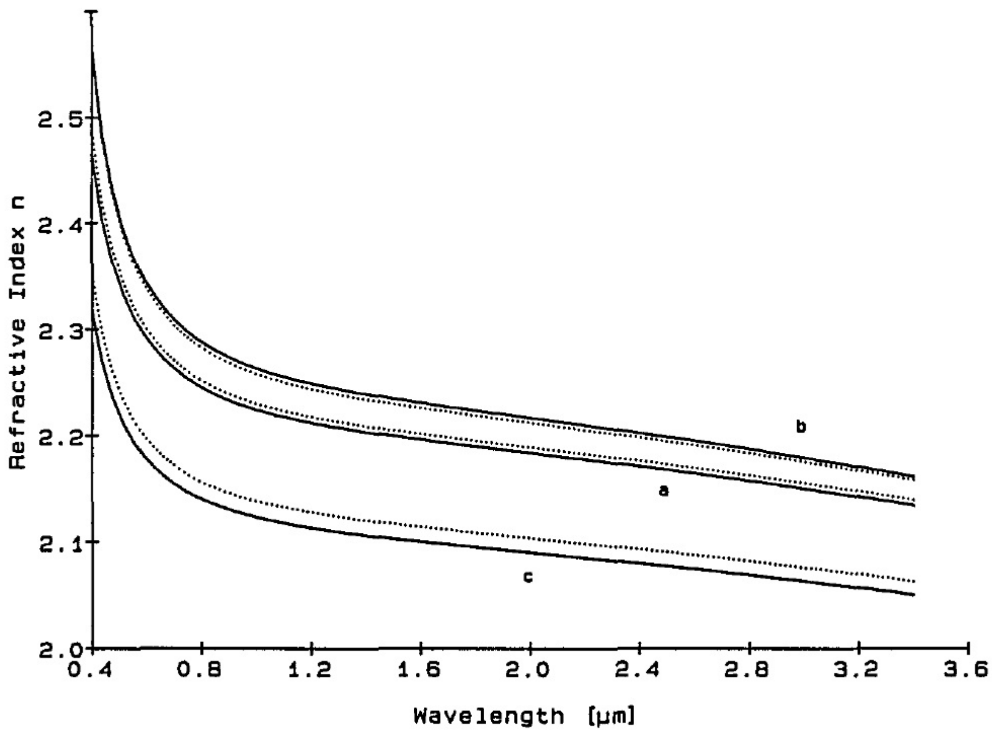


Figure 6.1: Overview of the refractive indices of $KNbO_3$ in the wavelength region 400-3400 nm. Solid curves: 22°C; dotted curves: 180°C [Zysset et al., 1992].

The tensor d_{ijk} of the nonlinear susceptibility has five independent coefficients according to the symmetry group mm2 and the Kleinmann symmetry rule. In the contracted notation it has the following form:

$$d_{ij} = \begin{pmatrix} 0 & 0 & 0 & 0 & -16.5 & 0 \\ 0 & 0 & 0 & 17.1 & 0 & 0 \\ -15.8 & -18.3 & -27.4 & 0 & 0 & 0 \end{pmatrix} pm/V$$

where the values are given for λ=1064 nm and T=22°C [Baumert, 1985]. These values are higher than those for $LiNbO_3$.

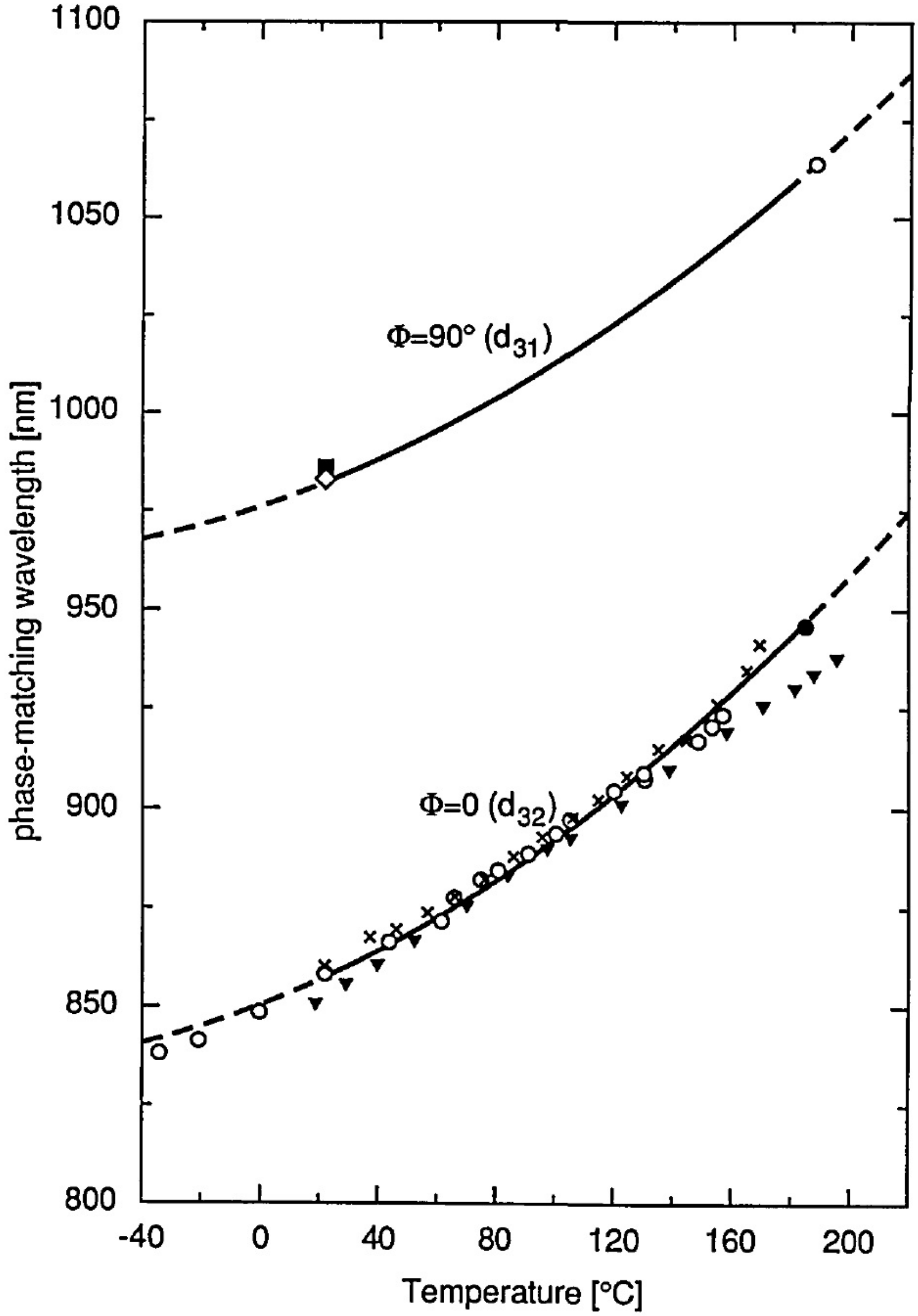


Figure 6.2: Temperature dependence of the wavelengths for noncritical 90° type I phase matching in $KNbO_3$ as calculated from the refractive-index data. Some measured data points are also shown [Biaggio et al., 1992].

$KNbO_3$ is well suited for temperature-tuned noncritical type I phase-matched second harmonic generation. Because of the relatively large temperature dependence of the refractive indices, the fundamental wavelength can be tuned over a large spectral range (see Fig. 6.2). Noncritical phase matching has the advantage of a larger angular acceptance bandwidth and a vanishing walk-off angle.
The first measurements of the linear electro-optic coefficients were accomplished by Günter in 1974. Table 6.1 shows the values which were remeasured later [Zgonik et al., 1993].

Table 6.1: Measured linear electro-optic tensor elements of $KNbO_3$ [Zgonik et al., 1993]. The values are given at a wavelength of 633 nm for a crystal at room temperature.

EO tensor element	unclamped [pm/V]	clamped [pm/V]
r_{113}	34±2	20.1±2
r_{223}	6±1	7.1±0.5
r_{333}	63.4±1	34.4±2
r_{131}	120±10	27.8±3
r_{232}	450±30	360±30

6.2 Potassium Sodium Tantalate Niobate (KNTN)

Electro-optic crystals of the potassium tantalate niobate ($KTa_{1-x}Nb_xO_3$ or KTN) family have attracted increased attention for integrated optics due to their large linear and quadratic electro-optic coefficients and the possibility of tuning the phase transition temperature by varying the Ta:Nb ratio as shown in Fig. 6.3. For integrated optics applications, epitaxially grown thin films promise to com-

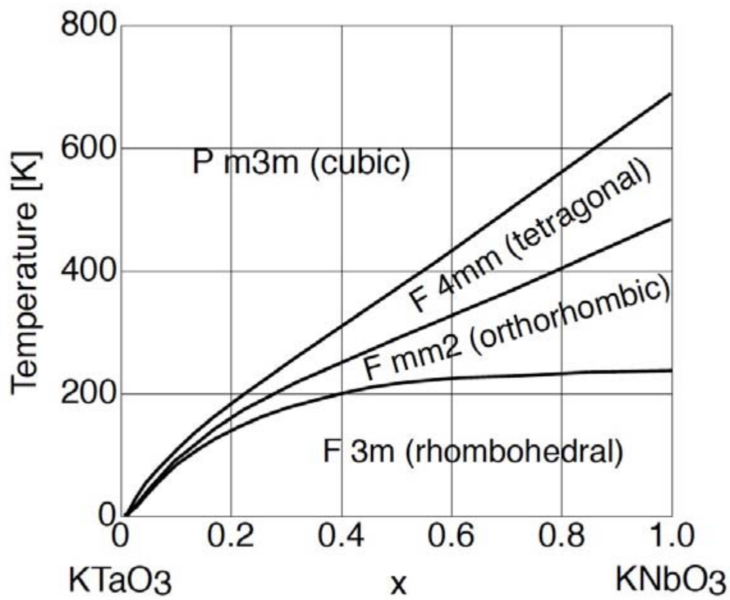


Figure 6.3: Phase diagram of the $KTa_{1-x}Nb_xO_3$ solid solution system. P and F indicate the para-electric and ferroelectric phases, respectively.

bine low optically losses with bulk-like dielectric and electro-optic properties as long as two requirements are met: good lattice matching at the substrate/film interface and an optically flat thin film surface. In our approach we used the liquid phase epitaxy technique to grow thin films of potassium sodium tantalate niobate ($K_{1-y}Na_yTa_{1-x}Nb_xO_3$ or KNTN) on potassium tantalate ($KTaO_3$ or KT) substrates. By substituting a small percentage of the potassium with sodium and performing the growth at a slow rate, a very small relative lattice mismatch of less then 10^{-4} and a surface roughness of approximately 20 nm rms were achieved [Herzog et al., 2007].

Slab and ridge waveguides have been fabricated and characterized from as-grown optically flat KNTN thin films. Polarization maintaining waveguiding has been demonstrated for both TE and TM modes. A film thickness of 3.4 μm and a refractive index contrast of 11×10^{-3} at a wavelength of 1550 nm have been

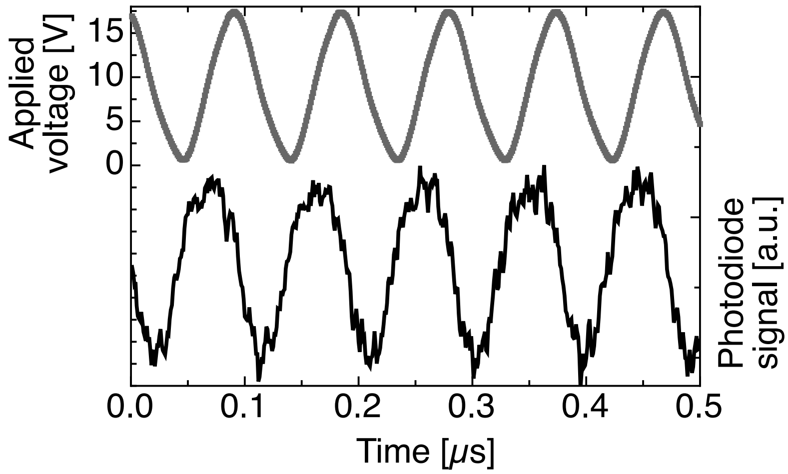


Figure 6.4: Amplitude modulation at $f = 10$ MHz and $\lambda = 633$ nm in an integrated $KNaTaNbO_3$ thin film phase modulator on a $KTaO_3$ substrate

determined using an optical profile reconstruction method. The dielectric and electro-optic coefficients of the thin film have been compared to the bulk values. Thin films show an electro-optic Kerr coefficient of $R_{11} = 8.2 \times 10^{-17}$ m^2/V^2. Finally, an electro-optic phase modulator was produced, having the half-wave voltage times length figure of merit of $V_\pi \times l = 38$ Vcm. Fig. 6.4 shows an amplitude modulation of light with $\lambda = 633$ nm at a frequency of 10 MHz.

6.3 Ion Implantation of Inorganic Crystals

Ion implantation is a widely used technique for production of planar optical waveguides in inorganic crystals. The Nonlinear Optics Laboratory has a long tradition in studying and employing this technique for the fabrication of optical waveguides in several nonlinear optical crystals. Initial implantation experiments were performed at ETH / PSI and in cooperation with the Institut für Schicht- und Ionentechnik in Jülich, Germany. Later on, the ion implantation experiments were continued at the ETH / PSI facilities. In 1991, the first optical waveguides in $KNbO_3$ were produced by implantation of 2 MeV He^+ ions [Strohkendl et al., 1991; Fluck et al., 1991]. These waveguides were of special interest for the blue light generation by the frequency doubling of red diode lasers. In order to produce ridge-type optical waveguides, the ion implantation technique was combined with lateral structuring by photolithography and Ar ion sputtering. This work led to the first demonstration of direct blue second harmonic light generation of laser diodes (see below). The same technique was successfully applied also for the fabrication of optical waveguides in other materials. These experiments were later extended to the ultraviolet (UV). Of special interest was β-BaB_2O_4 (beta barium borate, BBO) which is transparent in the UV wavelength range down to 190 nm. Apart from the deep UV light generation (see Section 6.5), also EO modulation was demonstrated in fabricated BBO waveguides at a wavelength of 257 nm [Degl'Innocenti et al., 2007]. Suitability of the ion implantation was studied also on other ferroelectric materials such as $Sn_2P_2S_6$ (SPS) [Guarino et al., 2006] and fluorescent crystals such as Cr:LiSAF [Majkic et al., 2007]. Fig. 6.5 shows a performance of a broadband fluorescence source based on a Cr:LiSAF waveguide pumped by a red diode laser.

In general, ion implantation with light ions such as He^+ leads to a buried optical barrier with a relatively small decrease of the material refractive index ($\Delta n < 0.1$). Consequently, such waveguides show high bending losses. On the other hand, a high dose He^+ implantation ($D \approx 5 \times 10^{16}$ ions/cm^2) can be used for the slicing of single-crystal thin films from bulk crystals. An ion-sliced film can be bonded onto a low index substrate. This method, which is particularly well suited for $LiNbO_3$, is described in Section 6.6.

Yet another more recently studied technique for the production of planar waveguides in $LiNbO_3$ is the implantation with medium-mass ions (O, F, Mg). Fluorine implantation with ion energies above 14 MeV at a significantly lower fluence ($D \approx 10^{14}$ ions/cm^2) yields waveguides with a step-like index profile and largely preserved nonlinear optical properties. In this case, the most important mechanism for the formation of the optical barrier is strong nucleus-electron interaction. The obtained wide optical barrier with a moderate index contrast provides a sufficient vertical confinement for bent waveguides with curvature radii as small as 40 μm. Fig. 6.6 shows a dependence of the barrier width and position as a function of fluorine ion fluence and energy. Using laser lithography patterning and Ar-ion sputtering, we have demonstrated ridge-type single-mode optical waveguides (see Figs. 6.7 and 6.8) and electro-optically tunable microring resonators in F-implanted $LiNbO_3$ wafers [Majkic et al., 2008].

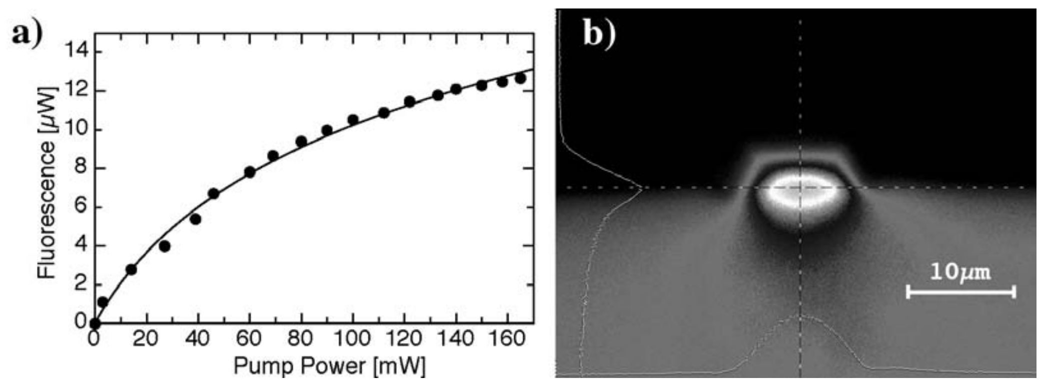


Figure 6.5: a) Emitted fluorescence power as a function of the input optical pump power. b) Fluorescence emission profile at the exit of a Cr:LiSAF waveguide.

