Light-induced waveguides, waveguide arrays and switches in photorefractive LiTaO$_3$ and Sn$_2$P$_2$S$_6$

A dissertation submitted to

ETH ZURICH

for the degree of

DOCTOR OF SCIENCES

presented by

FLURIN JUVALTA

Dipl. Phys. ETH
born September 15, 1979
citizen of Zuoz (GR), Switzerland

accepted on the recommendation of

Prof. Dr. P. Günter, examiner
Prof. Dr. G. Montemezzani, co-examiner

2008
Contents

Abstract v
Zusammenfassung vii

1 Introduction 1
   1.1 Nonlinear optics . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 2
   1.2 Photorefractive effect . . . . . . . . . . . . . . . . . . . . . . . . . . . . 3
   1.3 The interband photorefractive effect . . . . . . . . . . . . . . . . . . . . 10
   1.4 Optical waveguides . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 13
   1.5 Waveguide arrays . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 16

2 Electro-optical properties of LiTaO$_3$ 21
   2.1 Introduction . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 21
   2.2 Lithium Tantalate . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 23
   2.3 Refractive indices . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 24
   2.4 Electro-optic coefficients . . . . . . . . . . . . . . . . . . . . . . . . . . . 27
   2.5 Conclusions . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 31

3 Light-induced waveguides in Mg-doped near-stoichiometric LiTaO$_3$ 33
   3.1 Introduction . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 33
   3.2 Experimental . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 34
      3.2.1 Growth and preparation of Mg-doped near-stoichiometric LiTaO$_3$ . 34
      3.2.2 Light induced waveguides . . . . . . . . . . . . . . . . . . . . . . . . 35
      3.2.3 Holographic characterization . . . . . . . . . . . . . . . . . . . . . . . 37
   3.3 Results and discussion . . . . . . . . . . . . . . . . . . . . . . . . . . . . 38
      3.3.1 Depth profile of the interband grating . . . . . . . . . . . . . . . . . . 38
      3.3.2 Waveguide profile . . . . . . . . . . . . . . . . . . . . . . . . . . . . 39
      3.3.3 Waveguide dynamics . . . . . . . . . . . . . . . . . . . . . . . . . . . 41
      3.3.4 Quasi-fixing of waveguides and double waveguides . . . . . . . . . . . 43
   3.4 Conclusions . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 45
## CONTENTS

4 Light deflection through photoinduced waveguides 47

4.1 Introduction ................................................. 47
4.2 Experimental technique ..................................... 49
4.3 Beam splitting and deflection upon field removal ............... 51
4.4 Modeling of beam propagation .............................. 52
   4.4.1 Sr$_x$Ba$_{1-x}$Nb$_2$O$_6$ ............................... 52
   4.4.2 LiTaO$_3$ ........................................... 57
4.5 Waveguide anti-waveguide alternation ......................... 60
4.6 Conclusions ............................................... 62

5 Light-induced waveguides and waveguide arrays in Sn$_2$P$_2$S$_6$ 63

5.1 Introduction ............................................... 63
5.2 Light induced waveguides .................................. 65
5.3 Light induced waveguide arrays ............................ 67
5.4 Conclusions ............................................... 70

6 Main conclusions and outlook 71

A Materials 73
   A.1 Lithium Tantalate (LiTaO$_3$) ............................ 73
   A.2 Tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$) .................. 75

B Ion implantation in Sn$_2$P$_2$S$_6$ 79
   B.1 Ion Implantation ........................................ 79
   B.2 Barrier coupling method ................................. 81

Bibliography 95

List of publications 96

Acknowledgments 98

Curriculum vitae 99
Abstract

All-optical switching, in which one beam of light modulates, or switches, another, is one of the holy grails of modern photonics. It is a necessary technology for optical computing and for optical networks, where optical information is transferred from one location to another without conversion to electronic form. The dynamic control of light by light is the main focus of this thesis. Waveguides which confine the light in one or two dimensions are the most fundamental building blocks of integrated optical circuits. Different dynamic waveguide structures induced by lateral illumination of electro-optic crystals are generated and investigated. The materials under investigation are magnesium doped near-stoichiometric lithium tantalate (SLT:Mg) and nominally pure tin hypophosphosphate Sn$_2$P$_2$S$_6$ (SPS). In SLT:Mg the waveguides are controlled by deep UV light at $\lambda = 257$ nm and in pure Sn$_2$P$_2$S$_6$ in the visible at $\lambda = 514$ nm. The waveguide structures are induced a few ten micrometers beneath the surface of the crystals by the interband photorefractive effect, i.e. interband photoexcitation and drift or diffusion charge transport mechanism.

Using the interband photorefractive effect is much more effective in terms of use of the incident photons with respect to the conventional impurity-dominated effect, leading to a much faster response. In Sn$_2$P$_2$S$_6$ we measured the fastest formation of light induced waveguides in the visible up to present. The recording times are below 200 $\mu$s for intensities above 0.1 W/cm$^2$. Furthermore, structures written by interband light are very robust against sub-bandgap illumination. Thus, the waveguide structures can be probed nondestructively in the transverse direction with a beam at a longer wavelength.

Interband photorefraction was investigated in Mg-doped near-stoichiometric LiTaO$_3$ at the deep UV wavelength $\lambda = 257$ nm and Bragg diffraction experiments were performed. The formation of two distinct grating components, which are most likely associated with free carriers on the one hand and deep traps on the other hand, was observed in depth-resolved measurements. We demonstrated that the deep trap component can be quasi-fixed and is very robust against readout in the visible with lifetimes in the order of weeks.

We have shown that without sufficient background illumination also the light induced
waveguides could be quasi-fixed in Mg-doped near-stoichiometric LiTaO₃. The formation of a double waveguide upon removal of the applied electric field needed for waveguide formation was found after re-illuminating the unbiased crystal. This is based on a screening of the remaining space charge electric field. Deflection and modulation of the guided light is realized by alternately writing a waveguide and a double waveguide. Numerical simulations of the refractive index evolution and beam propagation show a good agreement with the observations.

Waveguide arrays or photonic lattices consist of a periodic arrangement of waveguides in one or two dimensions. The diffraction properties of such systems can be engineered opening up new possibilities for controlling the flow of light, which would have been impossible in the bulk. By interfering two light beams, dynamic waveguide arrays are generated in Sn₂P₂S₆ at λ = 514 nm with reconfiguration times below one millisecond. An applied electric field increases the refractive index modulation and induces a spatial shift to the array.

Lithium tantalate is a very promising material for applications at UV wavelengths. Since laser sources in the UV are becoming more and more available, there is an increasing interest in such short wavelength applications. Refractive indices and electro-optic coefficients \( r_{13} \) and \( r_{33} \) of near-stoichiometric and congruent lithium tantalate were measured from \( \lambda = 633 \text{ nm} \) down to the deep UV wavelength \( \lambda = 275 \text{ nm} \). Electro-optical coefficients of up to \( r_{33} = 52 \pm 1 \text{ pm/V} \) have been determined. We propose a simplified two-oscillator polarization potential model that very well describes our data near the absorption edge.
Zusammenfassung


Verglichen mit dem konventionellen photorefraktiven Effekt ist der interband photorefraktive Effekt viel effizienter hinsichtlich der benutzten Photonenergie, was zu einer schnelleren Antwort führt. In Sn$_2$P$_2$S$_6$ haben wir die bis heute schnellste Erzeugung lichtinduzierter Wellenleiter im sichtbaren Bereich gemessen mit Schreibzeiten kleiner als 200 $\mu$s für Intensitäten über 0.1 W/cm$^2$. Des Weiteren sind mit interband Licht geschriebene Strukturen sehr robust gegenüber der Beleuchtung durch Licht dessen Photonenergie kleiner ist als die Bandlücke des Materials. Deshalb werden bei einer Untersuchung mit Licht längerer Wellenlänge solche Wellenleiterstrukturen nicht beeinträchtigt.