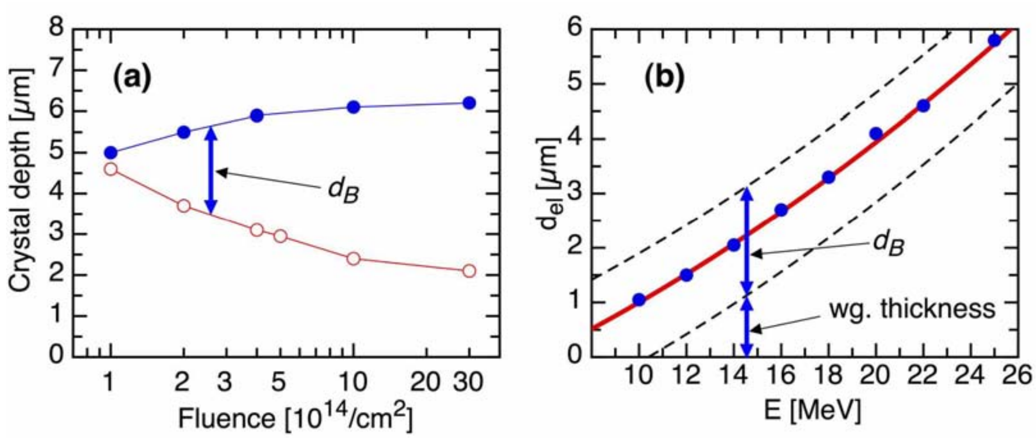


Figure 6.6: a) Depth position of the high-energy (open circles) and low-energy (closed circles) crystalline-amorphous boundaries as a function of implantation fluence. Barrier width d_B for fluence $\Phi = 2.5 \times 10^{14}$ ions/cm^2 is shown. b) Simulated depth position of the maximum electronic stopping power d_{el} in F-implanted $LiNbO_3$ as a function of ion energy. The barrier width d_B and the estimated waveguide thickness for $\Phi = 2.5 \times 10^{14}$ ions/cm^2 and E = 14.5 MeV are shown.

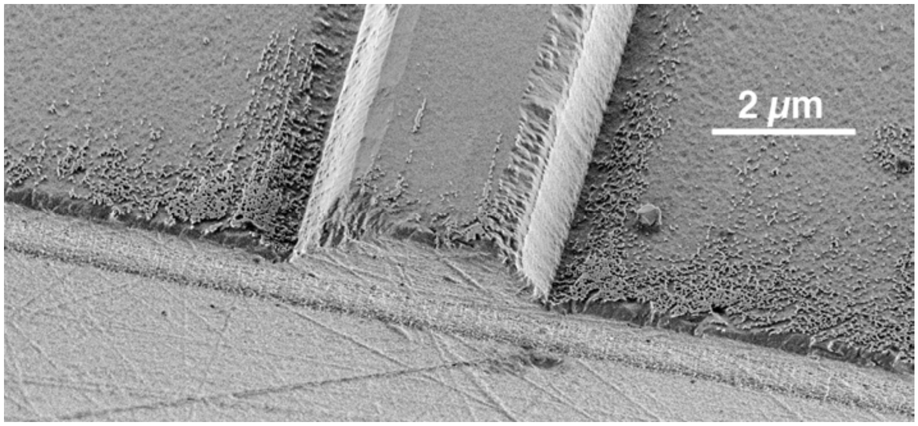


Figure 6.7: Scanning electron micrograph of a fabricated ridge waveguide on top of a F-implanted $LiNbO_3$ crystal. The waveguide cross-section is trapezoidal with a base width of 3.7 μm, a top width of 2.7 μm, and a ridge height of 1.2 μm. The amorphised optical barrier layer beneath the ridge is visible.

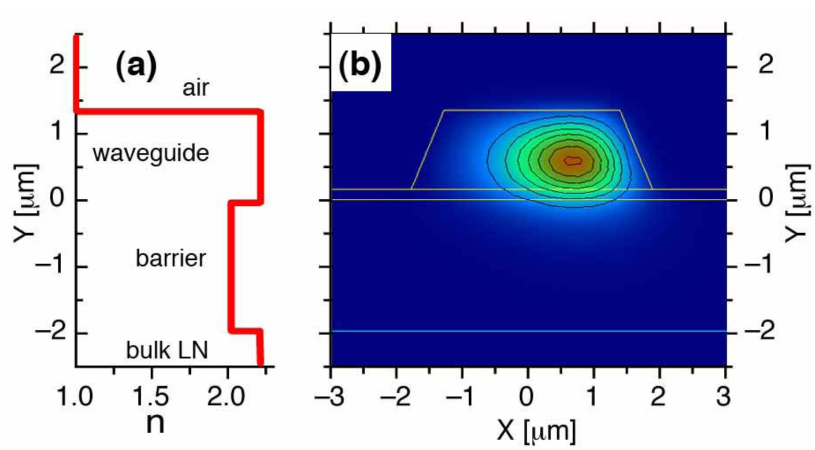


Figure 6.8: Simulation of light propagation in the waveguide of interest. (a) Vertical index profile of the waveguide structure (at X = 0). (b) Simulated electric field profile of the first TE optical mode at $\lambda = 1.55$ μm in the waveguide with a 80-μm bend radius. Optical barrier width is 2 μm. The waveguide cross-section is trapezoidal with a waveguide height of 1.35 μm, a ridge height of 1.2 μm, a base width of 3.7 μm, and a top width of 2.7μm.

6.4 Blue Lasers

The pioneering work in development of diode pumped frequency doubled blue lasers based on $KNbO_3$ was performed by Günter et al. already in 1979. Since then, the Nonlinear Optics Laboratory played a leading role in the further development of blue, compact all-solid-state lasers. The knowledge accumulated in the NLO Laboratory led to a very successful technology transfer to the ETH spin-off company Rainbow Photonics, who was the first producer of all-solid state blue lasers. Lasers with a wavelength of 430 and 488 nm were first exhibited at the Interational Laser Trade Fair in Munich in 1997, and at the Conference on Lasers and Electro-Optics (CLEO) in San Francisco in 1998.
Two different approaches were followed. In the first one, continuous wave infrared diode lasers (860 and 976nm) were frequency narrowed and stabilized using an external cavity. These diodes were used as a pump source for the second harmonic generation of the blue light (430 and 488nm) in $KNbO_3$ crystals. In order to increase the conversion efficiency, intensive research was performed in development of $KNbO_3$ waveguides. A review of this work is presented in [Fluck et al., 2000]. Fig. 6.9 shows an infrared tunable laser (Cr:LiSAF) with intracavity frequency doubling ($KNbO_3$ crystal) produced by Rainbow Photonics.

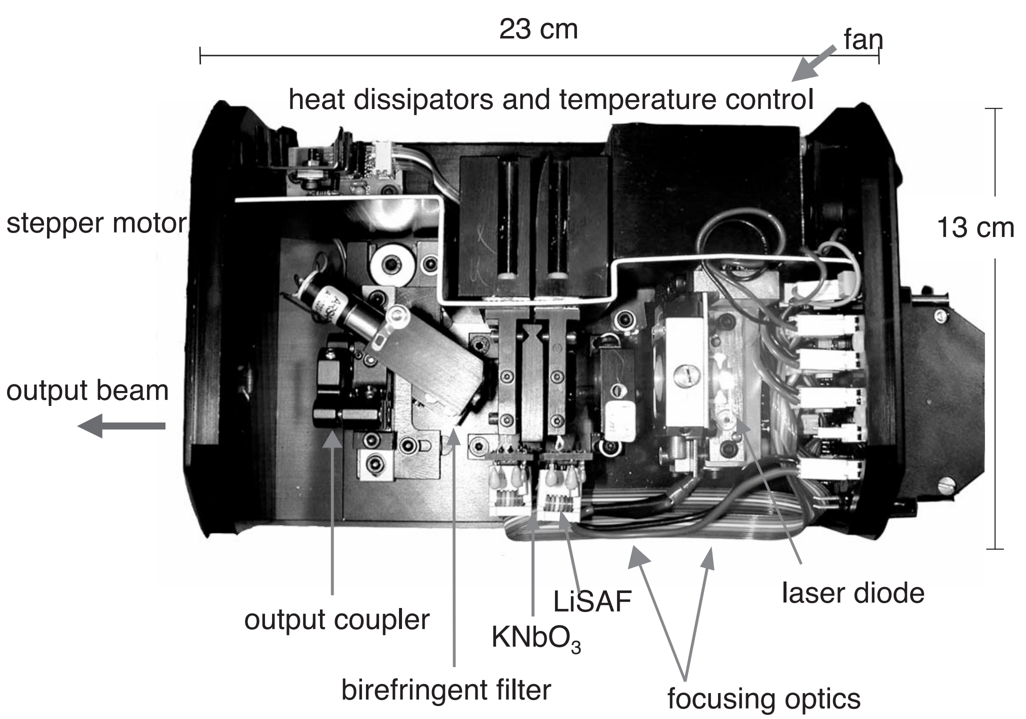


Figure 6.9: Tunable Cr:LiSAF laser with intracavity frequency doubling

6.5 UV SHG in β-BaB$_2$O$_4$ Waveguides

Compact, continuous-wave (CW) all-solid-state ultraviolet (UV) lasers operating in the wavelength range of 200-350 nm would be very attractive for several spectroscopic and biomedical applications, as well as in microlithography and optical data storage. Diode lasers emitting below 350 nm are extremely difficult to produce. Therefore, the only technique to build all-solid-state CW lasers operating below 350 nm at present is optical frequency conversion of visible lasers in UV transparent nonlinear optical crystals. Borate crystals, such as β-BaB$_2$O$_4$ (BBO) offer a wide transparency range down to 190 nm. Despite its relatively low nonlinearity ($d_{22} = 2.3$ pm/V), a high conversion efficiency has been demonstrated using BBO crystals in resonant cavities pumped by high power lasers. The last approach, however, requires an active stabilization of the cavity length and a single frequency pump laser, thus leading to a complex and expensive laser system.

In contrast, optical waveguides offer the advantage of maintaining high optical intensities over a long interaction length, enabling a high conversion of the pump beam in a single-pass configuration. Such waveguide-based laser systems are simple, compact, robust, and distinguished by less stringent spectral requirements for the pump laser. There is an increasing interest in realization of nonlinear optical waveguides for the deep UV laser light generation, since cheap and powerful green frequency-doubled Nd:YAG pump lasers are available on the market.

Recently, we have realized optical waveguides in BBO combining different techniques. Planar waveguides were produced by He$^+$ ion implantation, while the structuring of channel waveguides has been achieved by fs-laser ablation or photolithographic patterning combined with Ar plasma etching [Degl'Innocenti et al., 2006]. Although the photolithographic patterning is technically difficult to perform because of the hygroscopicity of BBO crystals, such waveguides show lower propagation losses (< 5 dB/cm for TM polarized pump light at 532 nm, and < 10 dB/cm for TE polarized generated light at 266 nm) and are thus advantageous for the SHG of the UV light.

Fig. 6.10 shows a front polished surface of a BBO crystal with photolithographically structured optical waveguides for the SHG generation of the UV light. Fig. 6.11 depicts the SHG power at 266 nm generated in BBO waveguides as a function of the fundamental internal input power at 532 nm. The maximum UV output power of 0.32 mW with a fundamental input power of 670 mW was achieved, corresponding to an internal conversion efficiency $\eta = P_{2\omega}/P_\omega^2$ of

0.07 %W^{-1} [Degl'Innocenti et al., 2008].

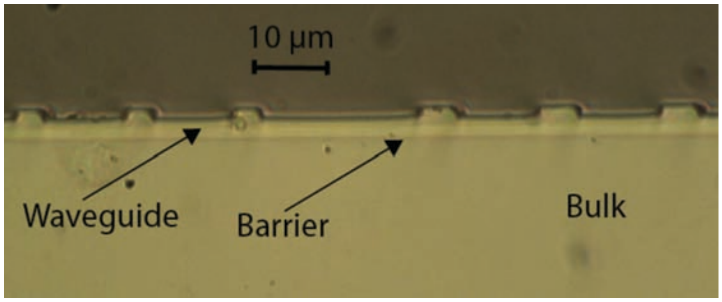


Figure 6.10: Front polished surface of a BBO crystal with structured optical waveguides for the SHG generation of the UV light. The waveguides having widths between 3 and 5 μm and a height of 1.7 μm were fabricated by lithographic patterning and Ar$^+$ ion etching. The guiding region and the implanted optical barrier are clearly seen.

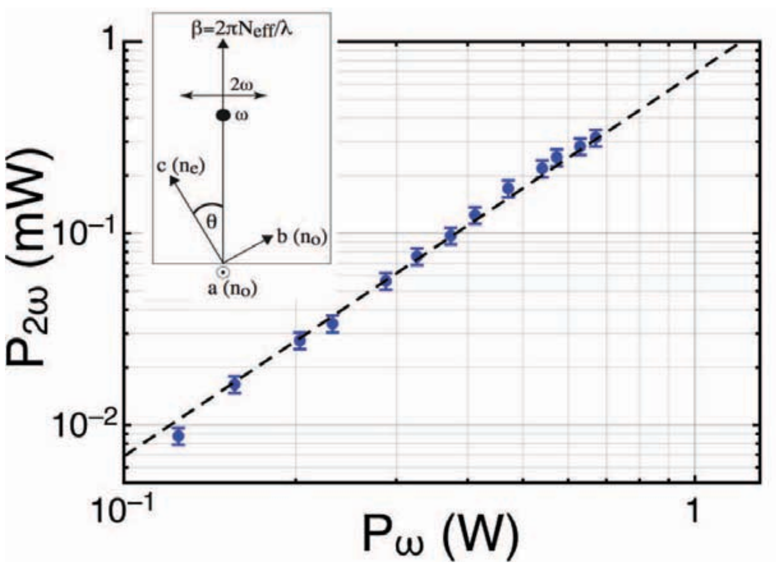


Figure 6.11: Second harmonic power at 266 nm generated in BBO waveguides as a function of the fundamental internal input power at 532 nm. The crystal temperature was 32.1°C. The dashed line represents a quadratic fit to the measured data. The inset shows the crystal orientation for type I SHG in BBO waveguides: the light propagation direction is along $\beta = 2\pi N_{eff}/\lambda$, where N_{eff} is the effective mode index, and a, b, c are the main crystallographic axes.

6.6 Ion-Sliced $LiNbO_3$ Thin Films and Photonic Devices

As optical components continue to replace electronics for optical signal processing devices, there is a growing impetus to integrate more photonic devices onto a single chip. Lithium niobate is an attractive ferroelectric material for the development of photonic chips due to its excellent electro-optic and nonlinear optic properties. However, at present there are no practical nonlinear optical integrated circuits of $LiNbO_3$ other than simple modulators and second harmonic generation devices. These are based on conventional waveguide fabrication techniques, such as metal in-diffusion and ion exchange. Although these waveguides show very low propagation losses, their optical confinement is very weak due to a relatively low refractive index contrast ($\Delta n < 0.1$). Consequently, these waveguides suffer from high bending losses, which prevent their use for high-density integrated optics.

A key step towards realization of high-index-contrast ferroelectric waveguides is to produce single-crystalline thin films, which could be structured and embedded in low-index dielectric materials. In our laboratory, we have developed an improved method for the fabrication of large area single-crystalline $LiNbO_3$ films on low-index substrates [Guarino et al., 2007]. This method generally known under the name of "Smart Cut" or "Crystal Ion-slicing" was originally discovered and applied for the fabrication of silicon-on-insulator (SOI) wafers. It is based on high-dose He^+ implantation for cleaving submicrometer thin $LiNbO_3$ films from a bulk crystal. Ion-sliced films have a very uniform thickness which can be easily controlled by adjusting the energy of implanted ions. One of the main challenges is the proper bonding of submicrometer thick films on handling substrates. We have successfully solved this technical problem by using an adhesion polymer benzocyclobutene (BCB) which has a good optical transmission and a low refractive index of 1.5 in the telecom wavelength range around 1.55 μm. Thus, the refractive index contrast between the $LiNbO_3$ films and the adhesion polymer is as high as 0.65.

The fabrication procedure is shown in Fig. 6.12. Its detailed description can be found in [Guarino et al., 2007; Poberaj et al., 2009]. Using our novel method, high quality $LiNbO_3$ thin films with a thickness of 600 nm and a surface of several cm^2 have been routinely produced. Fig. 6.13 (left) shows a photograph of a $LiNbO_3$ thin film (12 mm x 10 mm x 600 nm) bonded by a 2-μm thick BCB layer on a Cr-coated $LiNbO_3$ handling substrate. A high mechanical and

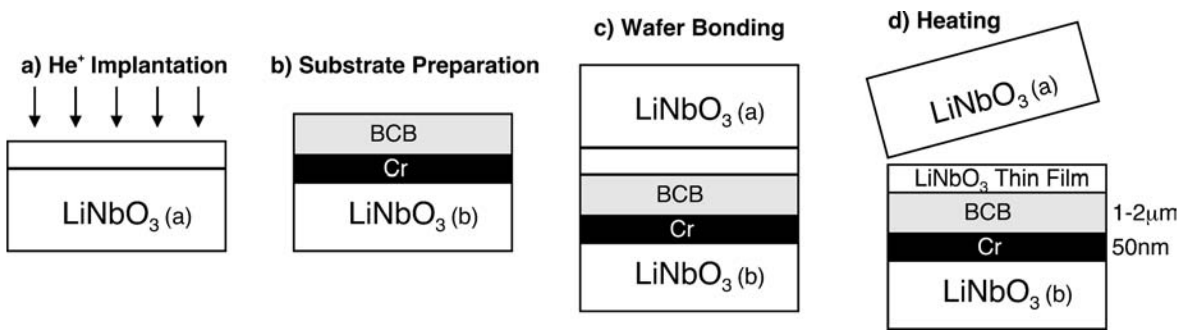


Figure 6.12: Schematic representation of the $LiNbO_3$ thin film fabrication method: (a) He^+ implantation ($E = 195$ keV, $D \sim 4 \times 10^{16}/cm^2$) of $LiNbO_3$ donor wafer; (b) preparation of $LiNbO_3$ receptor wafer (substrate): deposition of Cr electrode (50 nm) and spin-coating of BCB film (1 - 2 μm); (c) wafer bonding; (d) slow ramp thermal treatment: strong bonding, $LiNbO_3$ film split-off and annealing.

chemical stability of these $LiNbO_3$ films allows further processing steps such as lithographic structuring, cutting and polishing of the edges (see Fig. 6.13 (right)), making them very suitable as a platform for high-density integrated photonics devices.

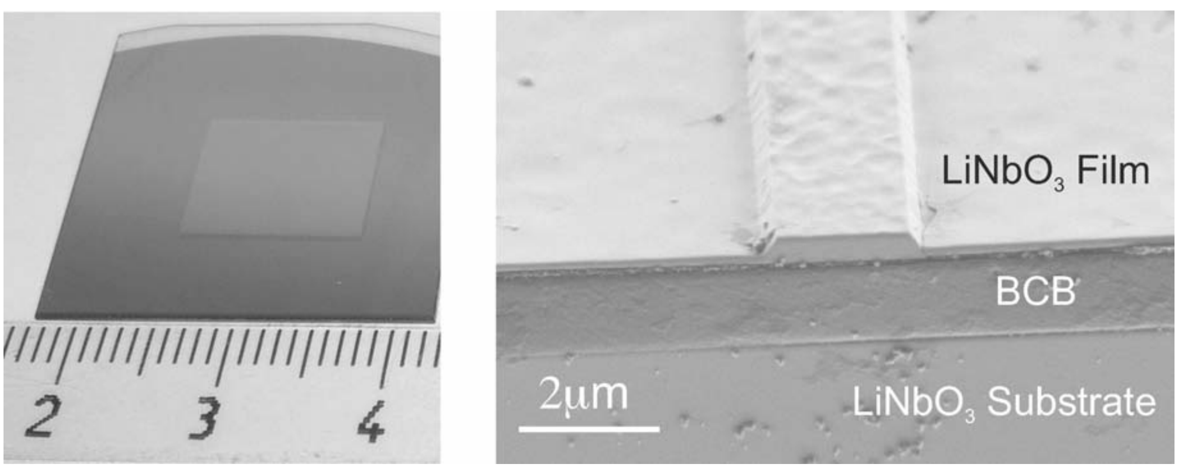


Figure 6.13: (left) Photograph of an ion-sliced $LiNbO_3$ thin film (12 mm x 10 mm x 600 nm) bonded on a Cr-coated $LiNbO_3$ substrate; (right) Polished edge of a $LiNbO_3$ thin film with a structured ridge waveguide for end-fire light coupling.

Our research has been focused on the development of electro-optically active microring resonators and photonic bandgaps. Both types of structures are distinguished by their small size and high functionality which is essential for the very large integration. For their prototyping we have used two different structuring techniques. Microring resonators and coupling waveguides with a typical width of 1 μm are patterned by a home-built laser lithography system and etched in Ar plasma. Fig. 6.14a shows a scanning electron micrograph of a race-tracked microring resonator with a free spectral range of 7 nm. Photonic bandgap structures in $LiNbO_3$ thin films have been structured by focused ion beam milling. Fig. 6.14b shows a triangular photonic crystal lattice with a lattice constant of 1μm. Cylindrical holes have a diameter of 250 nm and a depth of 1.5 μm.

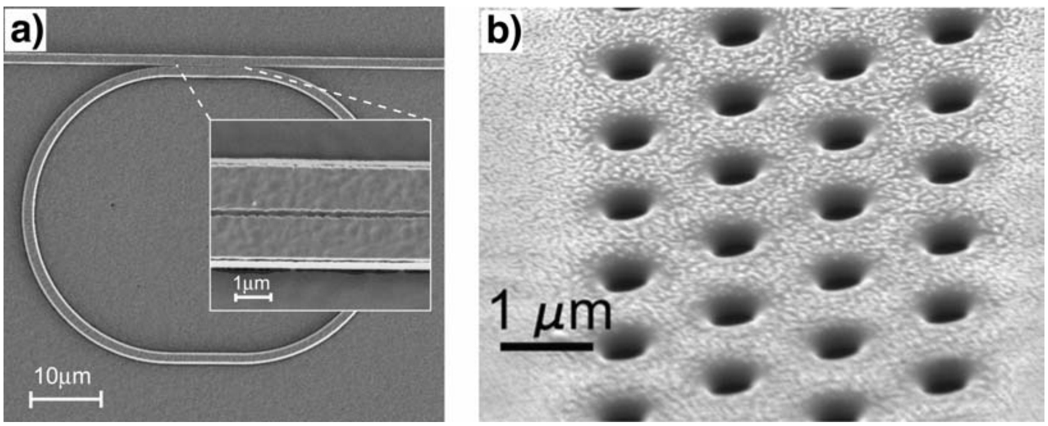


Figure 6.14: (a) Racetrack microring resonator in a $LiNbO_3$ thin film structured by the laser lithographic patterning and Ar^+ ion etching. (b) Scanning electron micrograph of a triangular photonic crystal lattice in a $LiNbO_3$ thin film fabricated by focused ion beam milling. The lattice constant is 1 μm, the air holes have a diameter of 250 nm and a depth of 1.5 μm. A 5-nm thick gold electrode was deposited on the top to obtain a sharp micrograph.

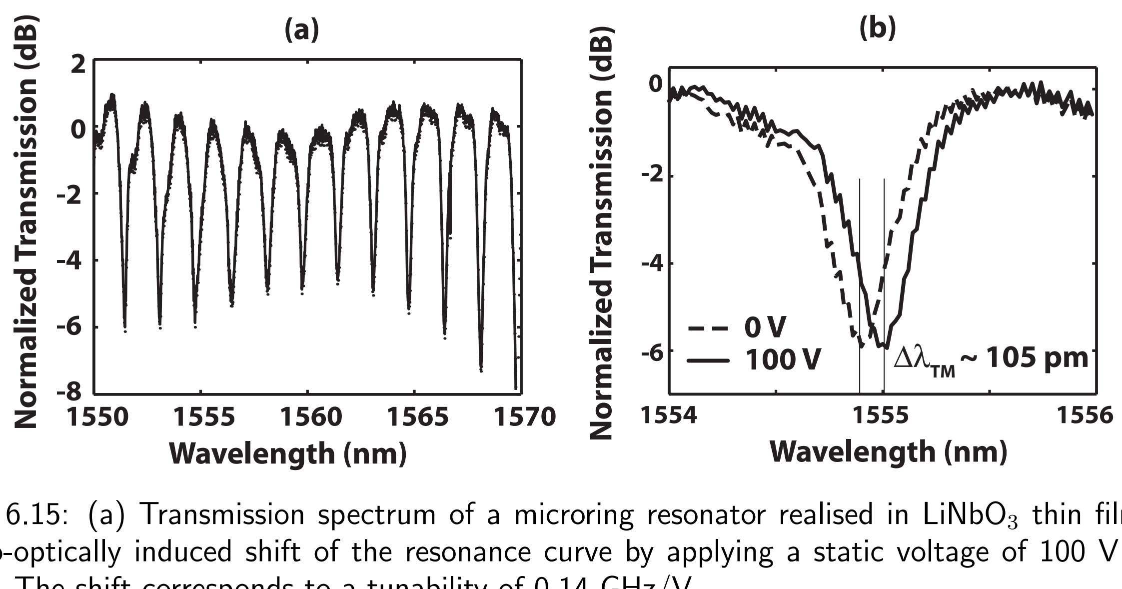


Figure 6.15: (a) Transmission spectrum of a microring resonator realised in $LiNbO_3$ thin film. (b) Electro-optically induced shift of the resonance curve by applying a static voltage of 100 V to the device. The shift corresponds to a tunability of 0.14 GHz/V.

The first electro-optically tunable microring resonators in $LiNbO_3$ thin films had a radius of 100 μm and a free spectral range of 1.6 μm [Guarino et al., 2007]. The measured transmission spectrum and electro-optical tunability (0.14 GHz/V) are depicted in Fig. 6.15. Further optimization of the electrode configuration will be performed in order to increase the tunability beyond 1 GHz/V.

Chapter 7

Photorefractive Optics

Photorefractive materials present a special group of nonlinear optical materials with the ability to detect, store and transmit light intensity patterns. This is possible already at very low light intensity levels due to the unique combination of different physical mechanisms involved in such materials: internal photoeffect, photoconductivity, charge trapping and the electro-optic effect. The richness of the physical mechanisms and a remarkable sensitivity of the effect to only moderate changes of material composition has led to a wide spectrum of different macroscopic effects that are interesting for a variety of applications in laser photonics. Particularly interesting are the possibility for the energy transfer between coherent beams, generation of self-pumped optical phase conjugated beams, self-organizing optical cavities, optical control of the group velocity of modulated signals, generation of dynamic optical waveguides and solitons, and many others, mostly based on dynamic holography and/or quasi-permanent holographic storage.

We have conducted research in the field of photorefractive optics considering three main directions, material development, characterization of materials and effects, and device oriented research. In 2006 and 2007, the state of photorefractive research worldwide has been reviewed in three book volumes co-edited by Prof. Peter Günter and published by Springer Series in Optical Sciences, covering basic effects, materials and applications. Some of our achievements are presented in the following sections.

7.1 Photorefractive Materials

We have been active in developing novel or modified materials, investigating the basic physical processes involved in the photorefractive effect in different materials and optimization for various spectral regimes and various applications. The core of our research has been based on inorganic crystals, e.g., $KNbO_3$, $BaTiO_3$, $LiTaO_3$, $LiNbO_3$, $Sn_2P_2S_6$, as well as organic crystals, polymers, and liquid crystals.

Potassium niobate ($KNbO_3$). Our laboratory has a worldwide leadership in preparing high-quality and large-size crystals of $KNbO_3$, a material with excellent electro-optic and nonlinear optical properties. By the optimized top-seeded solution growth $KNbO_3$ crystals contain different kinds and concentrations of dopants, such as Fe, Mn, Ni, Rh, and other ions were grown with the aim of optimizing their photorefractive performance [Zgonik et al., 1995]; some examples are shown in Fig. 7.1. A high temperature, post-growth reduction process in a CO/CO_2 atmosphere was developed and applied to the reproducible reduction of iron doped $KNbO_3$. Crystals produced in this way show a very high sensitivity at visible wavelengths, which is well beyond the sensitivities that can be obtained with most other materials. For example, reduced $KNbO_3$ crystals were shown to reach the fastest response times in the microsecond range for cw applications in the visible region [Voit et al., 1987]. Rh-doped $KNbO_3$ crystal submitted to the same kind of treatment showed an increase of the sensitivity at near infrared wavelengths by more than four orders of magnitude [Ewart et al., 1997].

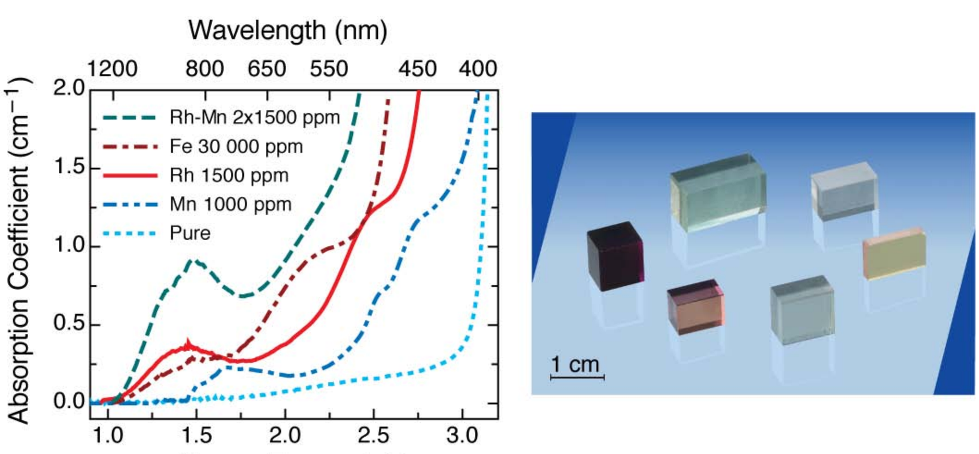


Figure 7.1: Absorption spectra and photographs of pure and doped $KNbO_3$ crystals.

Organic photorefractive materials. First demonstration of photorefraction in organic media was performed in our group in 1990 using single crystals of 2-cyclooctylamino-5-nitro-pyridine (COANP) doped with 7,7,8,8-tetracyanoquinodimethane (TCNQ) to induce the photoconductivity; see Fig. 7.2. We were also active in investigating low-glass-transition photorefractive polymers showing mesophase structures (Fig. 7.3) and photorefractive liquid crystals.

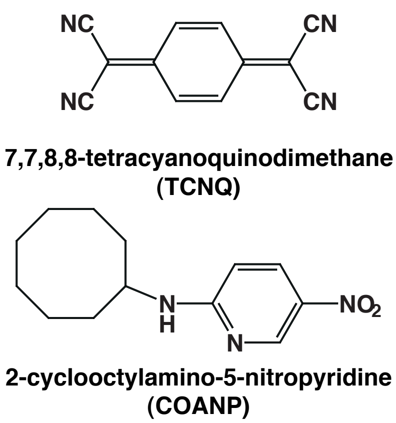


Figure 7.2: First organic photorefractive material: electro-optic crystal COANP, doped with TCNQ to induce photoconductivity [Sutter et al., 1990].

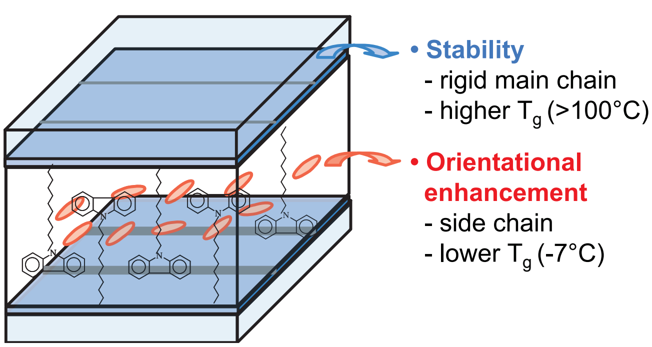


Figure 7.3: Photorefractive polymer composite based on the layered photoconducting PPT-CZ (poly(p-phenyleneterephthalate)-carbazole) polymer, the nonlinear optical chromophore diethylaminodicyanostyrene and the sensitizer C_{60}. Due to the low glass transition related to the side chains, no plasticizer is needed and therefore the phase stability is superior to the conventional composites [Kwon et al., 2003]. These composites also exhibit excellent photorefractive properties, gain of more than 250 cm^{-1} in transmission and 100 cm^{-1} in reflection grating geometries (at 633 nm and with an applied field of 60 V/μm).

Tin hypothiodiphosphate ($Sn_2P_2S_6$, SPS). During the last few years, we have concentrated our research on semiconducting ferroelectric $Sn_2P_2S_6$ crystals, a new and very promising material for optical processing in the near infrared. Complete characterization of its optical and nonlinear optical properties has revealed the following remarkable results: twenty times larger electro-optic figures of merit than in the inorganic standard $LiNbO_3$ and broad transparency range up to 8 μm with the possibility for phase-matched optical parametric generation, which can be interesting for the generation of infrared wavelengths not accessible with standard nonlinear optical materials [Haertle et al., 2005]. The temperature dependence of the linear and quadratic electro-optic coefficients is shown in Fig. 7.4.

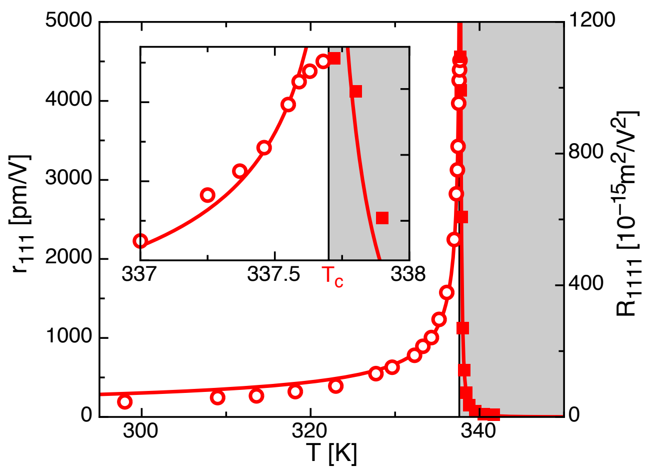


Figure 7.4: Temperature dependence of the linear electro-optic coefficient r_{111}^T ($T < T_c$) and of the quadratic electro-optic coefficient R_{1111}^T ($T > T_c$, shaded region) at 633 nm in $Sn_2P_2S_6$ [Haertle et al., 2003].