Durch die Interferenz zweier Lichtstrahlen der tiefen UV Wellenlänge $\lambda = 257$ nm haben wir photorefraktive Gitter in Mg-dottiertem, fast-stoichiometrischem LiTaO$_3$ geschrieben und haben Bragg-Beugung an diesen Gittern untersucht. Messungen in unterschiedlichen Tiefen unterhalb der Kristalloberfläche zeigten, dass sich zwei unterschiedliche Gitterkomponenten bilden, welche höchstwahrscheinlich den freien Ladungen auf der einen Seite und den tiefer gelegenen Störstellen auf der anderen Seite zugeord-
ZUSAMMENFASSUNG


Lithium Tantalat ist ein vielversprechendes Material für Anwendungen im ultraviolett Wellenlängenbereich. Aufgrund der steigenden Verfügbarkeit von UV Lichtquellen nimmt auch das Interesse an solchen kurzwelligen Anwendungen zu. Deshalb haben wir die Brechungsindizes und die elektro-optische Koeffizienten $r_{13}$ und $r_{33}$ von stoichiometrischem und kongruentem Lithium Tantalat im UV gemessen. Elektro-optische Koeffizienten von bis zu $r_{33} = 52 \pm 1$ pm/V wurden ermittelt. Wir haben ein vereinfachtes Zwei-Oszillatoren Polarisations-Potenzial Modell entwickelt, welches unsere Daten nahe der Absorptionskante im UV sehr gut wiedergibt.
Chapter 1

Introduction

Photonics is a key driving technology of the 21st century [1] and the basis of the present-day internet and long distance communications. Photonics is the science of generating, controlling, and detecting light, particularly in the visible and near infra-red spectrum range, but also extending to the extreme ultraviolet and long-wave infrared including the terahertz range. The term “ photonics” is used in analogy to electronics with the difference that photons instead of electrons build the smallest unit for transmission, processing, display, and storing of information. Photonics combines various disciplines such as quantum electronics, quantum optics, nonlinear optics, electro-optics, integrated optics, fiber optics, laser optics, optoelectronics, optical storage, etc.

Especially for communication networks, devices, and associated technologies photonics and optics are indispensable as they meet today’s and future requirements for ultrahigh bandwidth. All-optical switching, dynamic waveguiding and wave manipulation, and the ability to micro- and nanostructure electro-optical and nonlinear optical materials are very important tools and techniques to improve device performances.

The main aim of this thesis is the investigation of a dynamic concept for the fabrication of reconfigurable optical waveguide structures for integrated optics. It is shown that using the interband photorefractive effect, fast dynamic waveguide structures can be induced in different electro-optic materials, namely near-stoichiometric lithium tantalate (LiTaO$_3$) and tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$). These two materials are interesting for this kind of structures from a different point of view. LiTaO$_3$ is a promising material for applications in the deep UV wavelength range. Interband photorefraction was investigated and demonstrated in the deep UV at $\lambda_{UV} = 257$ nm. In addition, precise measurements and modeling of the electro-optic coefficient and the refractive index of LiTaO$_3$ down to $\lambda = 275$ nm are reported in Chapter 2. Up to present, Sn$_2$P$_2$S$_6$ is the only known material that allows for interband photorefraction in the visible, therefore elements like spatial light modulators (SLMs) can be used. It offers high photorefrac-
tive efficiency in the infrared up to the telecommunication wavelength 1.55 \( \mu m \) [2–4] and a large electro-optic coefficient \((r_{111} = 174 \text{ pm/V at } 633 \text{ nm}[5])\). Furthermore, the interband photorefractive response of \( \text{Sn}_2\text{P}_2\text{S}_6 \) is very fast [6].

This chapter gives a brief introduction to nonlinear optics, photorefractive optics, interband photorefractive effects, optical waveguides and waveguide arrays.

### 1.1 Nonlinear optics

In this section, basic concepts and models of nonlinear optics, which are important for this work, are introduced.

#### Linear and nonlinear optical effects

Linear and nonlinear optical effects can be described in terms of the linear polarization \( P^L \) and the nonlinear polarizations \( P^{NL} \) of the macroscopic material polarization \( P \) induced in a nonlinear medium by an external electric field \( E \) as

\[
P_i = P_i^0 + P_i^L + P_i^{NL} = P_i^0 + \varepsilon_0 \chi^{(1)}_{ij} E_j + \varepsilon_0 \chi^{(2)}_{ijk} E_j E_k + \varepsilon_0 \chi^{(3)}_{ijkl} E_j E_k E_l + \ldots
\]

(1.1)

using the Einstein convention for summation over common indices (with \( i, j, k, l = 1, 2, 3 \)) and with \( P^0 \) the spontaneous polarization, \( \varepsilon_0 \) the vacuum permittivity, \( \chi^{(1)} \) the linear or first-order, \( \chi^{(2)} \) the second-order, and \( \chi^{(3)} \) the third-order susceptibility. For symmetry reasons, the odd-order susceptibilities are present in any material, whereas the even-order ones only occur in noncentrosymmetric materials (in the dipole approximation). The susceptibility tensors \( \chi^{(n)} \) contain all the information about the macroscopic optical properties of the respective material.

#### The electro-optic effect

If the external electric field \( E \) is composed by the field \( E^\omega \) of an optical wave and a static electric field \( E^0 \), the nonlinear polarization \( P_i^{NL} \) in Eq. (1.1) will include terms of the same frequency as \( E^\omega \), the first such terms being

\[
P_i^{NL} = 2\varepsilon_0 \chi^{(2)}_{ijk} E_j^\omega E_k^0 + 3\varepsilon_0 \chi^{(3)}_{ijkl} E_j^\omega E_k^0 E_l^0 + \ldots
\]

(1.2)

This polarization depends linearly on the optical field \( E^\omega \) and can therefore be considered as a part of the linear polarization \( P^L \) with modified \( \chi^{(1)} \). This can be expressed as a
1.2 Photorefractive effect

change of the refractive index \((\chi^{(1)} = n^2 - 1)\), which is best written as the change of the
tensor \(\varepsilon^{-1} = n^{-2}\) in the form

\[
\Delta \left( \frac{1}{n^2} \right)_{ij} = r_{ijk} E^0_k + R_{ijkl} E^0_k E^0_l + \ldots,
\]

(1.3)

where \(r_{ijk}\) is the linear electro-optical tensor, related to \(\chi^{(2)}_{ijk}\), and \(R_{ijkl}\) is the quadratic
 electro-optical tensor, which is related to \(\chi^{(3)}_{ijkl}\). Contrarily to nonlinear optical effects,
the electro-optical effect does not depend on the intensity of the optical wave \(E^\omega\), but
only on the magnitude and the direction of the static field \(E^0\). For noncentrosymmetric
electro-optic materials and applied fields in the order of kV/cm, the quadratic effect is
usually negligible compared to the linear effect. Note that the present work concerns only
noncentrosymmetric materials and as a consequence just the linear electro-optic effect
will be considered in the following.

An effective coefficient \(r_{\text{eff}}\) can be calculated depending on the crystal symmetry
and the experimental geometry for simplifying calculations of \(\Delta n\). Since the electro-
optic changes of the refractive indices are very small, in the order of \(10^{-4}\) for practically
applicable fields, they can be expressed in a linear approximation as

\[
\Delta n = - \frac{1}{2} n^2 r_{\text{eff}} E^0.
\]

(1.4)

Now the refractive index \(n\) is a scalar value representing the refractive index in the specific
geometry and \(\Delta n\) the change of that index.

1.2 Photorefractive effect

The photorefractive effect was discovered by Ashkin and coworkers in 1966 [7], in the
beginning as an unwelcome beam distortion: they noticed, that a focused and intense
blue or green laser beam produced a local and temporary change of the optical properties
of crystalline ferroelectrics, in their experiment lithium niobate (LiNbO\(_3\)) and lithium
tantalate (LiTaO\(_3\)).

Initially, this effect was referred to as “optical damage”, as it limited the use of such
crystals with large electro-optic and nonlinear optic coefficients. Cause of the “damage”
were changes of the refractive index, induced by light, which gave rise to decollimation
and scattering of laser beams. This optically induced change of the refractive index of
electro-optic crystals due to charge transport is now referred to in literature as the “pho-
torefractive effect” [8,9]. The photorefractive effect can show very high light sensitivity,
permitting the observation of such effects at very low light powers (< µW). On the other
hand, detailed understanding of the photorefractive effects helped developing materials
with negligible light sensitivity, which can be then used without “damage” for electro-optics and nonlinear optics. Such materials are also of benefit for the non-conventional (band-to-band or interband) regime, which is exploited in this work.

In the following the basic theory of the photorefractive effect, starting from the band-transport model will be presented. The special case of direct band-to-band photoexcitation will be considered in section 1.3.

Physical Processes Involved in the Photorefractive Effect

The photorefractive effect can be described by the following four processes:

- Generation of charge carriers through photo-excitation. These carriers are either electrons excited to the conduction band or holes excited to the valence band. In most materials it is a combination of both, but one is dominating.