$Sn_2P_2S_6$ crystals also exhibit remarkable photorefractive properties. Stoichiometry control and doping led to an enhancement of the sensitivity in the infrared up to 1064 nm (see Fig. 7.5), with a two orders of magnitude faster response than achievable with any other photorefractive crystal in this wavelength range. For the first time in bulk ferroelectric crystals, photorefractive two-wave mixing was observed in the telecommunication wavelength range. In optimized $Sn_2P_2S_6$ crystals, we measured a relatively high photorefractive amplification gain of 6 cm^{-1} at 1550 nm using bulk Te-doped crystals, still achieving a very fast response in the order of 10 ms [Mosimann et al., 2007].

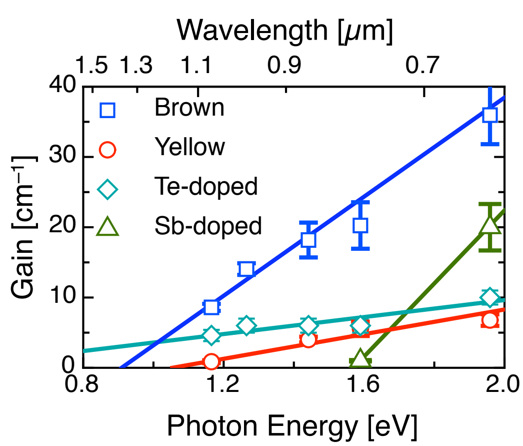


Figure 7.5: Measured maximal two-wave mixing gain coefficient as a function of the photon energy $h\nu$, respectively the wavelength λ, for different types of $Sn_2P_2S_6$ crystals [Bach et al., 2007].

7.2 Photorefractive Effects

Anisotropy of material properties. Along with other materials such as $BaTiO_3$, a full characterization of the linear optical, nonlinear optical, electro-optic, dielectric, piezoelectric, elastic and elasto-optic tensor properties has been performed [Zgonik et al., 1993; Zgonik et al., 1994]. The complete knowledge of these parameters is important in order to correctly evaluate the effective dielectric permittivity and the effective electro-optic coefficient that must be used in conjunction with the spatially modulated space-charge electric fields connected to the photorefractive effect; see also Fig. 7.6.

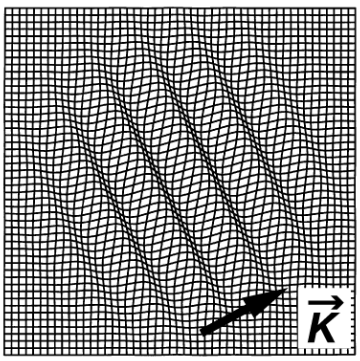


Figure 7.6: In photorefractive and related phenomena, neither the unclamped nor the clamped linear electro-optic coefficients should be used for calculating the refractive-index change due to the periodic space-charge field. We have shown that the total refractive-index change should be calculated by adding together the strain-free electro-optic contribution and the elasto-optic contribution, properly taking into account also the rotation of the axes of the optical indicatrix, which is produced by shear deformations [Gunter et al., 1991; Montemezzani et al., 2006].

We have shown that the effect of the strong anisotropy of different material tensors involved in the photorefractive effect must be correctly analyzed in order to explain many of the observed phenomena, and still forms the basic of finding optimal configurations for various volume holographic applications. As an example, the initially puzzling characteristic three-lobed structure of the beam fanning in $BaTiO_3$ shown in Fig. 7.7 was found to be a direct consequence of the interplay of the various anisotropic tensor properties.

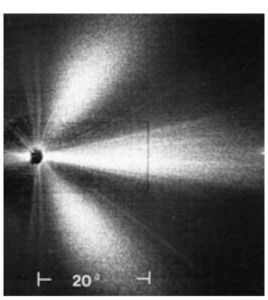


Figure 7.7: Experimentally observed far-field distribution of amplified scattered light (fanning) in $BaTiO_3$. The clamped or unclamped electro-optic tensors do not predict the upper two lobes. We could correctly predict the lobe structure by considering the full set of relevant electro-optic, piezoelectric, elastic, and elasto-optic tensor parameters [Montemezzani et al., 1995].

The determination of some material parameters like mobility anisotropy (Fig. 7.8) by holographic techniques was also only possible after taking into account the complete set of the related material tensor parameters.

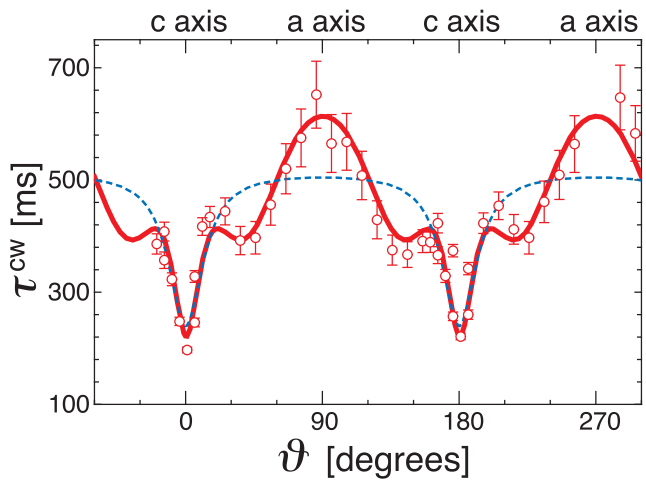


Figure 7.8: Relaxation time of the photoinduced space-charge grating in a $BaTiO_3$ crystal as a function of the angle between the grating vector $\boldsymbol{k}$ and the c-axis, which was measured to determine the mobility anisotropy in this crystal. The solid line represents the theoretical curve derived from the elastic and piezoelectric properties of $BaTiO_3$, resulting in hole-mobility anisotropy of $\mu_a/\mu_c = 19.6 \pm 0.6$. The dashed curve was obtained considering the uncorrected dielectric constants [Bernasconi et al., 1997].

Short-pulse photorefraction. We have investigated free-carrier mobility and free-carrier lifetime of photoexcited charges of low mobility electro-optic materials by using all-optical contactless methods, space-charge relaxation and holographic time-of-flight techniques with picosecond time resolution (see Fig. 7.9). Using ps laser pulses, the mobility was determined by observing the movement of electrons in the first few nanoseconds after the photoexcitation by means of a pump-probe technique and as a function of the grating spacing of the interference pattern. When performed as a function of temperature, such studies permit to elucidate the polaron nature of charge transport in inorganic crystals such as $KNbO_3$, $Bi_{12}SiO_{20}$, and others.

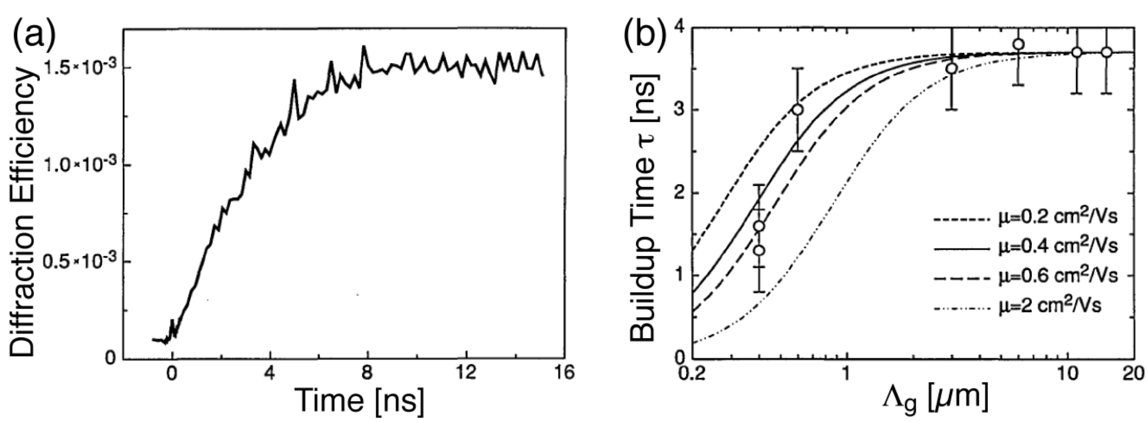


Figure 7.9: Holographic-Time-of-Flight technique (HTOF). (a) Buildup of a photorefractive grating in reduced $KNbO_3$ after illumination with 70-ps pulses. The grating is oriented along the c axis of the crystal and has a grating period of 0.8 μm. (b) Buildup time of the photorefractive grating after pulsed excitation as a function of the grating period Λ_g. The curves give theoretical predictions for various values of the carrier mobility μ. The large grating period limit gives the free electron lifetime [Biaggio et al., 1992].

Fixing mechanisms. The main advantage of the photorefractive effect is that it can be used for real-time dynamic holography, which usually means that the space-charge grating structures are erased as fast as they are written. For some applications, e.g., holographic optical memory, it is however desired that the written structures remain permanent or quasi-permanent. We have performed an extended experimental study and developed a detailed theory of the effect of hologram fixing through high-temperature charge compensation or ionic fixing [Montemezzani et al., 1993]. Figure 7.10 shows an example of high-temperature charge compensation in $KNbO_3$, as well as the theoretical evolution of the compensating fields during this process, which are connected to electrons and ionic charges, respectively. We have also proposed a new approach of photorefractive

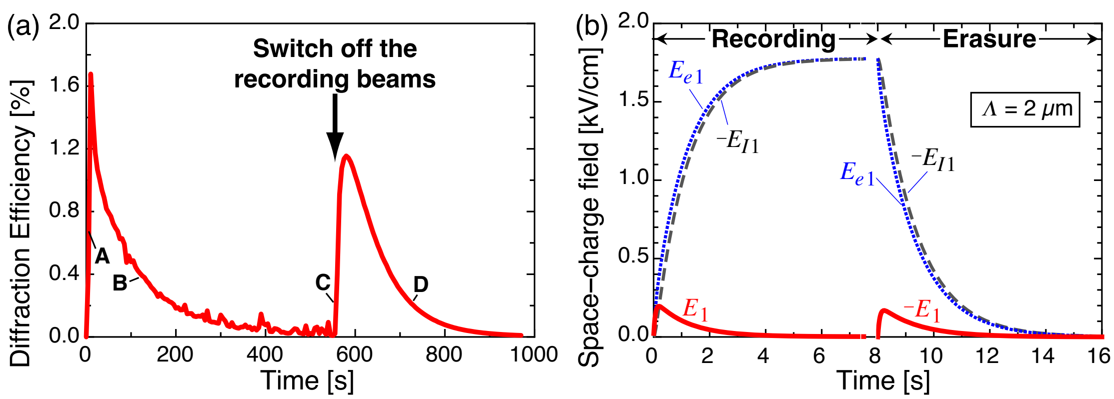


Figure 7.10: (a) Experimental dynamics of high temperature charge compensation (phase B) leading to fixing of photorefractive gratings in $KNbO_3$; the temperature is 95°C. (b) Corresponding theoretical evolution of the photorefractive space-charge field showing that the two compensating fields due to electrons (E_{e1}) and ions (E_{I1}) can be much larger than the "visible" resulting space-charge field (E_1, solid line). The speed of the dynamic evolution depends on temperature and on the grating spacing Λ. [Montemezzani et al., 1990; Montemezzani et al., 1993]

grating fixing at room temperature by means of ferroelectric domains (electrical fixing), in which a sample is depoled prior to grating recording [Cudney et al., 1994].

Interband photorefractive effects. In usual photorefractive experiments one relies on the electron or hole photoexcitation from impurity levels lying within the material energy band gap. However, band-to-band photoexcitation can also lead to charge separation and holographic grating formation. First experimental demonstration and the development of a full theoretical model of the interband photorefractive effect have been performed in our group. We have

shown that the photorefractive effect using band-to-band photoexcitation features several important advantages compared to the conventional excitation from the energy levels within the band gap. Due to the increased photoexcitation rate of band-to-band photoexcitation, the recording speed increases by two to three orders of magnitude compared to the conventional effect in the same material. Such gratings are also particularly robust against illumination at sub-bandgap photon energies [Montemezzani et al., 1994; Montemezzani et al., 2006].
Based on the interband photorefractive effect, we have proposed and demonstrated several application schemes; some of them are described in the next section.

7.3 Applications of the Photorefractive Effect

Light deflection and modulation. We have demonstrated that using anisotropic Bragg diffraction in a noncritical configuration makes it possible to achieve very large beam deflection angles, up to 5.7 degrees in $KNbO_3$ crystals for a wavelength of 633 nm. For writing, we used an argon-ion laser with a wavelength tuning range of 458–514 nm, without any beam tilting being required [Voit et al., 1986]. Fig. 7.11 presents an all-optical fiber multiplexer using the anisotropic Bragg diffraction to deflect the incoming signal beam and couple it into different output fibers. Anisotropic self-diffraction is also ideally suited for spatial light modulation or incoherent-to-coherent conversion [Voit et al., 1987].

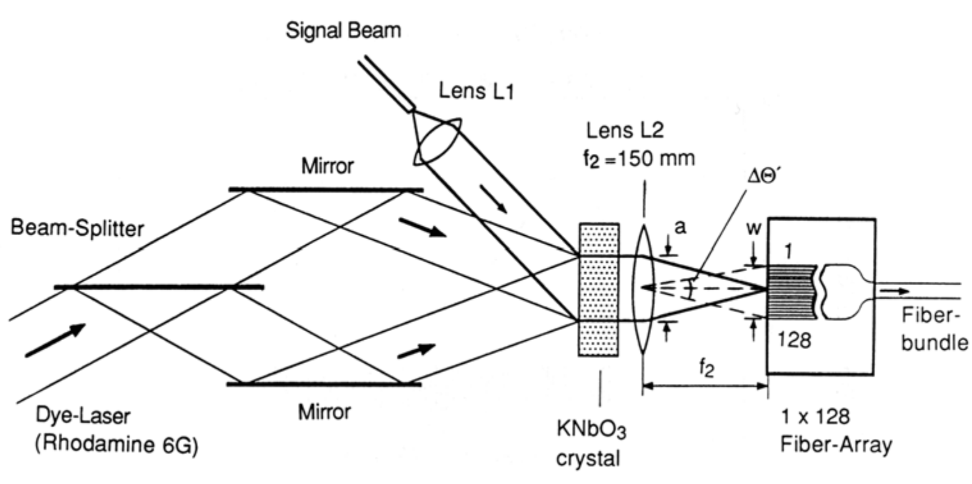


Figure 7.11: Set-up of an optically-controllable fiber-interconnection system. By changing the control wavelength (dye laser), the signal beam can be coupled into any of the linearly arranged fibers [Voit et al., 1989].

Associative memory. An associative memory is an element that is able to retrieve the whole information out of a partial input. The goal can be for example to mimic the associative power of the neural network of the human brain. Due to inherent parallelism and the possibility of spatially distributing the information content, optical and holographic techniques are very attractive for the implementation of simple associative memories. As an example, Figure 7.12(a) shows the schematic set-up of an associative memory that was implemented using a hybrid photorefractive-liquid crystal optical cavity [Ingold et al., 1992]. The comparison with the stored images occurs in the photorefractive crystal (PRC) within the non-resonant nonlinear cavity by the two-wave mixing process. The nematic liquid crystal cell (NLC) at the output of the cavity acts as a thresholding element for image discrimination, thanks to its bistable switching characteristics within the cavity. A simpler implementation, where the library images are stored in a holographic $LiNbO_3$ memory, is shown in Fig. 7.12(b) [Duelli et al., 1995]. Here the selection of the output-associated image is performed within a phase-conjugate feedback cavity containing a UV-activated saturable absorber for thresholding. The latter system can retrieve an image even if only 1/500 of it is presented at the input.

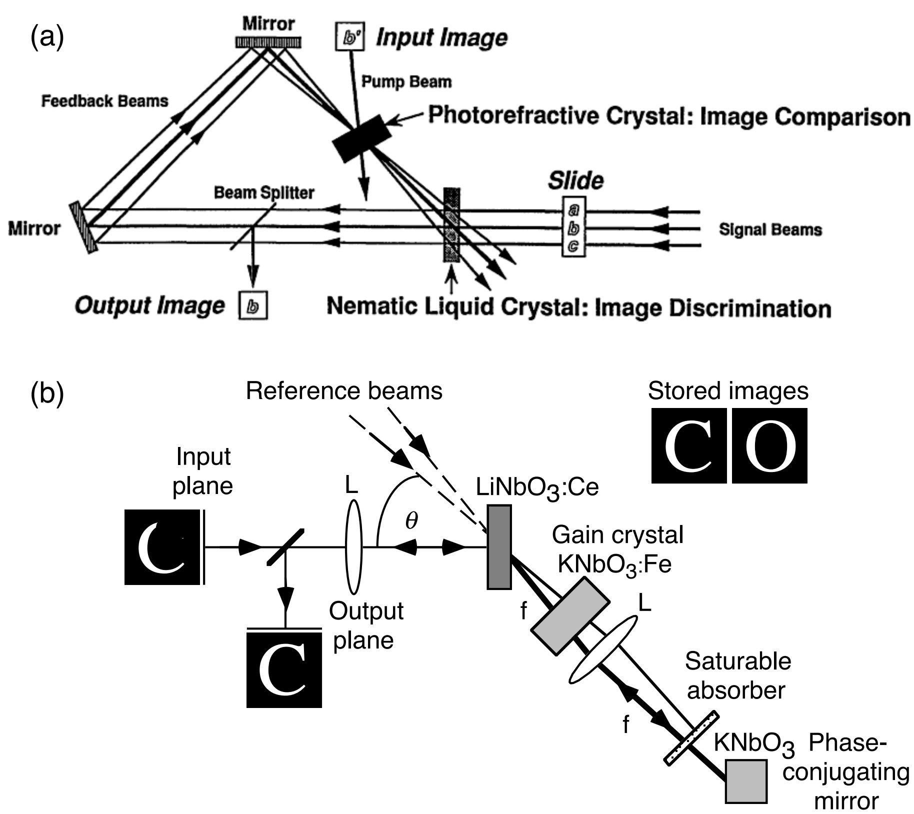


Figure 7.12: Schematic set-ups for all-optical associative memories. (a) System based on nonresonant hybrid cavity with image-bearing beams [Ingold et al., 1992]. (b) System using holographically stored images, phase conjugate feedback and an intensity-tunable saturable absorber for thresholding [Duelli et al., 1995].