- Transport of the excited, mobile charges due to diffusion or electric field forces (drift). Additionally, the bulk photogalvanic effect may contribute to the charge displacement.

- Recombination of the mobile charge carriers into trap states, e.g. defects, impurities, and self trapping. The resulting charge distribution is according to the light illumination, since charges recombined in illuminated regions can be re-excited, while charges recombined in dark regions remain there.

- Due to this charge redistribution, a space charge electric field is generated which leads to a change of the refractive index via the electro-optic effect, described in section 1.1.

Figure 1.1 illustrates the involved processes and steps for a particular case of electron diffusion. In Fig. 1.2 a simplified band-scheme and the most important processes for photo-excitation, displacement, recombination, and trapping of charge carriers are depicted.

In the following, we introduce the most important mathematical models used to describe the above mentioned physical processes.

Conventional Model

The so-called conventional model was elaborated by Kukhtarev et al. [10–12] considering only photo-excitation and recombination of one species of charge carriers between a single donor level and the corresponding conduction band. This single-level band scheme, along with the involved physical mechanisms, is depicted in Fig. 1.3. Such a simplification is
1.2 Photorefractive effect

The involved processes can be described by the following set of equations

\[ \frac{\partial N_D^+}{\partial t} = (s_e I + \beta_e)(N_D - N_D^+) - \gamma_e n N_D^+ \]  
\[ \frac{\partial n}{\partial t} = \frac{\partial N_D^+}{\partial t} + \frac{1}{e} \nabla J_e \]  
\[ J_e = e n \mu_e E + k_B T \mu_e \nabla n + e s_e I (N_D - N_D^+) L_{ph} \]  
\[ \nabla \cdot E_{sc} = \frac{e}{\varepsilon_0 \varepsilon_{eff}} (N_D^+ - n - N_A), \]

where the photoexcited charges are assumed to be electrons. The symbols in the above equations are:

- \( n \) the free electron concentration in the conduction band;
- \( N_D \) the donor concentration;
- \( N_D^+ \) the concentration of ionized donors;

Justifiable for light illumination with photon energies much smaller than the band-gap energy. For laser light with higher photon energies, additional impurity levels have to be taken into account.

Figure 1.1: Mechanisms involved in the photorefractive effect, here illustrated for the case of electron charge transport. \( \Lambda \) is the grating constant and \( \Phi \) is the spatial shift between light intensity and refractive index modulation. For charge transport dominated by diffusion one gets \( \Phi = \Lambda/4 \).
• $N_A$ the concentration of ionized donors (acceptors) in the dark;
• $J_e$ the electron current density;
• $E$ the total electric field;
• $E_{sc}$ the space charge field;
• $I$ the light intensity;
• $L_{ph}$ the photogalvanic transport length vector;
• $s_e$ the photoionization constant for electrons;
• $\beta_e$ the thermal (dark) generation rate for electrons;
• $\gamma_e$ the recombination constant for electrons;
• $\mu_e$ the electron mobility tensor;
• $\varepsilon_0$ the vacuum dielectric permittivity;
• $\varepsilon_{\text{eff}}$ the effective dielectric constant for the chosen configuration [13];
• $e$ the absolute value of the elementary charge;
• $k_B$ the Boltzmann constant;
• $T$ the absolute temperature.

Equation (1.5) is the rate equation for the concentration of the ionized donors. The first term describes the photoionization process ($s_e I$) and the thermal excitation of electrons from the donor level. The second term takes into account the recombination of the electrons ($\gamma_e$) into traps, in our case ionized donors. The second equation (1.6) is the continuity equation for the electron density. The additional term with respect to Eq. (1.5) describes the divergence of the electron current density. Eq. (1.7) describes the different contributions to the electron current density. The first term gives the drift current in the total electric field $E = E_{sc} + E_0$, where $E_0$ is the applied field. The second term describes the diffusion process of the electrons generated by the electron concentration gradient. Thus, this term only gives a contribution for inhomogenous illumination. The last term gives the photogalvanic current, if present. The last equation (1.8) is the Poisson equation for the electric field. It describes the spatially modulated part of the electric field generated by the nonuniform distribution of the charge carriers in the crystal. These four equations are valid in this form only for isotropic photoexcitation, i.e., if $s_e$ is independent on the light polarization and intensity, or if the intensity and polarization of the exciting light wave is invariant, i.e., constant, with respect to its propagation direction, like in the case of a plane wave.
1.2 Photorefractive effect

Figure 1.2: Simplified band scheme of possible charge transitions in a photorefractive material with the concentration of non-ionized donors $N_D - N_D^+$ and traps $N_D^+$. Excitation of charge carriers may occur via band-to-band (with a generation rate $s_{dir}I$), trap-to-band ($s_eI$), and band-to-trap transitions ($s_hI$). Excited charges are displaced by diffusion and drift. They can recombine through band-to-band electron-hole recombination (with a recombination rate $\gamma_{dir}$), or be trapped in mid-bandgap levels ($\gamma_e$ and $\gamma_h$). The arrows show the movements of the electrons and thermal excitation is not depicted.

Figure 1.3: Single level band scheme in the conventional model at low intensities. An electron is photoexcited to the conduction band from a mid band-gap level. Diffusion or drift result in a displacement of the electron. Near a trap center, the electron can be retrapped.
Photorefractive index gratings

Homogeneously irradiating a photorefractive material, will not generate a space charge field or refractive index grating, but only change some of the bulk material properties like the conductivity or the absorption. In contrast, for an inhomogeneous illumination the charges are locally redistributed and will result in a space charge field. An effective way to produce an intensity pattern is given by the interference of two coherent plane waves. Considering two plane waves with wave vectors $k_1$ and $k_2$ and electric field amplitudes $A_1$ and $A_2$ respectively, we obtain a sinusoidal intensity pattern of the form

$$I(x) = I_0 + I_1 \exp(iK \cdot x),$$

(1.9)

where $K = k_1 - k_2$ is the grating vector, $x$ is the position vector, $I_0 = \frac{1}{2} \sqrt{\varepsilon_0/\mu_0(|A_1|^2 + |A_2|^2)}$ the mean intensity and $I_1 = \sqrt{\varepsilon_0/\mu_0(|A_1||A_2|)}$ the intensity modulation. Assuming small modulation depth $m := I_1/I_0$, we can follow the usual linearization procedure and we obtain for the quantities $n$, $N_D^+$, $E$, and $J_e$ a sinusoidal expression of the type

$$F(x) = F_0 + F_1 \exp(iK \cdot x),$$

(1.10)

where only the real part corresponds to the value of the physical property. Since $F_1$ is a complex value, the modulation of the different quantities can exhibit a phase shift $\phi$ with respect to the light fringes. This implies a spatial shift of the maxima by $\phi/K$. The most important quantity for describing the photorefractive effect is the electric field and we will denote in the following the amplitude of the first spatial Fourier component ($E_1$) of the space charge field with $E_{sc}$. The exact amplitude of $E_{sc}$ for the conventional model will be given later in this section and for interband photorefraction in the next section.

In an electro-optic material the presence of a periodic space charge field $E_{sc}$ generates a refractive index grating that can be investigated by diffraction experiments of a third plane wave, eventually at a different wavelength. For non-centrosymmetric materials that exhibit the linear electro-optic effect (Pockels effect), the change of the dielectric permittivity tensor is defined as

$$\Delta\varepsilon^{-1} = r_{ef}(K)E_{sc},$$

(1.11)

where $\varepsilon$ is the dielectric permittivity tensor in the material at light wavelength and $r_{ef}(K)$ is a $3 \times 3$ matrix describing the electro-optic effect which considers the unclamped electro-optic tensor and the additional contributions from the piezo-electric effect [14]. Note that compared to Eq. (1.3) the spatial dispersion, i.e. the $K$-vector dependence, of the electro-optic tensor is considered. Since in Eq. (1.11), and alike in the following formula for $\Delta n$, the oscillating spatial term is not included, the components on the left-hand side of this equation are the complex first spatial Fourier components.
For a certain geometry and light polarization, the amplitude of the spatially periodic refractive index change $\Delta n$ can be expressed as

$$\Delta n \approx -\frac{1}{2}n^3 r_{\text{eff}} E_{\text{sc}},$$

(1.12)

where $n$ is the refractive index seen by the beam, and $r_{\text{eff}}$ is the effective electro-optic coefficient relevant for this geometry. In the following section the magnitude of the space charge field is considered in detail.

**Space charge field for sinusoidal illumination**

A solution of Eqs. (1.5)–(1.8) can be found in the case where the material is illuminated with a periodic intensity distribution as described by Eq. (1.9).

The space charge field is parallel to the grating vector and its amplitude is $|E_{\text{sc}}|$. The complex amplitude of the space charge field $E_{\text{sc}}$ in case of small modulation depth $m$ is given by [11]

$$E_{\text{sc}} = -im \frac{E_q(E_D - iE_0)}{E_q + E_D - iE_0}$$

(1.13)

where $i$ is the imaginary unit. For a better physical interpretation of $E_{\text{sc}}$ we have introduced the diffusion field $E_D$ and the trap limited field $E_q$. The first is defined as

$$E_D = \frac{k_B T}{e} K,$$

(1.14)

and corresponds to the field amplitude of a sinusoidal electric field that exactly counteracts the effect of the charge diffusion process. The trap limited field is

$$E_q = \frac{e}{\varepsilon_{\text{eff}} \varepsilon_0 K} N_{\text{eff}}$$

(1.15)

and gives a limit for the maximum $E_{\text{sc}}$ that can be generated with the available traps. In Eq. (1.15) $N_{\text{eff}}$ is the effective trap density defined as

$$N_{\text{eff}} = \frac{N_{D0}^+(N_D - N_{D0}^+)}{N_D}$$

(1.16)

where $N_{D0}^+ = n_0 + N_A$ is the spatial average of ionized donors with illumination switched on. Although $n_0$, the spatial average of electrons in the conduction band when the light is on, is dependent on the intensity $I_0$ of the light, $n_0$ is in most cases much smaller than $N_A$, so that $N_{D0}^+ \simeq N_A$. $N_{\text{eff}}$ and $E_q$ are independent of the light intensity in the conventional model.

In the case no electric field is applied, Eq. (1.13) can be simplified to

$$E_{\text{sc}} = -im \frac{E_q E_D}{E_q + E_D}$$

(1.17)
An important conclusion can be deduced from Eq. (1.17) which shows that if one of the two fields $E_q$ or $E_D$ is much smaller than the other, the total space-charge field is limited by this field. These limiting cases can be observed experimentally by varying the grating constant $K$. Furthermore, we see from Eq. (1.17) that without externally applied field the space-charge field is purely imaginary and the grating is shifted by $\phi_0 = \pi/2$ compared to the illumination grating.

For large grating spacing $\Lambda \gg 1\mu m$ the build-up time of such photorefractive gratings is defined by the dielectric time constant

$$\tau_D = \frac{\varepsilon_0 \varepsilon_{\text{eff}}}{\sigma} = \frac{\varepsilon_0 \varepsilon_{\text{eff}}}{\varepsilon \mu_c n_0}. \quad (1.18)$$

Here $\sigma = \varepsilon \mu_c n_0$ is the total conductivity of the material and depends on the light intensity through $n_0$. Solving Eq. (1.5) considering only the spatial average values gives $n_0 \propto I_0$ and therefore

$$\tau_D \propto \frac{1}{I_0} \quad (1.19)$$

for the conventional photorefractive effect. The correction factor in the build-up time for smaller grating spacing is in the order of 1 and is independent on the light intensity.

### 1.3 The interband photorefractive effect

The conventional model only considers photo-excitation of charges from one midgap impurity level to one of the conduction bands. Accordingly, the absorption constant is small and the excitation process rather slow (milliseconds to seconds). As shown in Fig. 1.2, other transitions are possible as well. Under light illumination with photon energies larger than the band gap, band-to-band transitions occur and dominate over impurity-to-band transitions, particularly when the impurity concentration is low. Because of a much higher absorption constant in this regime, many more free charge carriers are produced, thus leading to a much faster excitation process. This effect is known as the “interband photorefractive effect”.

### Basic equations

The most common set of equations describing the charge dynamics is given in [15], and includes one trap level in addition to the direct band-to-band excitation. No thermal excitations and no photogalvanic current are considered. The following equations describe the processes illustrated in Fig. 1.2:
1.3 The interband photorefractive effect

\[ \frac{\partial N_D^+}{\partial t} = s_e I(N_D - N_D^+) + \gamma_h p(N_D - N_D^+) - \gamma_e n N_D^+ \] (1.20)

\[ \frac{\partial n}{\partial t} = s_{\text{dir}} I(N_V - p) + s_e I(N_D - N_D^+) - \gamma_e n N_D^+ - \] \[ -\gamma_{\text{dir}} np + \frac{1}{e} \nabla J_e \] (1.21)

\[ \frac{\partial p}{\partial t} = s_{\text{dir}} I(N_V - p) + s_h I(N_V - p) N_D^+ - \gamma_h p(N_D - N_D^+) - \] \[ -\gamma_{\text{dir}} np - \frac{1}{e} \nabla J_h \] (1.22)

\[ J_e = e n \mu_e E_{\text{sc}} + k_B T \mu_e \nabla n \] (1.23)

\[ J_h = e p \mu_h E_{\text{sc}} - k_B T \mu_h \nabla p \] (1.24)

\[ \nabla E_{\text{sc}} = \frac{e}{\varepsilon_0 \varepsilon_{\text{eff}}}(N_D^+ + p - n - N_A) \] (1.25)

The symbols have the same meaning as in Eqs. (1.5–1.8) whereas the newly added ones are defined as follows: \( p \) is the free hole concentration in the valence band, \( N_V \) is the density of electrons close enough to the top of the valence band to be photoexcited, \( J_h \) the hole current density, \( s_{\text{dir}} \) the photoexcitation constant for direct band-to-band phototransitions, \( \gamma_h \) the recombination constant for the hole-donor interaction, and \( \mu_h \) the hole mobility tensor.

The mathematical complexity of this set of equations is already for one impurity level so high that no closed solution has yet been found. Analytic solutions for \( E_{\text{sc}} \) were found by applying some simplifications [15, 16]. Here we present only the solution for pure interband regime, i.e. without considering any trap level \((N_D = N_D^+ = N_A = 0)\). This limit well describes the effect for high light intensities, where the contribution from trap levels becomes negligible. In this limitation the hole and electron densities are equal and are given by:

\[ p_0 = n_0 = \sqrt{\frac{g I_0}{\gamma_{\text{dir}}}}, \] (1.26)

where the constant \( g \) is related to the absorption constant \( \alpha_{\text{dir}} \) for band-to-band excitation divided by the photon energy \( h \nu \):

\[ g \equiv s_{\text{dir}} N_v = \alpha_{\text{dir}}/h \nu. \] (1.27)

The resulting amplitude of the space charge field \( E_{\text{sc}} \) for a continuous sinusoidal illumination with a small light intensity modulation \( m \) is given by [15]

\[ E_{\text{sc}} = -i m \frac{E_{\text{ef}}[E_D(E_Rh - E_Re) - i E_0(E_Rh + E_Re)]}{(E_D + E_Re + E_Rh)(E_D + 2E_{\text{ef}}) + E_0^2 + i E_0(E_Re - E_Rh)} \] (1.28)
where $E_D = K k_B T / e$ represents the diffusion field, $E_{qf}$ the free carrier-limited field

$$E_{qf} = \frac{e}{\varepsilon_{\text{eff}} \varepsilon_0 K} \sqrt{\frac{g I_0}{\gamma_{\text{dir}}}},$$

(1.29)

and $E_{Re,Rh}$ the electron (hole) recombination fields

$$E_{Re,Rh} = \frac{1}{K \mu_{e,h}} \sqrt{g I_0 \gamma_{\text{dir}}},$$

(1.30)

The recombination field can be interpreted as the average electric field needed to drift one electron or hole by a distance $K^{-1} = \Lambda / 2\pi$ before a direct band-to-band recombination takes place.

As in the conventional photorefractive effect, in the case of pure diffusion charge transport mechanism ($E_0 = 0$) the space-charge field is purely imaginary, i.e. $\pi/2$ phase shifted with respect to the light fringes. Eq. (1.28) is a little bit more complex than Eq. (1.13), valid for the conventional single level model. Different regimes dominated by one of the fields $E_{qf}$, $E_D$, $E_{Rh}$, and $E_{Re}$ are possible.