Optical parallel processing. Photorefractive incoherent to coherent optical conversion, operating in the interband regime in $KNbO_3$ was demonstrated, which is interesting for high-speed and high-resolution optical processing in the UV [Bernasconi et al., 1999]. A fast read-out rate holographic image memory operating in a pulsed mode with a read-out rate exceeding 50'000 frames/s has been developed. Van der Lugt type and Joint Fourier Transform correlators based on this memory operating at a speed of 10'000 correlations per second were successfully demonstrated, see Fig. 7.13.

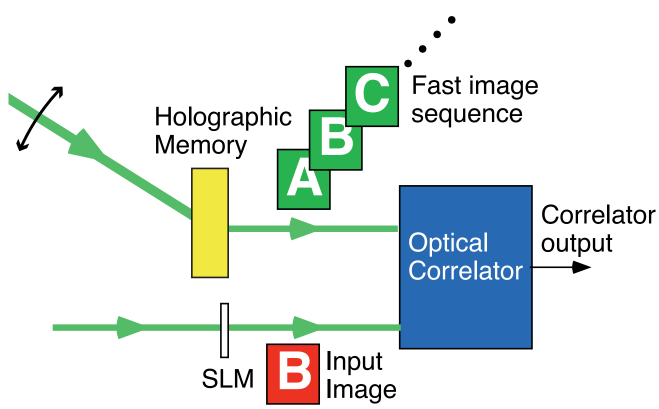


Figure 7.13: High-rate joint Fourier transform correlator: principle of operation. As a holographic memory, we used a thick $LiNbO_3$ crystal, in which we recorded angularly multiplexed volume holograms. The image correlation was performed in a thin dynamic hologram recorded in real-time in $Sn_2P_2S_6$ by the interband photorefractive effect. The correlator was working at a frame rate of 10 kHz using high repetition rate frequency-doubled Nd-YAG laser operating at 532 nm [Ryf et al., 2001].

Tunable Bragg filter. A high-wavelength-sensitivity tunable Bragg filter was developed that is interesting for wavelength division multiplexing (WDM) based on the interband effect. The filter was operating at telecommunication wavelengths near 1.55 μm and could be tuned in microseconds, which was achieved by using an acousto-optic deflector to control the direction of the recording beams and therefore the photorefractive grating spacing. As shown in Fig. 7.14, a bandwidth of 0.13 nm and fast tuning between the channels separated by 50 GHz were demonstrated using interband holographic gratings in $KNbO_3$.

Reconfigurable optical waveguides. We have shown that interband photorefraction is also very interesting for writing dynamic waveguiding structures that provide fast reconfigurable optical switching or routing elements. Compared to other techniques of writing photorefractive waveguides, this is the fastest, allowing for response times in the microsecond range. We have demonstrated such waveguides first in $KNbO_3$ using UV light at 364 nm. At 0.1 W/cm^2, we achieved a fast response of about 0.4 ms. We have also shown the applicability of

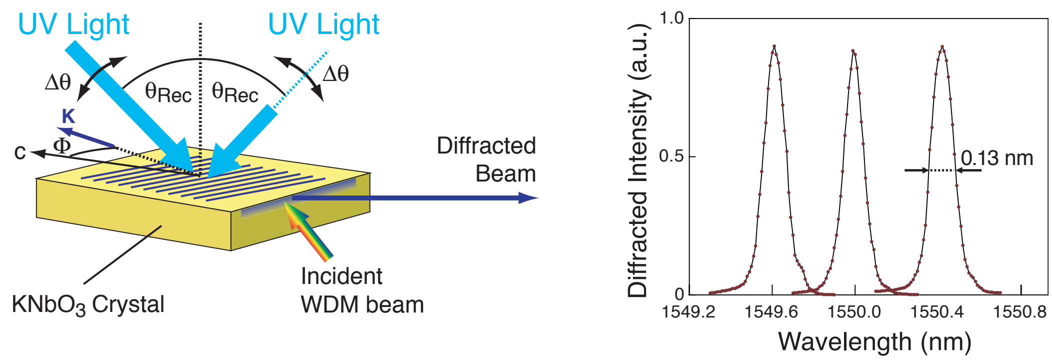


Figure 7.14: Tunable optical filter by volume dynamic holography. The Bragg angle of the filter is changed by changing the angle between the writing beams. From the incident WDM signal containing multiple wavelengths, only the one that satisfies the Bragg condition is diffracted. The result on the right was obtained by tuning the writing-beam angles to three different grating constants [Dittrich et al., 2002].

the effect to form different reconfigurable optical interconnects, either using prefabricated masks or spatial light modulators for writing the desired waveguiding structures; an example is shown in Fig. 7.15.

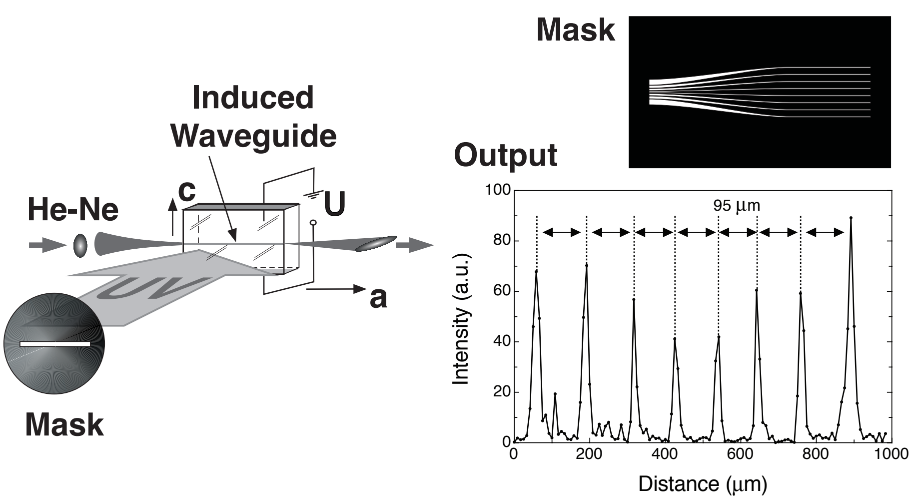


Figure 7.15: Dynamic waveguides are induced in a photoconducting electro-optic crystal by a simple principle: biasing the crystal will decrease the refractive index in the whole sample by the electro-optic effect. Where illuminated, free charges drift and screen the electric field: the refractive index in this region increases again. For certain parameters, such a structure is able to support waveguiding. The figure on the right shows the output of a dynamic 1x8 coupler using a nominally pure $KNbO_3$ crystal and the mask that is shown on the top [Dittrich et al., 1999].

The fastest response of the light-induced waveguides was obtained using pure $Sn_2P_2S_6$ crystals. The response time was below 200 μs for intensities above 0.1 W/cm^2 [Juvalta et al., 2009]. Additionally, visible green light of lower photon energy could be used for photoexcitation, which is interesting due to the widely available laser sources and optical elements (like spatial light modulators) in this wavelength range.

We have also demonstrated fixing of dynamic waveguiding structures using near-stoichiometric Mg doped $LiTaO_3$ (Mg:SLT) and deep UV controlling light at 257 nm. The structures could be either changed dynamically in the ms range or fixed during several days, i.e., the writing light and the electric field could be turned off and the structures remained in the crystal, and could be also easily removed by deep UV light illumination. This mechanism is based on the deep energy levels of $LiTaO_3$, where the structures can be stored, and it was also used to induce double dynamic and fixed waveguides [Juvalta et al., 2006].

Photorefractive waveguides. Ion implantation using MeV protons was successfully used for preparing $KNbO_3$ for special photorefractive applications, where a relatively thin active photorefractive layer is sufficient, like in optical waveguides. Samples subjected to ion implantation show a consistent and reproducible reduction state that can be varied by adjusting the irradiation dose. Very high photorefractive gain coefficients of up to 34 cm^{-1} at a wavelength of 514.5 nm were achieved in ion implanted $KNbO_3$ and the fastest build-up time of 34 μs at an intensity of 200 W/cm^2 (corresponding to a power of only 2.5 mW in the interaction region) [Zha et al., 1993]. By proton implantation, the photorefractive sensitivity in the waveguides could be extended up to the telecommunication wavelength of 1550 nm. Although the photorefractive amplification gain was relatively low at this wavelength (0.9 cm^{-1}, see Fig. 7.16), this was the first demonstration of the photorefractive effect in the telecommunication C-band range in ferroelectric crystals [Brulisauer et al., 1996].

Photorefractive two-wave mixing was also demonstrated in He^+ implanted $Sn_2P_2S_6$ optical waveguides. The high optical nonlinearity is preserved after implantation and at the telecommunication wavelength 1.55 μm, a maximal two-wave mixing gain of 2.5 cm^{-1} has been measured in Te-doped waveguides [Mosimann et al., 2009].

Self-pumped optical phase conjugation. For many applications, it is desirable to obtain cheap, reliable, and easy to handle high-power laser sources with a good beam quality. Laser diodes are being steadily improved, and nowadays emit optical powers up to kilowatt ranges. Their beam characteristics, however, are not yet sufficient for many applications, where a high focusabil-

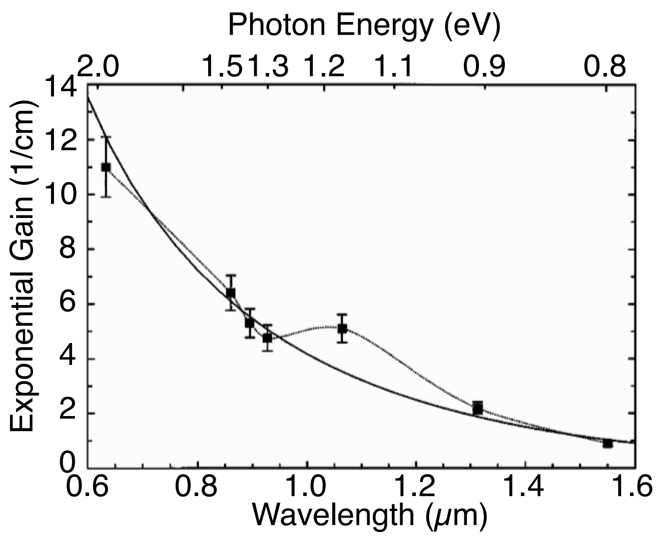


Figure 7.16: Photorefractive gain as a function of wavelength in a proton-implanted Fe-doped $KNbO_3$ waveguide [Brulisauer et al., 1996].

ity or a narrow spectral distribution is needed. Intensive research is under way worldwide to develop optical elements and configurations improving the beam quality of laser diodes. A promising method is to use a phase-conjugate injection using a photorefractive medium. We have shown that reduced Rh:doped $KNbO_3$ and various $Sn_2P_2S_6$ crystals are very promising materials to achieve this goal. High-efficiency CAT-type self-pumped optical phase conjugating element has been developed for the near-infrared wavelengths in $KNbO_3$ [Medrano et al., 1994], and ring-type in $Sn_2P_2S_6$ [Jazbinsek et al., 2005]. Particularly $Sn_2P_2S_6$:Te crystals are very interesting because of the relatively fast build-up of the phase-conjugate beam at the technologically important wavelength of 1.06 μm, see Fig. 7.17. Optical double-phase conjugation in $Sn_2P_2S_6$ has been also demonstrated and used for spectral clean-up of an emitter of a high-power laser diode bar [Bach, 2008].

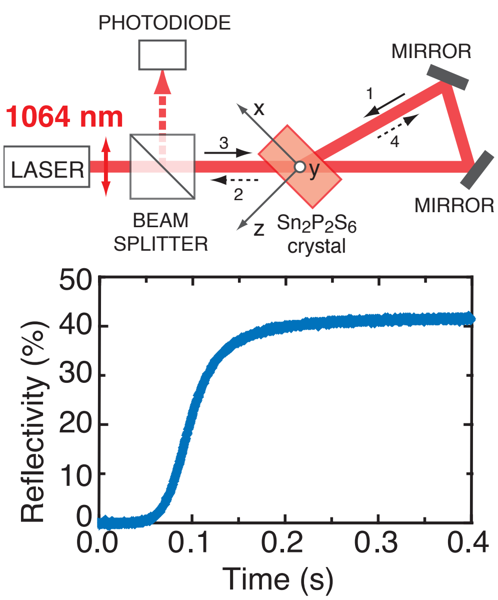


Figure 7.17: *Top:* Ring-type configuration to achieve self-pumped optical phase conjugation in photorefractive crystals is relatively simple and robust: Light entering from the left (beam 3) after passing the crystal is fed back to the crystal (beam 1) by using two mirrors forming a loop. Under certain conditions (high non-local nonlinearity), beam 2, a phase-conjugated version of the pump beam 3, will emerge, which is here monitored by a photodiode. *Bottom:* Phase conjugate reflectivity ($P_{\text{beam 2}}/P_{\text{beam 3}}$) at 1064 nm of more than 40% in Te-doped $Sn_2P_2S_6$ with a rise time of below 100 ms at an intensity of 20 W/cm^2. This is more than two-orders of magnitude faster than in commonly used Rh-doped $BaTiO_3$ crystals [Bach et al., 2005].

Chapter 8

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156. I. Liakatas, Ch. Cai, M. Bösch, M. Jäger, Ch. Bosshard, P. Günter, Ch. Zhang and L.R. Dalton
"Intermolecular Interactions of Highly Nonlinear Optical Molecules for Electro-Optic Polymer Applications", Proceedings of "Organic Thin Films for Photonics Applications"
Santa Clara, CA, USA, Technical Digest, 11-13 (1999)

157. M. Jäger, S. Lecomte, U. Gubler, Ch. Bosshard, P. Günter, L. Gobbi and F. Diederich
"Reversible Structuring of Optical Waveguides using Optical Switch Molecules", Proceedings of "Organic Thin Films for Photonics Applications"
Santa Clara, CA, USA, Technical Digest, 194-196 (1999)

158. G.I. Stegeman, R. Fuerst, R. Malendevich, R. Schiek, Y. Baek, I. Baumann, W. Sohler, G. Leo, G. Assanto, Ch. Bosshard and P. Günter
"Unique Properties of Quadratic Solitons"
Proceedings of the IV International Workshop NOA'98, Miedzyzdroje, PL, Acta Physica Polonica A 95 (5), 691-703 (1999)

159. U. Gubler, Ch. Bosshard, P. Günter, M.Y. Balakina, J. Cornil, J.L. Bredas, R. Martin and F. Diederich
"Scaling Laws of Second-Order Hyperpolarizabilities in Molecular Wires"
Proceedings of Conference on Lasers and Electro-Optics (CLEO 2000), San Francisco, California, USA, Technical Digest CMI1, 44-45 (2000)

160. I. Liakatas, C. Cai, M. Bösch, C. Fischer, M. Jäger, Ch. Bosshard and P. Günter
"Highly Efficient and Stable Bithiophene-Based Nonlinear Optical Chromophores for Polymer Electro-Optic Applications"
Proceedings of Conference on Lasers and Electro-Optics (CLEO 2000), San Francisco, California, USA, Technical Digest CMI2, 45 (2000)

161. Ch. Bosshard, U. Gubler, P. Kaatz, U. Meier and P. Günter
"Calibration of Third-Order Nonlinear Optical Susceptibilites via Non-Phasematched Optical Third-Harmonic Generation in Noncentrosymmetric Media"
Proceedings of Quantum Electronics and Laser Science Conference (QELS 2000), San Francisco, California, USA, Technical Digest QThC5, 171-172 (2000)

162. I. Biaggio
"Pulse-Length Dependent Acoustic Phonon Contributions to Degenerate Four Wave Mixing in Noncentrosymmetric Materials"
Proceedings of Quantum Electronics and Laser Science Conference (QELS 2000), San Francisco, California, USA, Technical Digest QFE7, 264-265 (2000)

163. P. Günter and Ch. Bosshard (Editors)
Proceedings of the 5th Int. Conf. on Organic Nonlinear Optics (ICONO-5), Nonlinear Optics 25 (1-4), 1-508 (2000)

164. M. Kiy, I. Gamboni, I. Biaggio and P. Günter
"Ultra-high Vacuum Reveals Interface Dependent and Impurity-gas Dependent Charge-Injection in Organic Light-emitting Diodes"
Organic Light-Emitting Materials and Devices IV, Zakya H. Kafafi, Editor, Proceedings of SPIE Vol. 4105, 290-298 (2001)

165. M.J. Orozco-Arellanes, F. Alonso, L.A. Ros and R.S. Cudney
"Indirectly Seeded Optical Parametric Generation in PPLN"
"Optics for the Quality of Life"
19th Congress of the International Commission for Optics (ICO XIX), Technical Digest (A. Consortini, G.C. Righini, Eds.), SPIE Proceedings Volume 4829, 417-418 (2002)

166. P. Günter, G. Montemezzani, D. Haertle, M. Jazbinsek and A.A. Grabar
"$Sn_2P_2S_6$ a Novel Photorefractive Material for Photonic Applications in the Visible and Infrared"
"Optics for the Quality of Life"
19th Congress of the International Commission for Optics (ICO XIX), Technical Digest (A. Consortini, G.C. Righini, Eds.), SPIE Proceedings Volume 4829, 905 (2002)

167. R. Blinc, B. Zalar, R. Kind and P.M. Cereghetti
"H-Bond Dynamics, Slater Ice Rules and Takagi Group Motion in a KDP-Type Deuteron Glass"
"Fundamental Physics of Ferroelectrics"
AIP Conference Proceedings 626, 74-80 (2002)

168. D. Haertle, G. Caimi, A. Haldi, G. Montemezzani, P. Günter, A.A. Grabar, I.M. Stoika, Yu.M. Vysochanskii

"Electro-optical Properties of Photorefractive $Sn_2P_2S_6$"
Proceeding of Photorefractive Effects, Materials, and Devices, Trends in Optics and Photonics Series (TOPS) 87, 73-78 (2003)

169. A.A. Grabar, I.V. Kedyk, I.M. Stoika, Yu.M. Vysochanskii, M. Jazbinsek, G. Montemezzani and P. Günter
"Enhanced photorefractive Properties of Te-Doped $Sn_2P_2S_6$"
Proceeding of "Photorefractive Effects, Materials, and Devices, Trends in Optics and Photonics Series (TOPS) 87, 10-14 (2003)

170. M. Jazbinsek, D. Haertle, G. Montemezzani, P. Günter, A.A. Grabar, I.M. Stoika and Yu. M. Vysochanskii
"$Sn_2P_2S_6$ Crystals for Fast Near Infrared Photorefraction and Phase Conjugation"
Proceeding of Photorefractive Effects, Materials, and Devices, Trends in Optics and Photonics Series (TOPS) 87, 190-194 (2003)

171. O-P. Kwon, G. Montemezzani, P. Günter and S.-H. Lee
"High Performance Layered Photorefractive Polymers in Reflection Grating Geometry"
Proceeding of Photorefractive Effects, Materials, and Devices, Trends in Optics and Photonics Series (TOPS) 87, 262-266 (2003)

172. Ph. Dittrich, G. Montemezzani and P. Günter
"Interband Photorefraction for Tunable Optical Filters at Telecom Wavelength"
Proceeding of Photorefractive Effects, Materials, and Devices, Trends in Optics and Photonics Series (TOPS) 87, 620-624 (2003)

173. U. Gubler, R. Negres, R.E. Martin, D. Hagan, Ch. Bosshard, P. Günter and F. Diederich
"White-Light-Continuum Spectroscopy to Determine Third-Order Nonlinear Optical Properties"
Linear and Nonlinear Optics of Organic Materials, Proc. SPIE 4461, Eds. M. Eich and M. Kuzyk, 90-104 (2001)

174. R. Degl'Innocenti, G. Poberaj, C. Medrano and P. Günter
"Blue and UV Solid State Waveguide Lasers"
Glass Science and Technology, Novel Optical Technologies, Proceedings of

the 3rd DGG Symposium held at the 79th Annual Meeting of the Deutsche Glastechnische Gesellschaft (DGG), Würzburg, Germany Glass Sci. Technol. 78 C , 77-83 (2005)

175. P. Günter, D. Haertle, T. Bach, M. Jazbinsek, G. Montemezzani, A.A. Grabar, I.M. Stoika and Yu.M. Vysochanskii
"$Sn_2P_2S_6$ for Near-Infrared Photorefractive Applications"
Proceedings of the 10th International Conference on Photorefractive Effects, Materials and Devices (PR05), Sanya, Hainan, China, TOPS 99, Optical Society of America, 2-7 (2005)

176. F. Juvalta, Ph. Dittrich, M. Jazbinsek, P. Günter, G. Montemezzani, K. Kitamura and Y. Furukawa
"Interband Photorefraction in Pure and Mg-Doped Near-Stoichiometric $LiTaO_3$"
Proceedings of the 10th International Conference on Photorefractive Effects, Materials and Devices (PR05), Sanya, Hainan, China, TOPS 99, Optical Society of America, 96-100 (2005)

177. -P. Kwon, S.-J. Kwon, M. Jazbinsek, P. Günter and S.-H. Lee
"High Performance Photorefractive Materials Based on Nematic-like Mesophase Photoconductive Polymer"
Nonlinear Optics, Quantum Optics 34, (1-4), 155-158 (2005)

178. G. Poberaj, P. Rabiei and P. Günter
"Smart-Cut $LiNbO_3$ Thin Film Waveguides for Integrated Optics" "Lithium Niobate from Material to Device, from Device to System" *Metz, France, 23-25.5.05, MOPS, Suplec, Metz Technical Digest, 3-8 (2005)*

179. T. Bach, M. Jazbinsek, G. Montemezzani, P. Günter, A.A. Grabar, I.M. Stoika, Yu.M. Vysochanskii
"Enhanced Near-infrared Photorefractive Properties of Te-doped $Sn_2P_2S_6$"
Holography 2005: Intl. Conf. on Holography, Optical Recording, and Processing of Information (Eds. V. Sainov and E. Stoykova), Proceedings of SPIE 6282, 625208-1-5 (2006)

180. F. Juvalta, Ph. Dittrich, G. Montemezzani, M. Jazbinsek, P. Günter, S. Takekawa and K. Kitamura
"Holographic Gratings in Pure and Mg-doped Near-stoichiometric LiTaO3

Induced by Deep-ultraviolet Light"
Holography 2005: Intl. Conf. on Holography, Optical Recording, and Processing of Information, (Eds. V. Sainov and E. Stoykova), Proceedings of SPIE 6252, 62520A-1-5 (2006)

181. A. Schneider, M. Stillhart and P. Günter
"Generation of THz and IR Radiation in DAST Crystals"
Proceedings of SPIE 6100, 61001C 1-8 (2006)

182. A. Schneider, M. Stillhart, Z. Yang, F. Brunner, P. Günter
"Improved Emission and Coherent Detection of Few-cycle Terahertz Transients Using Laser Pulses at 1.5 μm"
Nonlinear Optics and Applications II (ed. M. Bertolotti), Proceedings of SPIE 6282, 658211-1 (2007)

Chapter 9

Teaching

9.1 Lectures

- Prof. P. Günter
  Physics I + II
  Since 1997 (every second year for students of the following departments):
  - Maschinenbau und Verfahrenstechnik
  - Materialwissenschaften
  - Informationstechnologie und Elektrotechnik
  - Bau, Umwelt und Geomatik
  - Umweltwissenschaften

- Prof. P. Günter
  - Quantenelektronik I (WS 97/98, WS 99/00, WS 00/01)
  - Elektro-Optik from 1982 - 1987 (every year)
  - Nichtlineare Optik (every second year)

- Prof. R. Kind
  - Kernresonanzspektroskopie

- PD Ch. Bosshard
  - Nichtlineare Optik
  - Nichtlineare Optische Spektroskopie

- PD G. Montemezzani
  - Elektro-Optik
  - Holographie

- PD I. Biaggio
  - The Interaction of Laser Radiation with Matter, Spring Semester 2002
  - Nonlinear Optics, Fall Semester 2001

- Dr. M Jazbinsek
  - Elektro-Optik from 2004-2009 (every year)

9.2 Group Seminars

1. Winter Semester 84/85
   "Dielectric Materials and Optical Effects"

2. Summer Semester 86
   "Nonlinear Optical Effects in Molecular Crystals"

3. Winter Semester 86/87
   "Active Photorefractive Defects in Oxygen-Octaedra Ferroelectrics"

4. Winter Semester 86/87
   "Optical Signal Processing and Optical Associative Memory"

5. Summer Semester 87
   "Ultra-short Laser Pulses"

6. Summer Semester 88
   "Nonlinear Optical Effects in Organic Materials"

7. Winter Semester 88/89
   "Optical Bistability"

8. Summer Semester 89
   "Nonlinear Optical Effects in Optical Waveguides"

9. Winter Semester 89/90
   "Photorefractive Effects in Electro-optical Crystals"

10. Summer Semester 90
    "Nonlinear Optical Effects at Interfaces"

11. Winter Semester 90/91
    "Electro-optical and Nonlinear Optical Properties of Dipolar Liquids"

12. Summer Semester 91
    "Non-classical Light States and Propagation"

13. Winter Semester 91/92
    "Electro-optical and Nonlinear Optical Effects in 2-dimensional Structures"

14. Summer Semester 92
    "Matrix Methods in Optics"

15. Winter Semester 92/93
    "Optical Computing"

16. Summer Semester 93
    "Polymers in Optics"

17. Winter Semester 93/94
    "Photonic Band Gaps"

18. Summer Semester 94
    "Nonlinear Dynamics of Optical Systems"

19. Winter Semester 94/95
    "Ferro- and Pyro-Electricity"

20. Summer Semester 95
    "Imaging of Atoms and Molecules - Nanoscopical Methods"

21. Winter Semester 95/96
    "Linear and Nonlinear Optical Spectrocopy"

22. Summer Semester 96
    "(All-)optical Signal Processing: Applications vs. Basic Research"

23. Winter Semester 96/97
    "Photorefractive and Spatial Solitons"
24. Summer Semester 97
    "Electro-luminescence in Organic Materials"
25. Winter Semester 97/98
    "Charge Transport in Dielectric Materials"
26. Summer Semester 98
    "Lightwave Communication"
27. Winter Semester 98/99
    "Terahertz Waves and Nonlinear Optics"
28. Summer Semester 2000
    "Photoinduced dynamic optical gratings in femtoseconds and picoseconds range"
29. Summer Semester 2001
    "Microresonators for Photonics"
30. Summer Semester 2002
    "Optical Properties of Photonic Crystals"
31. Winter Semester 04/05
    "Terahertz Waves for Imaging and Spectroscopy"
32. Summer Semester 07
    "Photonic Crystals"

9.3 Habilitation Theses in Physics

1. Dr. Dieter Suter
   "Beyond the Two-level Atom - Methods for Atomic Sublevel Spectroscopy"
   ETH Zürich
   1993

2. Dr. Christian Bosshard
   "Third-Order Nonlinear Optics in Polar Materials"
   ETH Zürich
   1997

3. Dr. Uwe Siegner
   "Femtosecond Dynamics in Semiconductors for Ultrabroadband Optical Excitation"
   ETH Zürich
   1999

4. Dr. Bert Müller
   "Natural Formations of Nanostructures: From Fundamentals in Metal Heteroepitaxy via Islanding of Organic Compounds to Cell Behavior on Strained Ge/Si-Nanopattern"
   ETH Zürich
   2000

5. Dr. Ivan Biaggio
   "Nonlinear Optical Wave Interaction in Noncentrosymmetric Materials: Sub-Nanosecond Charge Migration and Degenerate Four Wave Mixing"
   ETH Zürich
   2001

6. Dr. Germano Montemezzani
   "Optical Wave Manipulation and Signal Processing in Anisotropic Photorefractive Materials"
   ETH Zürich
   2002

9.4 PhD Theses in Physics

1. Colla, E.L.
   "Incommensurability, Phase Transitions and Memory Effects in the Substitutionally Disordered System $[(CH_3)_4N]_2ZnCl_{4-x}Br_x$"
   ETH Nr. 8455, 1989
   (Prof. Dr. H. Gränicher & PD Dr. R. Kind & Prof. Dr. W. Petter)
   Working Place after ETH: EPFL Lausanne, Switzerland

2. Liechti, O.
   "Phase Transitions in Mixed Crystal Systems: Solid Solutions of $[(CH_3)_4N]_2$-$CuBr_xCl_{4-x}$ and of $Rb_{1-x}(NH_4)_xH_2PO_4$"
   ETH Nr. 8688, 1989
   (Prof. Dr. P. Günter & Prof. Dr. Kind)

3. Voit, Eugen André
   "Photorefractive Properties and Applications of $KNbO_3$ Crystals"
   ETH Nr. 8555, 1989
   (H. Gränicher Prof. & PD Dr. R. Kind)
   Working Place after ETH: Leica Geosystems, Switzerland

4. Rosatzin, Martin
   "Optisch induzierte, transiente Spinphänomene in atomaren Grundzuständen"
   ETH Nr. 9302, 1990
   (Prof. Dr. P. Günter & Prof. Dr. J. Mlynek)

5. Bosshard, Christian
   "Optical and Nonlinear Optical Properties of 2-Cyclooctylamino-5-Nitropyridine and 2-Docosylamino-5-Nitropyridine Molecules, Crystals and Langmuir-Blodgett-films"
   ETH Nr. 9407, 1991
   (Prof. Dr. P. Günter & Prof. Dr. H. Gränicher)
   ETH medal award for excellent PhD work
   Working Place after ETH: CSEM Alpnach, Switzerland

6. Ingold, Matthias
   "Optical Bistability, Beam Competition and Associative Memory based on Nematic Liquid Crystals and Photorefractive Crystals"
   ETH Nr. 9506, 1991

(Prof. Dr. P. Günter & Prof. Dr. H. Gränicher)
Working Place after ETH: ABB Baden, Switzerland

7. Kerkoc, P.
   "Growth of Bulk Crystals and Single Crystal Cord Fibers of 4-(N,N-Dimethylamino)-3-Acetamidonitrobenzene (DAN) and their Nonlinear Optical Properties"
   ETH Nr. 9413, 1991
   (Prof. Dr. H. Gränicher & Prof. Dr. P. Günter)
   Working Place after ETH: Gymnasium Tolmin, Slovenia

8. Amrhein, Peter
   "Photorefractive Spatial Light Modulation in $KNbO_3$ Crystals"
   ETH Nr. 9670, 1992
   (Prof. Dr. P. Günter & Prof. Dr. H. Gränicher)
   Working Place after ETH: UBS, Switzerland

9. Sutter, Kurt A.
   "Photorefractive Effects in Molecular Crystals"
   ETH Nr. 9671, 1992
   (Prof. Dr. P. Günter & Prof. Dr. H. Gränicher)
   Working Place after ETH: European Patent Attorney at E. Blum + Co. AG, Switzerland

10. Schwyn Thöny, Sylvia
    "Rf Sputter Deposition of Waveguding Epitaxial $LiNbO_3$ and $KNbO_3$ Thin Films"
    ETH Nr. 9929, 1992
    (Prof. Dr. P. Günter & Dr. H. W. Lehmann)

11. Korner, Norman
    "From Long Range Order to Glass Order: Static and Dynamic Properties of the Solution $Rb_{1-x}(ND_4)_xD_2PO_4$"
    ETH Nr. 9952, 1992
    (Prof. Dr. R. Kind & Prof. Dr. P. Günter & Prof. Dr. E. Brun)

12. Biaggio, Ivan
    "Photorefractive Effects Induced by Short Light Pulses"
    ETH Nr. 10009, 1992

(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
Working Place after ETH: Lehigh University, USA

13. Montemezzani, Germano
"Photorefractive Gratings: Fixing and Recording in the Visible and Ultraviolet"
ETH Nr. 10013, 1992
(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
Working Place after ETH: University of Metz, France

14. Gutmann, Roland
"Liquid Phase Epitaxy of Para- and Ferroelectric $KTa_{1-x}Nb_xO_3$"
ETH Nr. 10095, 1993
(Prof. Dr. P. Günter & Prof. Dr. N. Setter)

15. Pfändler, Stephan
"Local Structural Heterogeneities in Polymer Glasses and Perturbed Single Crystal Lattices: Investigations on Poly (Methyl/Methacrylate) and Potassium Niobate"
ETH Nr. 10924, 1994
(Prof. Dr. R. Kind & Prof. Dr. P. Günter & Prof. Dr. B. Meier)

16. Küpfer, Manfred
"Nonlinear Optics of Organic Monolayers at the Air/Water Interface and of Langmuir-Blodgett Film Waveguides"
ETH Nr. 11000, 1995
(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
Working Place after ETH: Leica Geosystems, Switzerland

17. Blasberg, Tilo
"Coherent Raman Scattering for Optical Detection of NMR in Pr^{3+}: $YAlO_3$"
ETH Nr. 11027, 1995
(Prof. Dr. P. Günter & Prof. Dr. M. Mehring & PD Dr. D. Suter)

18. Duelli, Markus
"Nonlinear Holographic Associative Memories"
ETH Nr. 11078, 1995
(Prof. Dr. P. Günter & Prof. Dr. U.P. Wild)
Working Place after ETH: Duelli Corp., Sunnyvale, California, USA

19. Knöpfle, Georg
”Nonlinear Optical Characterization and Crystalline Arrangement of Organic Molecules”
ETH Nr. 11159, 1995
(Prof. Dr. P. Günter & Prof. Dr. U.W. Suter)