In the interband photorefractive effect the steady state of the space charge field depends not only on the grating spacing $\Lambda$ but also on the light intensity $I_0$. For high intensities ($E_{qf} \gg E_D$) and small grating spacings $\Lambda$ the $E_{sc}$ will grow proportional to $\sqrt{I_0}$.

The dynamics of the build-up is given by a double exponential function [16], and the build-up time constants are proportional to

$$\tau_{ib} \propto \frac{1}{\sqrt{I_0}}$$

(1.31)

in the pure interband regime. The different intensity dependence of the build-up time for conventional and interband effects, compared with Eq. (1.19), can be used to easily identify different regimes.

For conventional photorefractive crystals, oxde ferroelectrics, interband photorefractive experiments must be performed with ultraviolet light, since the bandgap of these materials lies in that energy range, e.g. LiTaO$_3$ has a bandgap of 4.8 eV. Sn$_2$P$_2$S$_6$ has a smaller bandgap of 2.3 eV and thus visible laser lines, such as that of an Argon ion laser, can be used. More characteristic properties of LiTaO$_3$ and Sn$_2$P$_2$S$_6$ are given in Appendix A.
1.4 Optical waveguides

This section summarizes the basic principles of optical waveguiding, considering a planar
dielectric waveguide [17,18].

Basics and theory

A waveguide is defined as any structure capable of guiding electromagnetic radiation in a
direction parallel to its axis, while substantially confining the radiation to a region within
and adjacent to its surfaces or interfaces. Guiding of optical waves, generally, is achieved
by an appropriate spatial variation of the refractive index. A core dielectric material
with higher refractive $n_1$ index is surrounded by other dielectric materials with lower
refractive index $n_{2,3}$, the cladding. Depending on the geometry it can either be a planar
(1 dimensional) or a channel (2 dimensional) waveguide. The change of the refractive
index can be step-wise (step-index waveguide) or gradual (graded-index waveguide).

In the following, we will consider a planar dielectric waveguide with a step-index
profile. Figure 1.4 illustrates the waveguide layout and depicts the corresponding refrac-
tive index step-profile. In the special special case of $n_2 = n_3$ the waveguide is called
symmetric.

In the following, we will consider a planar dielectric waveguide with a step-index
profile. Figure 1.4 illustrates the waveguide layout and depicts the corresponding refrac-
tive index step-profile. In such a waveguide, guided light propagating in the core is reflected
internally by total reflection at the core-cladding interface. Figure 1.5 shows the angle
convention used. In the $x$-direction the structure has infinite extent.

Total reflection in the core is only possible if the complementary reflection angle $\tilde{\theta}$ is
larger than the critical angle for total reflection $\tilde{\theta}_c$:

$$\tilde{\theta} > \tilde{\theta}_c = \arcsin \frac{n_2}{n_1},$$  \hspace{1cm} (1.32)

For total reflection from a dielectric, a phase shift $\phi_r$ is induced which depends on the
angle $\tilde{\theta}$, the polarization of the light and the involved refractive indices.
Figure 1.5: Angle convention and illustration of light propagation in a planar waveguide. For any mode, the guided wave is in phase with itself after two reflections.

The condition for the development of a mode, i.e., the wave being in phase with itself after two reflections, leads to the mode equation of an asymmetric waveguide:

\[ k \cdot AC - \phi_{1}^{1-2} - \phi_{1}^{1-3} = 2\pi m, \quad m = 0, 1, 2, \ldots \]  \hspace{2cm} (1.33)

\[ 2k n_{1} d \sin \theta_{m} - \phi_{1}^{1-2}(\theta_{m}) - \phi_{1}^{1-3}(\theta_{m}) = 2\pi m, \quad m = 0, 1, 2, \ldots \]  \hspace{2cm} (1.34)

Here, \( m \) is the mode number and \( \phi_{1}^{1-2,3} \) is the phase shift induced by total reflection at the interface between the core (medium 1) and the corresponding cladding (medium 2 or 3). This transcendental equation can only be solved numerically or graphically [17, 18]. We present here only the result for TE modes, depicted in Fig. 1.6. For \( m = 0, 2, 4, \ldots \) the field distribution is symmetric, for \( m = 1, 3, 5, \ldots \) it is antisymmetric.

The actual propagation constant \( \beta_{m} \) depends on the mode number \( m \). With this propagation constant, an effective refractive index \( N \), defining the phase velocity of the guided light, can be defined according to

\[ \beta_{m} = k n_{1} \cos(\theta_{m}) = k N, \]  \hspace{2cm} (1.35)

\[ N = n_{1} \cos(\theta_{m}). \]  \hspace{2cm} (1.36)

The number of TE modes \( M \) of a planar dielectric waveguide is the smallest integer larger (indicated by the symbol “\( \geq \)”) than \( 2d n_{1} \theta_{c}/\lambda_{0} \), which means

\[ M \geq 2 \frac{d}{\lambda_{0}} NA, \]  \hspace{2cm} (1.37)

where

\[ NA = \sin \theta_{a} = (n_{1}^{2} - n_{2}^{2})^{(1/2)}, \]  \hspace{2cm} (1.38)

is the numerical aperture. In order to have a single mode waveguide, the waveguide has to be sufficiently thin. For symmetric planar waveguides, at least one mode is always existing.
Waveguides in integrated optics

Waveguides are indispensable and one of the fundamental building blocks for integrated optical applications and devices, including lasers, modulators, filters, switches, and couplers (e.g. [19]). The one or two dimensional confinement of the propagating light permits to improve the device characteristics such as lower losses and savings in drive voltage and power. For nonlinear-optical applications that need a high optical intensity, such as harmonic generation or parametric mixing, waveguides allow to maintain the high intensity over a much longer interaction length, and may therefore increase the conversion efficiency by several orders of magnitude. Since the nonlinear field increases with pump intensity and propagation distance, lateral confinement is a key issue for efficient interactions.

Also for optical switches and connectors in telecommunication devices, waveguides play an important role: they allow the coupling between different input and output channels without inflicting serious losses. Especially for such applications, dynamic and reconfigurable waveguides are desirable. In this thesis we investigated a method that allows to create such dynamic, reconfigurable waveguides in conducting electro-optic crystals based on the interband photorefractive effect.
1.5 Waveguide arrays

Waveguide arrays, also known as photonic lattices are material systems with a dielectric constant periodically modulated along one or two transverse directions in space. Compared to photonic crystals, the modulation period of a photonic lattice is larger than the light wavelength, and the resulting system can effectively behave as an array of coupled optical waveguides.

Light propagation in linear and nonlinear waveguide arrays exhibits behaviour characteristics of that encountered in discrete systems. The diffraction properties of these systems can be engineered, which opens up new possibilities for controlling the flow of light that would have been otherwise impossible in the bulk: these effects can be exploited to achieve diffractionless propagation and minimize the power requirements for nonlinear processes. This section gives a brief overview over the field of photonic lattices. For a complete review see e.g. refs. [20, 21].

Coupled mode theory

The standard theoretical approach for describing light propagation in photonic lattices is the coupled mode theory. The main assumption of this theory is that the lattice consists of weakly coupled identical single-mode waveguides. Thus, only coupling between nearest neighbors is considered. Furthermore, all details of the optical field transfer between the waveguides is packed up into a single uniform parameter — the coupling constant $C$.

In the case of an ideal, infinite-size array (without losses) with nonlinearity, this set of equation takes the form [22]

$$
\left[i \frac{d}{dz} + \beta \right] E_n + C (E_{n-1} + E_{n+1}) + \gamma(|E_n|^2) E_n = 0,
$$

where $E_n$ is the mode amplitude in the $n^{th}$ waveguide, $\beta$ is the propagation constant in a single waveguide, and $\gamma(|E_n|^2)$ is the nonlinear response of the material, which depends on the optical intensity in the $n^{th}$ waveguide $I_n = |E_n|^2$ and on the kind of nonlinearity. No second-order derivatives appear in Eq. (1.39) because all dispersive and diffractive effects are ignored in each waveguide. This set of equations is referred to as the discrete nonlinear Schrödinger equation. It describes the physical situation reasonably well when a CW beam is launched into a large array where each waveguide supports a single mode and confines it in both transverse dimensions (no diffraction).

Discrete diffraction relation

Even though no diffraction occurs within each waveguide, an optical beam can still spread over the whole array because of the coupling among the waveguides. This spreading is
1.5 Waveguide arrays

commonly referred to as discrete diffraction. The linear terms in Eq. (1.39) govern the
diffractive properties of the whole array. This equation can be solved analytically when
the nonlinear term is neglected. The solution is in the form of plane waves [22]:

$$E_n = E_0 e^{i n k_x d + i k_z z}.$$ \hspace{1cm} (1.40)

where $d$ represents the center-to-center spacing between the waveguides in the $x$-direction
and $k_x$ and $k_z$ are the wave-vector components in $x$ and $z$ direction respectively. Substituting
this solution into Eq. (1.39) without nonlinearity ($\gamma = 0$), we obtain the following
dispersion relation between $k_x$ and $k_z$:

$$k_z = \beta + 2C \cos(k_x d).$$ \hspace{1cm} (1.41)

An important property of this relation is its periodicity in $k_x$. Since $k_x$ is a continuous
variable, the spectrum of the propagation constant $k_z$ has a band structure where only
values between $\beta - 2C$ and $\beta + 2C$ are allowed. Due to the periodicity of $k_z$, there is
an infinite number of angular frequencies $k_x$ propagating with the same $k_z$. Using the
standard terminology from the field of solid-state physics, it suffices to consider only the
first Brillouin zone, which is formed in the range $|k_x d| < \pi$.

Following the standard definition of the group-velocity dispersion for optical pulses
[23], we can introduce a diffraction parameter for the entire array using

$$D = \frac{\partial^2 k_z}{\partial k_x^2}.$$ \hspace{1cm} (1.42)

The most striking feature of this equation in the context of photonic lattices is the
change in the sign of the diffraction parameter $D$ in the outer parts of the Brillouin zone.
The positive values of $D$ in the range $\pi/2 < |k_x d| \leq \pi$ are analogous to optical pulses
experiencing anomalous dispersion. In this region, diffractive properties are anomalous
in the sense that they are opposite to those experienced in nature. Moreover, diffraction
completely disappears around the two points $k_x d = \pm \pi/2$.

The parameter $k_x d$ corresponds to the phase difference between adjacent waveguides
and is also referred to as the “Bloch momentum”. Experimentally, such a phase difference
can be induced by tilting the input beam with respect to the array. The results
agree reasonably well with theory if the input beam covers at least four waveguides.

Fig. 1.7 shows the most spectacular consequences of anomalous diffraction. In a) and c),
measurement and modelling show that diffraction is comparable for two different input
angles, but different than in an isotropic medium: the beam tilted by 2.2° exits the
array at the same location as the untilted beam. Fig. 1.7(b) shows the beam tilted by
1.1° which corresponds to $k_x d = \pi/2$. The beam tilted at that angle crosses the array
without diffraction as theoretically predicted [24].
Figure 1.7: Discrete diffraction: Measured output intensity profile (top) and simulated propagation for a Gaussian excitation (bottom) with several input tilts (Figure from Ref. 24).

When only one waveguide is excited initially such that $E_n = 0$ at $z = 0$ for all $n \neq 0$, the relative phase between adjacent waveguides is not defined. However, Eq. (1.39) is analytically integrable in the linear case ($\gamma = 0$) and the solution is given by [22]

$$E_n(z) = E_0^n J_n(2Cz) e^{i\beta z},$$  \hspace{1cm} (1.43)

where $J_n$ is the $n^{th}$ order Bessel function. Physically, as the CW beam propagates along the waveguides, its power spreads into many nearby waveguides in a symmetric fashion such that the intensities are distributed as $J_n^2(2Cz)$ at any $z$. At a distance corresponding to $2Cz \approx 2.405$, $J_0$ vanishes and all the power disappears from the original waveguide. A part of the power reappears with further propagation, as dictated by the zeroth-order Bessel function.

**Band-Gap Structure of photonic lattices**

A more general approach, in which photonic lattices are regarded as an example of a general one-dimensional periodic structure, is the Floquet-Bloch analysis. For wave propagation in $z$ direction and a refractive index modulated periodically in $x$-direction, the solution for the electric field can be written as

$$E = E(x)e^{i\omega t - ikx}.$$  \hspace{1cm} (1.44)
Inserting Eq. (1.44) into the wave equation results in the Eigenmode equation

\[
\frac{\partial^2}{\partial x^2} + k_0^2 n^2(x) E(x) = k_z^2 E(x),
\]

(1.45)

with periodic refractive index \(n(x+d) = n(x)\). According to Blochs theorem, the solutions of Eq. (1.45) are of the form

\[
E_{nk_z}(x) = e^{ik_x x} u_{nk_z}(x),
\]

(1.46)

where \(u(x)\) is a periodic function with the same periodicity than \(n(x)\): \(u(x + d) = u(x)\). This analysis predicts that the spectrum of eigenvalues \(k_z\) (propagation-constant) of the array’s eigenmodes is divided into bands, separated by gaps, in which propagation modes do not exist [25, 26]. This bands are denoted by the index \(n\) in Eq. (1.46). A typical band-gap structure of a photonic lattice is shown in Fig. 1.8, where the propagation constant \(k_z\) is plotted versus the Bloch wavenumber \(k_x\). The right side of Fig. 1.8 shows a photograph of the modal intensity distribution at the array output for each band. There is a characteristic distribution for each band. Clearly, the Floquet -Bloch modes belonging to higher bands have much of their energy distributed in the lower index regions (between the waveguides). The coupled mode analysis from the previous section describes only propagation within the first of these bands, where the energy is concentrated in the high index waveguides. The diffraction curve of this first band is nearly sinusoidal, as predicted by the coupled-mode theory.

**Figure 1.8:** Left: reduced band-gap diagram of a photonic lattice. The shaded regions represent the photonic-lattice gaps. Right: Highly magnified images of pure FB modes excited in each band. (Figure from Ref. 26).
Introduction

Discrete solitons

Discrete solitons are self-localized wave packets, whose energy resides primarily in distinct waveguide array sites (hence discrete) and exist through a balance of discrete diffraction (coupling effects) and material nonlinearity.

Depending on the nature of the underlying nonlinear process, different families of discrete solitons are possible. To date, arrays made from materials with Kerr [27], quadratic [28] or photorefractive nonlinearities [29] have been shown to support such discrete localized states. For a given material, the nonlinear mechanism can be either self-focusing or defocusing. In the self-focusing case, the refractive index increases locally with intensity, whereas in the defocusing case it decreases.

In a nonlinear lattice, the optical field perturbs the refractive index, inducing a defect within an otherwise perfect photonic lattice. Consequently, the propagation constant \( k_z \) of this nonlinear defect-like state moves into a band-gap, and therefore its wavefunction decays rapidly away from the perturbed region, since propagation in the unperturbed array is not allowed for that \( k_z \). Hence the field becomes self-localized and a discrete soliton is formed. Note that as the power of this optical field increases, the deeper its propagation value moves into the gap, resulting in a more confined and transversely immobile discrete soliton.

The perturbation of the photonic lattice, which is induced by a soliton, is also seen by other low intensity beams or solitons. Therefore, their propagation through the lattice is significantly affected. This fact offers the possibility to control light by light, i.e. all-optical signal processing. This field covers a large part of recent investigations in photonic lattices – theoretically and experimentally [30,31].

Optically induced photonic lattices

There is an increasing interest in optically induced photonic lattices [20,29]. The advantages of such lattices are the saturable screening nonlinearity, allowing solitons to exist already at milliwatt power levels, the possibility to induce 2-dimensional lattices and of course the fact, that such lattices are produced in real time and can be reconfigured at any time, limited only by the response time of a certain material. The formation of such optically induced lattices is based on the photorefractive effect by interfering two or more plane waves.

In Chapter 5 we present a 1-dimensional array induced in the fast photorefractive material Sn\(_2\)P\(_2\)S\(_6\) by the interference of two light beams at \( \lambda = 514 \text{ nm} \).
Chapter 2

Electro-optical properties of near-stoichiometric and congruent lithium tantalate at ultraviolet wavelengths†

We measured the unclamped electro-optic coefficients $r_{13}$ and $r_{33}$ of near-stoichiometric and congruent lithium tantalate from $\lambda = 633$ nm down to the absorption edge at about 275 nm. Electro-optical coefficients up to $r_{33} = 52 \pm 1$ pm/V have been determined at $\lambda = 275$ nm. The reduced half-wave-voltage at 275 nm is $v_\pi = 300 \pm 10$ V for $r_{33}$ and $v_\pi = 560 \pm 20$ V for the configuration employing $r_c = r_{33} - (n_o/n_e)^3 r_{13}$ respectively. We propose a two-oscillator polarization potential model which very well describes our data in the UV. In the investigated wavelength range the values for stoichiometric and congruent LiTaO$_3$ differ by less than 3%, which is below the accuracy of our measurements. In addition, the refractive indices in the UV were measured and more precise Sellmeier parameters for the UV and visible were determined based on the previous and new refractive index data.