20. Prêtre, Philippe
”Relaxation Mechanisms in Nonlinear Optical Polymers”
ETH Nr.11193, 1995
(Prof. Dr. P. Günter & Prof. Dr. U.W. Suter)
Working Place after ETH: Mems AG, Switzerland

21. Fluck, Daniel
”Ion-Implanted $KNbO_3$ Waveguides for Blue-Light Second-Harmonic Generation”
ETH Nr. 11225, 1995
(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
Working Place after ETH: Pro Archive AG, Switzerland

22. Schlesser, Raoul
”Organic Electro-Optic Crystals and Thin Films: Optical Characterization and Molecular Beam Deposition”
ETH Nr. 11456, 1996
(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
Working Place after ETH: North Carolina State University + Hexatech Inc., USA

23. Gitmans, Frank
”Growth of $LiTaO_3$ and $LiNbO_3$ Thin Films by Molecular Beam Epitaxy”
ETH Nr. 11693, 1996
(Prof. Dr. P. Günter & Prof. Dr. M. Erbudak)

24. Marty, Thomas
”Laser Cooling of Cesium Atoms: An Experimental Study of the Magneto-Optical Trap”
ETH Nr. 11771, 1996
(Prof. Dr. P. Günter & Dr. P. Thomann)
Working Place after ETH: Zuehlke Engineering AG, Schlieren, Switzerland

25. König, Thomas
    "Cluster Dynamics Processes in the Solid Solution $Rb_{1-x}(ND_4)_xD_2PO_4$ Investigated by Means of ^{87}Rb NMR Techniques"
    ETH Nr. 12027, 1997
    (Prof. Dr. R. Kind & Prof. Dr. P. Günter & Dr. J. Roos)

26. Pliska, T.
    "Potassium Niobate Channel Waveguide for Blue Light Second-Harmonic Generation"
    ETH Nr. 12222, 1997
    (Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
    Working Place after ETH: JDS Uniphase AG, Zurich, Switzerland

27. Ewart, Michael John
    "Reduced $KNbO_3$ for Photorefractive Applications"
    ETH Nr. 12484, 1997
    (Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
    Working Place after ETH: APM Technica AG, Switzerland

28. Brülisauer, Simon
    "Control of the Photorefractive Effect in $KNbO_3$ by Ion-Implantation"
    ETH Nr. 12625, 1998
    (Prof. Dr. P. Günter & PD Dr. Ch. Buchal)
    Working Place after ETH: Zhlke, Switzerland

29. Follonier, Stéphane
    "Light-Induced Charge Transport and Refractive Index Changes in Organic Electro-Optic Materials"
    ETH Nr. 12681, 1998
    (Prof. Dr. P. Günter & Prof. Dr. U.W. Suter)
    Working Place after ETH: CSEM Landquart, Switzerland

30. Bernasconi, Pietro
    "Physics and Applications of Ultraviolet Light Induced Photorefractive Gratings"
    ETH Nr. 12761, 1998
    (Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
    Working Place after ETH: Bell Labs, Alcatel-Lucent, USA

31. Seuret, Christophe
"Atomic Force Microscopy of Organic Nonlinear Optical Molecules at the Gas-Liquid and Gas-Solid Interface"
ETH Nr. 12905, 1998
(Prof. Dr. P. Günter & Prof. Dr. R. Zenobi)

32. Spreiter, Rolf
"Electro-Optical Effects in Organic Crystals"
ETH Nr. 13170, 1999
(Prof. Dr. P. Günter & Prof. Dr. U.W. Suter)
Working Place after ETH: Bruker Biospin AG, Switzerland

33. Jeitziner, Christian Anton
"NMR Studies of the Low-Temperature Structure and Dynamics of the Pseudo-Spin Glass D-RADP-X"
ETH Nr. 13257, 1999
(Prof. Dr. R. Kind & Prof. Dr. P. Günter & Dr. J. Roos)
Working Place after ETH: Siemens Schweiz AG, Zurich, Switzerland

34. Perhöfer, Harald
"Investigation of Epitaxial Ferroelectric $K_{1-y}Na_yNb_{1-x}Ta_xO_3$ on Ba Doped $KTaO_3$ Substrates"
ETH Nr. 13459, 1999
(Prof. Dr. P. Günter & Prof. Dr. M. Zgonik)

35. Ryf, Roland
"Optical Parallel Processing Based on the Photorefractive Effect"
ETH Nr. 13546, 2000
(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)
Working Place after ETH: Bell Labs, Alacatel-Lucent, USA

36. Gubler, Ulrich
"Third-Order Nonlinear Optical Effects in Organic Materials"
ETH Nr. 13605, 2000
(Prof. Dr. P. Günter & Prof. Dr. F. Diederich & PD Dr. Ch. Bosshard)
Working Place after ETH: Leister, Switzerland

37. Cereghetti, Paola Maria
"On the Dynamics of Glassy Phase States: An NMR Investigation"

ETH Nr. 13806, 2000
(Prof. Dr. Kind & Prof. Dr. B. Meier)
Working Place after ETH: Lehigh University, USA

38. Liakatas, Ilias
"Polymer Electro-optic Modulators: Materials and Devices"
ETH Nr. 13994, 2000
(Prof. Dr. P. Günter & Prof. Dr. U. Suter & PD Dr. Ch. Bosshard)
Working Place after ETH: Geitonas School Athens, Greece

39. Abplanalp, M.
"Piezoresponse Scanning Force Microscopy of Ferroelectric Domains"
ETH Nr. 14048, 2001
(Prof. Dr. P. Günter & Prof. Dr. J. Fousek)
Working Place after ETH: ABB Research, Switzerland

40. Bösch, Martin
"Electro-Optic Polymers: Photochemical Stability and In-line Modulator"
ETH Nr. 14192, 2001
(Prof. Dr. P. Günter & Prof. Dr. U.W. Suter & PD Dr. Ch. Bosshard)

41. Hagn, Gerhard
"Electro-Optic Effects and Their Applications in Indium Phosphide Waveguide Devices for Fiber Optic Access Networks"
ETH Nr. 14353, 2001
(Prof. Dr. P. Günter & Prof. Dr. H. Melchior)

42. Kiy, Michael
"Charge Injection and Transport in Organic Semiconductors"
ETH Nr. 14631, 2002
(Prof. Dr. P. Günter & Prof. Dr. B. Batlogg & PD Dr. I. Biaggio)
Working Place after ETH: VistaPrint Research + Technology, Switzerland

43. Dittrich, Philipp
"Ultraviolet Interband Photorefraction for Dynamic Waveguides and Filters, and Femtosecond Photo-structuring"
ETH Nr. 15414, 2004
(Prof. Dr. P. Günter & Prof. Dr. Esslinger & PD Dr. G. Montemezzani)
Working Place after ETH: Julius Bär, Switzerland

44. Tapponnier, Axelle
”Charge Transport and Nonlinear Optical Effects in Organic Thin Films”
ETH Nr. 15911, 2005
(Prof. Dr. P. Günter & Prof. Dr. M. Erbudak)
Working Place after ETH: Physikinstitut der Kantonsschulen Rämiühl, Switzerland

45. Haertle, Daniel
”Photorefractive and Nonlinear Optical Properties of $Sn_2P_2S_6$”
ETH Nr. 16107, 2005
(Prof. Dr. P. Günter, Prof. Dr. M. Sigrist & Prof. Dr. G. Montemezzani)
Working Place after ETH: University of Bonn, Germany

46. Schneider, Arno
”Generation and Detection of Terahertz Pulses in Organic Crystals”
ETH Nr. 16132, 2005
(Prof. Dr. P. Günter, Prof. Dr. M. Sigrist & PD Dr. Ch. Bosshard)
Working Place after ETH: ETH, Nonlinear Optics Laboratory, Zurich, Switzerland

47. Guarino, Andrea
”Electro-optic Microring Resonators in Inorganic Crystals for Photonic Applications”
ETH Nr. 17029, 2007
(Prof. Dr. P. Günter & Prof. Dr. V. Sandoghdar)
Swiss Physical Society (SPG) Prize for Applied Physics 2008
Working Place after ETH: Oclaro, Switzerland

48. Degl’Innocenti, Riccardo
”Optical Waveguides in BaB_2O_4 for Second Harmonic Generation and Electro-optic Modulation in the Deep UV”
ETH Nr. 17145, 2007
(Prof. Dr. P. Günter & Prof. Dr. M. Sigrist)
Working Place after ETH: Scuola Normale Superiore, NEST, CNR-INFM, Pisa, Italy

49. Herzog, Christian
”$K_{1-y}Na_yTa_{1-x}Nb_xO_3$ Thin Films for Integrated Electro-Optics”
ETH Nr. 17275, 2007

(Prof. Dr. P. Günter & Prof. Dr. G. Montemezzani)
Working Place after ETH: ETH Zurich, Physics Department, IT Service Group, Zurich, Switzerland

50. Mutter, Lukas
"Nonlinear Optical Organic Crystals for Photonic Applications"
ETH Nr. 17482, 2007
(Prof. Dr. P. Günter & Prof. Dr. M. Zgonik)
Working Place after ETH: EPFL, Lausanne, Switzerland

51. Losio, Paolo Antonio
"Charge Transport and Mirrorless Lasing in Organic Semiconductors"
ETH Nr. 17492, 2007
(Prof. Dr. P. Günter & Prof. Dr. B. Batlogg & Prof. Dr. J. Blatter)
Working Place after ETH: OC Oerlikon Solar, Switzerland

52. Ruiz Brunner, Blanca Estela
"Development of Organic Nonlinear Optical Crystals for Integrated Optics and THz Generation"
ETH Nr. 17514, 2007
(Prof. Dr. P. Günter & Prof. Dr. U.W. Suter & PD Dr. Ch. Bosshard)
Working Place after ETH: ETH, Nonlinear Optics Laboratory, Zurich, Switzerland

53. Rezzonico, Daniele
"Microring Modulators in Electro-optic Polymers"
ETH Nr. 17570, 2007
(Prof. Dr. P. Günter & Prof. Dr. U.W. Suter & PD Dr. Ch. Bosshard)
ETH medal award for excellent PhD work
Working Place after ETH: Fluxim AG, Switzerland

54. Bach, Tobias
"Beam Clean Up of High-Power Laser Diodes Using Photorefractive $Sn_2P_2S_6$"
ETH Nr. 17826, 2008
(Prof. Dr. P. Günter & Prof. Dr. G. Montemezzani & Dr. M. Jazbinsek)
Working Place after ETH: ETH, Nonlinear Optics Laboratory, Zurich, Switzerland

55. Hunziker, Christoph
"Crystalline Thiophene and Phenolic Polyene Derivatives for Organic Elec-

tronic and Photonic Applications"
ETH Nr. 18032, 2008
(Prof. Dr. P. Günter & Vahid Sandoghdar & Dr. M. Jazbinsek)
Working Place after ETH: AWK Group, Switzerland

56. Juvalta, Flurin
"Light-induced Waveguides, Waveguide Arrays and Switches in Photorefractive $LiTaO_3$ and $Sn_2P_2S_6$"
ETH Nr. 18105, 2008
(Prof. Dr. P. Günter & Prof. Dr. G. Montemezzani)
Working Place after ETH: Avelon Cetex Technologie AG, Switzerland

57. Mosimann, Roger
"Photorefractive Effects in $Sn_2P_2S_6$ at Near Bandgap and Telecommunication Wavelengths"
ETH Nr. 18140, 2008
(Prof. Dr. P. Günter & Prof. Dr. G. Montemezzani & Dr. M. Jazbinsek)
Working Place after ETH: AWK Group, Switzerland

58. Majkic, Aleksej
"Electro-optically Tunable Microring Resonators in Fluorine-implanted Lithium Niobate"
ETH Nr. 18179, 2008
(Prof. Dr. P. Günter & Prof. Dr. M. Zgonik & Dr. G. Poberaj)
Working Place after ETH: University of Nova Gorica, Laboratory of Organic Matter Physics, Nova Gorica, Slovenia

59. Figi, Harry
"Single-Crystalline Organic Electro-Optic Microring Filters and Modulators"
ETH Nr. 18365, 2009
(Prof. Dr. P. Günter & Prof. Dr. M. Sigrist & Dr. M. Jazbinsek)
Working Place after ETH: University of Washington, USA

60. Kwon, Seong-Ji
"Bulk and Thin Film Crystals of Configurationally Locked Polyene for Integrated Optics and THz Wave Generation"
ETH Nr. 18498, 2009
(Prof. Dr. P. Günter & Prof. Dr. U. Suter & Dr. M. Jazbinsek)

61. Fabian Brunner
"Generation and Detection of Terahertz Pulses in the Organic Crystals OH1 and COANP"
ETH Nr. 18733, 2009
(Prof. Dr. B. Batlogg & Prof. Dr. P. Günter)

62. Marcel Stillhart
"DSTMS Crystals for THz Wave Generation, Detection and Spectroscopy"
ETH Nr. 18779, 2009
(Prof. Dr. B. Batlogg & Prof. Dr. P. Günter)

63. Manuel Koechlin
"Electro-optical Microresonators in Ion-sliced Lithium Niobate"
ETH Nr. 18811, 2009
(Prof. Dr. B. Batlogg & Prof. Dr. P. Günter)

64. Frederik Sulser
"Photonic Bandgap Devices in $LiNbO_3$"
(to be submitted in Spring 2010)

65. Gelu-Marius Rotaru
"Critical Behavior in Ferroelectrics"
(to be submitted in Spring 2010)

9.5 Diploma Theses in Physics

1. S. Giannini
   "Verwendung von Halbleitenden Elektroden zur Photoelektrolyse von Wasser"
   ETH Zürich, März 1977
   (Prof. Dr. P. Günter)

2. H. Rytz
   "Optische und Elektro-optische Eigenschaften von $PbHPO_4$"
   ETH Zürich, Februar 1978
   (Prof. Dr. P. Günter)

3. H.J. Hagenbucher
   "Optische und Elektro-optische Eigenschaften von $CsCuCl_3$"
   ETH Zürich, Februar 1978
   (Prof. Dr. P. Günter)

4. J.C. Baumert
   "Nichtlinear-optische Eigenschaften von $PbH_{1-x}D_xPO_4$"
   ETH Zürich, Oktober 1979
   (Prof. Dr. P. Günter)

5. R. Sanctuary
   "Optische Parametrische Verstärkung und Oszillation in $KNbO_3$"
   ETH Zürich, Oktober 1979
   (Prof. Dr. P. Günter)

6. M. Hug
   "Holographie und Kohärente Bildverstärkung in Elektro-optischen Kristallen"
   ETH Zürich, Oktober 1980
   (Prof. Dr. P. Günter)

7. E. Voit
   "Elastische Elasto-elektrische und Elasto-optische Eigenschaften von $KNbO_3$"
   ETH Zürich, Oktober 1983
   (Prof. Dr. P. Günter)

8. D. Jundt
   "Nichtlinear-optische Effekte in der Inkommensurablen und Kommensurablen Phase von Rb_2ZnCl_4"

ETH Zürich, Oktober 1984
(Prof. Dr. P. Günter)

9. P. Steiner
"Elektro-optische und Akusto-optische Eigenschaften von Rb_2ZnCl_4"
ETH Zürich, Dezember 1984
(Prof. Dr. P. Günter)

10. I.P. Gienal
"Räumliche Lichtmodulatoren: Aufbau und Physikalische Eigenschaften"
ETH Zürich, März 1985
(Prof. Dr. P. Günter)

11. O.M. Funariu
"Optische, Elektro-optische und mechanische Eigenschaften von Rb_2ZnCl_4"
ETH Zürich, März 1985
(Prof. Dr. P. Günter)

12. L. Ingold
"Optische Bistabilität in Photorefraktiven Materialien"
ETH Zürich, September 1985
(Prof. Dr. P. Günter)

13. J. Tscherry
"$KNbO_3$ Elektro-optische Deflektoren und Elastische/Piezoelektrische Eigenschaften"
ETH Zürich, September 1985
(Prof. Dr. P. Günter)

14. C. Bosshard
"Optische, Elektro-optische und Nichtlinear-optische Eigenschaften von 6-Cyclooctylamin-3-Nitropyridin (COANP) udn Methylaminopropanoat-2,4-dinitrophenyl (MAP)"
ETH Zürich, April 1986
(Prof. Dr. P. Günter)

15. K. Ensslin
"Optische und Elektro-optische Eigenschaften von Betainarsenat"
ETH Zürich, April 1986
(Prof. Dr. P. Günter)

16. M. Nussbaumer
”Integrierte Optik mit Molekularen Dünnfilmen”
ETH Zürich, Sommer 1986
(Prof. Dr. P. Günter)

17. Ch. Wirz
”Optische Frequenzverdoppelung von $Ga_{1-x}Al_xAs$ Laserdioden in $KNbO_3$”
ETH Zürich, Sommer 1986
(Prof. Dr. P. Günter)

18. R. Kesselring
”Räumliche Lichtmodulation unter Ausnützung der Anisotropen Selbstbeugung in Photorefraktiven $KNbO_3$-Kristallen”
ETH Zürich, September 1986
(Prof. Dr. P. Günter)

19. P. Amrhein
”Anisotrope Bragg-Beugung in Photorefraktiven $KNbO_3$-Kristallen”
ETH Zürich, September 1986
(Prof. Dr. P. Günter)