2.1 Introduction

The electro-optic effect is widely used in scientific and technical applications for modulation, deflection or processing of optical signals. There are many electro-optical materials which are used at visible or infrared wavelengths, but only a few of them are suitable

†The results of this Chapter have been published in the Journal of the Optical Society of America B, 23 (2), pp. 276–281 (2006).
for applications in the UV. Since laser sources in the UV are becoming available, there is an increasing interest in such low wavelength applications. Using UV light, a resolution proportional to \( \lambda \) can be achieved in applications like holographic interferometry or lithography. For volume holographic data storage, the storage density is increasing proportional to \( \lambda^{-3} \) when going to smaller wavelengths. Another important advantage of using lower wavelengths is the smaller divergence of such light beams.

A promising material for applications at UV wavelengths is stoichiometric lithium tantalate. It is transparent down to 260 nm where most of the standard electro-optical materials, e.g. LiNbO\(_3\) or K\(_2\)NbO\(_3\), show large light absorption. At 320 nm, congruent LiNbO\(_3\) exhibits an absorption constant of about 20 cm\(^{-1}\) while in congruent LiTaO\(_3\), this value is reached at 275 nm [32]. We have demonstrated earlier that in stoichiometric materials many physical properties are substantially different than in conventionally used congruent materials [32,33]. For example, the absorption edge is shifted deeper into the UV down to 300 nm for stoichiometric LiNbO\(_3\) and down to 260 nm for stoichiometric LiTaO\(_3\) [32]. Therefore also the refractive indices and electro-optic coefficients are expected to be different.

There is an increasing interest in the use of LiTaO\(_3\) for photorefractive and other applications at UV wavelengths. Besides lower absorption edge, stoichiometric LiTaO\(_3\) as compared to LiNbO\(_3\) shows higher Curie temperature, higher dark resistivity [32], smaller optical damage in the visible [7,32,34], smaller coercive fields [35], one order of magnitude longer dark-storage time of holograms [36] and smaller birefringence, which can even be adjusted to zero by controlling the stoichiometry and/or the temperature [37].

Recent investigations such as dynamic holography at the deep UV wavelength 257 nm [33,38]) and two-beam coupling at 364 nm [32] or two-color holographic recording with gating light at 350 nm [36] showed the potential of near-stoichiometric lithium tantalate for several applications in the UV, like tunable optical filters [38], non-volatile holographical data storage [39], high-speed optical processing with high resolution [40], light induced waveguides [41] or frequency doubling into the UV using periodically poled crystals [42]. For all of these applications, the material properties such as refractive indices and electro-optic coefficients are required.

Up to now, there exists no data on these coefficients at UV wavelengths in lithium tantalate. For congruent LiTaO\(_3\), the electro-optical coefficients have been known at 633 nm [43] and have recently been measured at near-infrared wavelengths up to 1558 nm [44]. Also refractive indices were known only down to 400 nm [45,46] and not in the UV wavelength range.

In this paper we determine the optical properties of congruent and near-stoichiometric lithium tantalate, namely the electro-optic coefficients and the refractive indices partic-
2.2 Lithium Tantalate

Congruent LiTaO$_3$ crystals can be grown by the traditional Czochralski method [48]. These crystals are intrinsically nonstoichiometric owing to a shift of the melt composition from the stoichiometry towards the Ta side. Consequently, to grow stoichiometric crystals, the melt composition has to be kept Li rich. This is achieved by adding stoichiometric powder to the melt at a rate equal to the crystal growth. This so-called double crucible Czochralski method is described in detail in Ref. 49.

In this paper one congruent (CLT) and one near-stoichiometric (SLT) lithium tantalate crystal grown by K. Kitamura et al. at the National Institute for Material Science, Tsukuba, have been investigated. The degree of stoichiometry of the SLT crystal was Li/(Ta+Li) = 49.9% with a Curie temperature of 684 °C. The absorption edge of our SLT crystal is slightly shifted from 275 nm to 270 nm [50]. The properties of the two investigated crystals are summarized in Table 2.1.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Dimension $a \times b \times c$ (mm$^3$)</th>
<th>Curie Temperature (°C)</th>
<th>Li/(Ta+Li) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CLT</td>
<td>$7.3 \times 0.975 \times 10.2$</td>
<td>605 ± 1</td>
<td>48.6</td>
</tr>
<tr>
<td>SLT</td>
<td>$1.958 \times 7.9 \times 8.2$</td>
<td>684 ± 1</td>
<td>49.9</td>
</tr>
</tbody>
</table>

Below the Curie temperature, the point-group symmetry of LiTaO$_3$ is uniaxial $3m$ with the optical axis, the crystallographic $c$-axis, along the three-fold symmetry axis. It has therefore two different refractive indices $n_a = n_b = n_o$ for ordinary and $n_c = n_e$ for extraordinary polarization, which we both determined for the two crystals. According to the $3m$ point group symmetry, lithium tantalate has eight nonzero elements of the electro-
optic tensor in the reduced notation, with only four independent values: $r_{12} = -r_{22} = r_{61}$, $r_{51} = r_{42}$, $r_{13} = r_{23}$ and $r_{33}$. When applying a field in the $c$-direction, we are able to measure the highest coefficient $r_{33}$ as well as $r_{13}$.

### 2.3 Refractive indices

Interferometric refractive index measurements have been performed using a standard Michelson interferometer as depicted in Fig. 2.1 and vertically polarized light. The crystal was mounted onto a rotation stage in one arm of the interferometer. It can be rotated around an axis perpendicular to the light beam and parallel to the light polarization. When the crystal is rotated by an angle $\theta$ away from the position of normal incidence, the optical path length can be determined by counting the number $m$ of interference fringes at the interferometer output. With $\theta$ and $m$ we can calculate the refractive index $n$ using [51]

$$n = \frac{\alpha^2 + 2(1 - \cos \theta)(1 - \alpha)}{2(1 - \cos \theta - \alpha)},$$

where $\alpha = (m \cdot \lambda)/(2 \cdot L)$, $\lambda$ is the wavelength at which the refractive index was measured and $L$ is the crystal thickness. Using this technique, refractive indices could be measured with a precision of about 0.01. The refractive indices were measured at 5 different

![Figure 2.1: Interferometric setup for measuring the refractive indices and electro-optical coefficients. For the determination of refractive indices, the crystal was mounted onto a rotation stage. The unclamped electro-optic coefficients were measured by applying a low frequency sine voltage.](image)
wavelengths between 275 nm and 364 nm using an Ar$^{++}$ laser (Spectra-Physics) for the ordinary and the extraordinary wave. The results are shown in Fig. 2.2 for the CLT and SLT crystal. The data above 400 nm had already been reported before and the wavelength dispersion of the refractive index was described by the Sellmeier equation [45, 46]:

$$n^2(\lambda) = A + \frac{B}{\lambda^2} - C - D\lambda^2. \quad (2.2)$$

This function is plotted in Fig. 2.2 (dashed lines) using the Sellmeier coefficients $A$, $B$, $C$ and $D$ from Ref. 45 and Ref. 46 for CLT and SLT respectively. The measured data in the UV deviates from these curves. Therefore we adjusted the coefficients to match the data also at smaller wavelengths (solid lines). To give the Sellmeier coefficients a more physical meaning we have also rewritten Eq. (2.2) as explained in the following.