20. K. Sutter
”Linear optische Eigenschaften und Phasenanpassung von 6-Cyklooktylamin-3-Nitropyridin (COANP), 3-Methyl-N-Methyl-4-Aminomitrobenzol (MNMA), 6-Alpha-Methylbenzylamin-3-Nitropyriden (MBANP)”
ETH Zürich, September 1986
(Prof. Dr. P. Günter)

21. B. Rüegg
”Strukturelle und Nichtlinear-optische Eigenschaften von COANP, MNMA, DAN und MBANP”
ETH Zürich, September 1986
(Prof. Dr. P. Günter)

22. M. Ingold
”Photorefraktive Effekte in Cer-dotiertem $LiNBO_3$”
ETH Zürich, Oktober 1986
(Prof. Dr. P. Günter)

23. B. Senning
    "Optische Frequenzverdoppelung von Gepulsten $Ga_{1-x}Al_xAs$, Nd:YAG udn Styril-9 Farbstoff-Lasern in $KnBO_3$"
    ETH Zürich, April 1987
    (Prof. Dr. P. Günter)

24. G. Montemezzani
    "Optische und Thermische Fixierprozesse in $LiNbO_3$ und $KNbO_3$ Kristallen"
    ETH Zürich, April 1987
    (Prof. Dr. P. Günter)

25. P. Stillhard
    "Elektro-optische Eigenschaften von COANP Kristallen und Dünnfilmen"
    ETH Zürich, April 1987
    (Prof. Dr. P. Günter)

26. R. Staub
    "Untersuchung von KNb1-xTaxO_3 Kristallen für die integrierte Optik"
    ETH Zürich, April 1987
    (Prof. Dr. P. Günter)

27. G. Surmely
    "Linear und nichtlinear optische Eigenschaften von PNP, 2-(N-prolinol)-5-nitropyridin"
    ETH Zürich, März 1988
    (Prof. Dr. P. Günter)

28. A. Fischer
    "Elektrooptische und photorefraktive Eigenschaften von $Sr_{0.75}Ba_{0.25}Nb_2O_6$"
    ETH Zürich, März 1988
    (Prof. Dr. P. Günter)

29. J. Keller
    "Optische Bistabilität mit einem nematischen Flssigkristall und photorefraktiver Verstärkung in $KNbO_3$"
    ETH Zürich, Oktober 1988
    (Prof. Dr. P. Günter)

30. J. Arnold
    "Beeinflussung der Photorefraktiven Effekte in $KNbO_3$ durch äussere elek-

trische Felder"
ETH Zürich, Oktober 1988
(Prof. Dr. P. Günter)

31. L. Baraldi
"Nichtlinear-optische und elektro-optische Eigenschaften von 2-(N-prolinol)-5-nitropyridin (PNP)"
ETH Zürich, März 1989
(Prof. Dr. P. Günter)

32. M. Küpfer
"Nichtlinear optische Eigenschaften von 2-Docosylamino-5-Nitropyridin (DCANP) Wellenleiterschichten"
ETH Zürich, Oktober 1989
(Prof. Dr. P. Günter)

33. K. Totland
"Optisch bistabile Kavitäten mit flüssigkristallinen und photorefraktiven Elementen für Bildspeicher"
ETH Zürich, Oktober 1989
(Prof. Dr. P. Günter)

34. D. Fluck
"Photorefraktiver Effekt in stark reduzierten $KNbO_3$-Kristallen mit sehr hoher Leitfähigkeit"
ETH Zürich, Oktober 1989
(Prof. Dr. P. Günter)

35. G. Knoöpfle
"Feldinduzierte Harmonischenerzeugung an 2-Docosylamino-5-Nitropyridin (DCANP) und 2-Cyclooctylamino-5-Nitropyridin (COANP)"
ETH Zürich, Oktober 1989
(Prof. Dr. P. Günter)

36. W. Ferri
"Untersuchung des photorefraktiven Effektes in $KNbO_3$ unter gepulster Beleuchtung"
ETH Zürich, Oktober 1989
(Prof. Dr. P. Günter)

37. N. Saupper
    "Linear- und nichtlinear-optische Eigenschaften von N-(4-Nitro-2-pyridinyl)-phenylalaninol (NPPA)"
    ETH Zürich, April 1990
    (Prof. Dr. P. Günter)

38. R. Bopp
    "Intrakavität-Frequenzverdopplung und Intrakavität-Summenfrequenzerzeugung"
    ETH Zürich, April 1990
    (Prof. Dr. P. Günter)

39. H. Ammann
    "Photorefraktive Effekte in 2-Cyclooctylanimo-5-Nitropyridin (COANP) dotiert mit 7,7,8,8,-Tetracyanochinodimethan (TCNQ)"
    ETH Zürich, Oktober 1990
    (Prof. Dr. P. Günter)

40. M. Duelli
    "Aufbau und Untersuchung eines optischen Assoziativspeichers"
    ETH Zürich, Oktober 1990
    (Prof. Dr. P. Günter)

41. S. Pfändler
    "Photorefraktive und elektro-optische Eigenschaften von $Bi_4Ge_3O_{12}$ im ultravioletten Spektralbereich"
    ETH Zürich, Oktober 1990
    (Prof. Dr. P. Günter)

42. Ph. Prêtre
    "Nichtlinear optische Suszeptibilitäten organischer Moleküle und Kristalle bei der Wellenlänge 1,9 μ m"
    ETH Zürich, Oktober 1990
    (Prof. Dr. P. Günter)

43. M. Rossi
    "Nichtlinear optische Effekte in gepolten Seitenkettenpolymeren"
    ETH Zürich, Oktober 1990
    (Prof. Dr. P. Günter)

44. Th. Borer
"Nichtlineare Optik und Wellenleitung in 2-Docosylamino-5-Nitropyridin (DCANP) Langmuir-Blodgett (LB) Filmen"
ETH Zürich, Oktober 1990
(Prof. Dr. P. Günter)

45. U. Bertele
"Zeitaufgelöste Messungen des Aufbaus und Zerfalls photorefraktiver Gitter in reduzierten $KNbO_3$-Kristallen"
ETH Zürich, Oktober 1990
(Prof. Dr. P. Günter)

46. R. Schlesser
"Optische und elektro-optische Eigenschaften von 4'-Nitrobenzylidine-3-acetamino-4-methoxyaniline (MNBA)"
ETH Zürich, April 1991
(Prof. Dr. P. Günter)

47. Y. Schumacher
"Kristallzüchtung von COANP (2-Cyclooctylamino-5-nitropyridin) dotiert mit TCNQ (7,7,8,8-Tetracyanchinodimethan)"
ETH Zürich, April 1991
(Prof. Dr. P. Günter)

48. M. Bopp
"Photorefraktive Untersuchungen im ultravioletten Spektralbereich in undotierten $LiNbO_3$- und KD_2PO_4-Kristallen"
ETH Zürich, April 1991
(Prof. Dr. P. Günter)

49. H. Klepel
"Beobachtung optisch induzierter Kohärenztransferprozesse mittels zweidimensionaler Spektroskopie"
ETH Zürich, April 1991
(Prof. Dr. P. Günter)

50. W. Baumann
"Linear und nichtlinear optische Eigenschaften von 21'-Docosenyl-2-Amino-5-Nitropyridin-(VECANP)-Wellenleiterschichten"

ETH Zürich, Oktober 1991
(Prof. Dr. P. Günter)

51. C. Keller
"Assoziativ-Speicher mit sättigbarem Absorber"
ETH Zürich, Oktober 1991
(Prof. Dr. P. Günter)

52. B. Binder
"Frequenzverdoppelung in planaren $KNbO_3$ Wellenleitern"
ETH Zürich, Oktober 1991
(Prof. Dr. P. Günter)

53. St. Altner
"Kohärente Spinphänomene im Cäsium-Grundzustand"
ETH Zürich, Oktober 1991
(Prof. Dr. P. Günter)

54. J. Moll
"Optische Frequenzverdopplung in ein- und zweidimensionalen Wellenleitern in $KNbO_3$"
ETH Zürich, April 1992
(Prof. Dr. P. Günter)

55. P. Nosbaum
"Messung von nichtlinear optischen Suszeptibilitäten dritter Ordnung in organischen Molekülen und Kristallen bei der Wellenlänge 1.9 μm"
ETH Zürich, April 1992
(Prof. Dr. P. Günter)

56. L. Jaussi
"Photorefraktive Korrelatoren und ihre Anwendung im optischen Assoziativspeicher"
ETH Zürich, April 1992
(Prof. Dr. P. Günter)

57. Ch. Beyeler
"Photorefractive Effect in $KNbO_3$: Ni in the Red and Near Infrared"
ETH Zürich, April 1992
(Prof. Dr. P. Günter)

58. Ch. Pfammatter
"NMR Relaxationsverhalten des Mischkristallsystems $Rb_{1-x}(ND_4)_xD_2PO_4$"
ETH Zürich, April 1992
(Prof. Dr. P. Günter)

59. P. Rogin
"Photorefraktive Effekte in $KNbO_3$-Kristallen mit Band-zu- Band-Anregung durch ultraviolettes Licht"
ETH Zürich, Oktober 1992
(Prof. Dr. P. Günter)

60. M. J. Ewart
"Characterisation of Photoexcited Electrons in Reduced $KNbO_3$ with Pulsed Photorefractive Effects"
ETH Zürich, Oktober 1992
(Prof. Dr. P. Günter)

61. A. Bohren
"Elektro-Optische Eigenschaften Gepolter Seitenkettenpolymere"
ETH Zürich, Oktober 1992
(Prof. Dr. P. Günter)

62. S. Krucker
"Untersuchungen & Beschreibungen von Photoreftaktivem $KNbO_3$ im Roten & NIR"
ETH Zürich, Oktober 1992
(Prof. Dr. P. Günter)

63. A. Caprez
"Fixierung von Fourierhologrammen in $LiNbO_3$ und Anwendung auf optische Assoziativspeicher"
ETH Zürich, Oktober 1992
(Prof. Dr. P. Günter)

64. Th. Pliska
"Frequenzverdopplung von Laserdioden in $KNbO_3$Streifenwellenleitern"
ETH Zürich, Oktober 1992
(Prof. Dr. P. Günter)

65. J. Steinfort
”Scanning Force Microscopy of Cadmium Arachidate Langmuir-Blodgett Films with Submolecular Resolution”
ETH Zürich, Dezember 1992
(Prof. Dr. P. Günter)

66. F. Brechlin
”Photorefraktive Eigenschaften von ionenimplantierten $KNbO_3$-Wellenleitern”
ETH Zürich, März 1993
(Prof. Dr. P. Günter)

67. R. Ducret-Stich
”Optische und nichtlinear optische Eigenschaften von 4’-Dimethylamino-N-methyl-4-stilbazolium Tosylat (DAST)”
ETH Zürich, März 1993
(Prof. Dr. P. Günter)

68. D. Erni
”Hochtemperaturreduktion von $KNbO_3$ Kristallen für photorefraktive Anwendungen”
ETH Zürich, März 1993
(Prof. Dr. P. Günter)

69. P. Bernasconi
”Optical Communication Links with Photorefractive Crystals”
ETH Zürich, Oktober 1993
(Prof. Dr. P. Günter)

70. U. Meier
”Relaxationsmechanismen elektro-optischer Polymere”
ETH Zürich, Oktober 1993
(Prof. Dr. P. Günter)

71. B. Schmidt
”Kraftmikroskopische Untersuchungen an DCANP und VECANP Langmuir-Blodgett Filmen”
ETH Zürich, Oktober 1993
(Prof. Dr. P. Günter)

72. Ch. Seuret
    ”Dynamische Eigenschaften der Estermethyl-Rotation in teildeuteriertem poly(methylmethacrylat) (PMMA)”
    ETH Zürich, Oktober 1993
    (Prof. Dr. P. Günter)

73. J. Weiss
    ”Zweistrahlkopplung in photorefraktiven ionenimplantierten $KNbO_3$ Wellenleitern”
    ETH Zürich, Oktober 1993
    (Prof. Dr. P. Günter)

74. F. Mayer
    ”Frequenzverdopplungs- und Dämpfungseigenschaften von $KNbO_3$ Wellenleitern”
    ETH Zürich, März 1994
    (Prof. Dr. P. Günter)

75. L.-D. Pivéteau
    ”L'origine de la non-linéarité optique des verres dopes au Cd(S,Se)”
    ETH Zürich, März 1994
    (Prof. Dr. P. Günter)

76. M. Aufdenblatten
    ”Frequenzverdopplung in speziell orientierten $KNbO_3$ Streifenwellenleitern”
    ETH Zürich, Oktober 1994
    (Prof. Dr. P. Günter)

77. M. Friedrich
    ”Ferroelektrika und Kraftmikroskopie”
    ETH Zürich, Oktober 1994
    (Prof. Dr. P. Günter)

78. E. Gamper
    ”Elektro-optische Wellenleitermodulatoren auf $KNbO_3$”
    ETH Zürich, Oktober 1994
    (Prof. Dr. P. Günter)

79. B. Münch
”Wachstum und Charakterisierung von organischen nichtlinear optischen Dünnschichten”
ETH Zürich, Oktober 1994
(Prof. Dr. P. Günter)

80. R. Spreiter
”Bestimmung der nichtlinearen Brechungsindizes n2 in $KNbO_3$ und Molekularkristallen”
ETH Zürich, Oktober 1994
(Prof. Dr. P. Günter)

81. P. Wyss
”Bestimmung der Hyperpolarisierbarkeiten von Hydrazon Derivaten”
ETH Zürich, Oktober 1994
(Prof. Dr. P. Günter)

82. P. Blanc
”Nonlinear Transverse Patterns in a Undirectional Ring Resonator”
ETH Zürich, Oktober 1994
(Prof. Dr. P. Günter)

83. M. Boscacci
”Photorefractive and Absorptions Gratings with Interband Excitation”
ETH Zürich, März 1995
(Prof. Dr. P. Günter)

84. P. Cereghetti
”2D NMR Investigations on Partially Deuterated PMMA at Low Temperatures”
ETH Zürich, März 1996
(Prof. Dr. P. Günter)

85. A. Lötscher
”Photorefraktive räumliche Solitonen in $KNbO_3$”
ETH Zürich, März 1996
(Prof. Dr. P. Günter)

86. M. Lardelli
”Zeitaufgelste Untersuchung der Photorefraktion in eisen- und rhodium-

dotiertem $KNbO_3$"
ETH Zürich, Juli 1996
(Prof. Dr. P. Günter)

87. M. Pouchon
"Photorefraktiver Effekt in protonenimplantierten $KNbO_3$ Wellenleitern"
ETH Zürich, Juli 1996
(Prof. Dr. P. Günter)

88. M. Wiki
"Photorefraktive räumliche Solitonen in $KNbO_3$"
ETH Zürich, September 1996
(Prof. Dr. P. Günter)

89. M. Fierz
"Photorefractive Effect and Photoconductivity in 4-N,N-Dimethylamino-4'-N'-Methylstilbazolium Toluene-p-Sulfonate (DAST) Crystals"
ETH Zürich, Oktober 1996
(Prof. Dr. P. Günter)

90. W. Mazerant
"Bestimmung der nichtlinear optischen Eigenschaften 3. Ordnung von polaren Materialien"
ETH Zürich, Oktober 1996
(Prof. Dr. P. Günter)

91. St. Fischer
"Nichtlinear optische Eigenschaften 3. Ordnung von 4-N,N-Dimethylamino 4'-N'-Methyl-4-Stilbazolium Tosylat (DAST)"
ETH Zürich, September 1997
(Prof. Dr. P. Günter)

92. P. Messmer
"Zweidimensionale Photorefraktive Solitonen und Lichtinduzierte Wellenleiter in $KNbO_3$"
ETH Zürich, September 1997
(Prof. Dr. P. Günter)

93. H. Wyss
"Frequenzverdopplung von Laserdioden mit Wellenlängenstabilisierung durch

externe Bragg-Gitter"
ETH Zürich, März 1998
(Prof. Dr. P. Günter)

94. M. Achermann
"Ultrafast Electron Dynamics in Silver Films and Nanoparticles"
ETH Zürich, März 1998
(Prof. Dr. P. Günter)

95. M. Hübscher
"Numerische Simulation der Frequenzverdopplung in $KNbO_3$ Wellenleitern"
ETH Zürich, Oktober 1998
(Prof. Dr. P. Günter)

96. C. Tacchella
"Photorefractive Holographic Storage System with High Read-Out Frame-Rate"
ETH Zürich, Oktober 1998
(Prof. Dr. P. Günter)

97. M. Wintermantel
"Characterization of Electron-Transport and Polaron-Models in $Bi_{12}SiO_{20}$"
ETH Zürich, Oktober 1998
(Prof. Dr. P. Günter)

98. U. Fischer
"Hyperpolarisierbarkeiten 2. Ordnung und Kerreffekt von organischen Molekülen"
ETH Zürich, März 1999
(Prof. Dr. P. Günter)

99. P. Saieva
"Temperatur- und Wellenlängenabhängigkeit der Lumineszenz von organischen Materialien"
ETH Zürich, März 1999
(Prof. Dr. P. Günter)

100. Th. Schibli
"Stabilitätsanalyse von Faser- und Wellenleiter-Lasern mit hohen Repeti-

tionsraten"
ETH Zürich, März 1999
(Prof. Dr. P. Günter)

101. O. Schwarz
"Optical Correlation Based on Photorefractive Materials"
ETH Zürich, März 1999
(Prof. Dr. P. Günter)

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