**Figure 2.2:** Refractive indices of (a) CLT and (b) SLT. The dashed lines and the data above 400 nm are taken from literature [45, 46]. The solid lines correspond to the new coefficients (Table 2.2). There is a small correction at lower wavelengths due to the new data in the UV.
Wemple and DiDomenico [52] introduced a simple oscillator model for describing the dispersion of the refractive index:

\[ n^2(\omega) - 1 = \sum_i \frac{f_i}{\omega_i^2 - \omega^2}, \quad (2.3) \]

This equation separates all interband transitions into individual oscillators with frequency \( \omega_i \) and strength \( f_i \). Energy band calculation showed that the conduction band of oxygen-octahedra ferroelectrics like lithium tantalate can be described by considering two sub-bands which can be related to the two main oscillators [52]. This leads to the two oscillator Sellmeier equation:

\[ n^2 - 1 = \frac{S_\gamma \lambda_\gamma^2}{1 - (\lambda_\gamma/\lambda)^2} + \frac{S_\epsilon \lambda_\epsilon^2}{1 - (\lambda_\epsilon/\lambda)^2} = \frac{E_{d\gamma}E_\gamma}{E_\epsilon^2 - E^2} + \frac{E_{d\epsilon}E_\epsilon}{E_\gamma^2 - E^2}. \quad (2.4) \]

Here, \( S_i = f_i/(2\pi c)^2 \) are the oscillator strength factors and \( \lambda_i = 2\pi c/\omega_i \) the resonant light wavelengths for the two oscillators \((i = \epsilon, \gamma)\). The expression on the right hand side is the energy description of Wemple and DiDomenico with the oscillator energies \( E_i = h\omega_i \) and the dispersion-energy parameters \( E_{di} = S_i h^2 c^2 / E_i \). Since one oscillator is resonant far in the UV at about 9 eV and one at about 5 eV, i.e. \( \lambda_\gamma \ll \lambda_\epsilon < \lambda \) [52], one can simplify the first term of Eq. (2.4) by a constant \( A = S_\gamma \lambda_\gamma^2 = E_{d\gamma} / E_\gamma \) and leaving the second term, which corresponds to the dominating oscillator, unchanged.

With this simplification, Eq. (2.4) describes the measured data well in the wavelength range below 500 nm. In this model only interband transitions are considered, and no optical lattice vibrations which occur at far infrared wavelengths. We can include these vibrations by another oscillator \( S_{IR} \lambda_{IR}^2 / (1 - (\lambda_{IR}/\lambda)^2) \). Since the lattice vibrations occur at higher wavelengths \( \lambda_{IR} \gg \lambda \), we can reduce this additional term to \(-D\lambda^2\) with \( D = S_{IR} \) and get

\[ n^2(\lambda) - 1 = A + \frac{S_\epsilon \lambda_\epsilon^2}{1 - (\lambda_\epsilon/\lambda)^2} - D\lambda^2. \quad (2.5) \]

It is easy to assign the coefficients \( A, S_\epsilon, \lambda_\epsilon, D \) to the coefficients \( A, B, C, D \) of Eq. (2.2). In the energy description, Eq. (2.5) can be rewritten as

\[ n^2(E) - 1 = \frac{E_{d\epsilon}}{E_\gamma} + \frac{E_{d\gamma}E_\epsilon}{E_\epsilon^2 - E^2} - \frac{E_{dIR}E_{IR}}{E^2}. \quad (2.6) \]

This form of the Sellmeier equation is used in the next section for the derivation of the polarization potential model for describing the dispersion of the electro-optic coefficient.

The Sellmeier coefficients in Table 2.2 correspond to the oscillator description (Eq. (2.5)) with the oscillator energy \( E_\epsilon = hc/\lambda_\epsilon \). They were evaluated taking into account the measured data in the UV and the literature data from Ref. 45 and Ref. 46 for CLT and
Table 2.2: Sellmeier parameters for describing the dispersion of $n_o$ and $n_e$ in CLT and SLT crystals. The given values are the fit parameters for calculating the electro-optic coefficient using Eq. (2.5) and $E_\epsilon = \frac{hc}{\lambda \epsilon}$.

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>$A$</th>
<th>$S_\epsilon$ (µm$^{-2}$)</th>
<th>$E_\epsilon$ (eV)</th>
<th>$D$ (µm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CLT - $n_o$</td>
<td>1.08</td>
<td>68.66</td>
<td>6.59</td>
<td>0.0241</td>
</tr>
<tr>
<td>SLT - $n_o$</td>
<td>1.72</td>
<td>43.45</td>
<td>6.05</td>
<td>0.0327</td>
</tr>
<tr>
<td>CLT - $n_e$</td>
<td>1.22</td>
<td>62.02</td>
<td>6.43</td>
<td>0.0242</td>
</tr>
<tr>
<td>SLT - $n_e$</td>
<td>1.68</td>
<td>45.57</td>
<td>6.16</td>
<td>0.0320</td>
</tr>
</tbody>
</table>

SLT respectively. The values in Table 2.2 are fit parameters for calculating the refractive indices at any wavelength, but the physical properties exhibit an error of about ±12%.

Note that the empirical law of Wemple and DiDomenico [47,52] that the ratio $E_0/S_0$ is a constant for most inorganic crystals, is only valid for the one-oscillator Sellmeier equation, where all oscillators are considered in a mean oscillator with strength $S_0$ and energy $E_0$.

If we compare the new Sellmeier parameters (solid lines in Fig. 2.2) with the old ones (dashed lines), there is a small variation at lower wavelengths. This is because the old parameters have been evaluated from refractive index data above 400 nm only, not considering the main dispersion range in the UV. We have used the newly determined Sellmeier parameters to evaluate electro-optic coefficients from the measurements as described in the following section.

### 2.4 Electro-optic coefficients

The electro-optic coefficients were measured with the same Michelson interferometer as was used for the determination of refractive indices. The setup is depicted in Fig. 2.1. Similar measurements have been performed in other materials like LiNbO$_3$ [53], COANP, PNP [54] or Sn$_2$P$_2$S$_6$ [5]. For the measurement, the crystal was orientated with the $c$-axis in the propagation plane and perpendicular to the incoming laser beam. An electric field was applied along the $c$-axis using silver-paste electrodes. The field induced phase shift is then given by

$$\varphi = -\frac{2\pi L}{\lambda} n^3 r_{\text{eff}} E + \frac{4\pi L}{\lambda} (n - 1) d_{\text{eff}} E,$$

where $L$ is the length of the crystal, $n$ the refractive index and $r_{\text{eff}}$ the eo-coefficient of
the corresponding light polarization ($n_e$ and $r_{33}$ for horizontally polarized light, $n_o$ and $r_{13}$ for vertically polarized light), $d_{\text{eff}}$ is the effective piezoelectric coefficient. The first term of Eq. (2.7) is the phase shift due to the electro-optic effect while the second term describes the contribution due to the piezoelectric change of the crystal length. With the known value of $d_{\text{eff}} = -3 \text{ pm/V}$ [55], this piezoelectric effect represents a small correction in the order of about 1% to $r_{33}$ and about 5% to the smaller coefficient $r_{13}$.

If the applied field is a low frequency modulated sine-wave signal ($\Omega \simeq 1\text{kHz}$) with a small amplitude $\delta V$, the induced phase shift is also modulated with this frequency as well as the measured intensity at the interferometer output. Using a lock-in amplifier,

![Figure 2.3: Measured dispersion of the electro-optic coefficients $r_{33}$ and $r_{13}$ of CLT and SLT. The solid lines correspond to Eq. (2.12), while the dashed lines correspond to Eq. (2.11) which describes the dispersion at longer wavelengths. The parameters for the theoretical curves are listed in Table 2.3.](image_url)
we are then able to measure very small intensity modulations $\delta I$. The working point of the interferometer was adjusted by means of a piezoelectric movable mirror as depicted in Fig. 2.1. If the working point is set to maximum modulation, the refractive index can be determined using

$$r_{\text{eff}} = \frac{-\delta I}{\delta V} \frac{d}{d\pi n^3 \Delta I} + \frac{2(n-1)}{n^3} d_{\text{eff}}. \quad (2.8)$$

Here, $d$ is the distance between the electrodes and $\Delta I = I_{\text{max}} - I_{\text{min}}$ the intensity contrast of the interferometer which was determined by moving the piezo-mirror to detect minimum and maximum intensity at the interferometer output. This configuration allows us to determine the unclamped electro-optic coefficients $r_{33}$ and $r_{13}$. The measurements were performed at different wavelengths between 275 nm and 528 nm (Ar$^+$ laser) and at 633 nm (HeNe laser). The absorption at 275 nm was about 1 cm$^{-1}$ in SLT and about 15 cm$^{-1}$ in CLT. Therefore transmission experiments were still possible since the crystals were relatively thin (CLT: 1 mm, SLT: 2 mm). By measuring the field induced absorption, we found that the electro-absorption did not influence the electro-optic measurements. At low frequencies of the applied electric field ($\Omega < 500$ Hz) we observed a significant photoconductive screening of the applied field, which disappeared if the electric field frequency was increased to $\Omega \approx 1$ kHz. Therefore all the measurements were performed at $\Omega \approx 1$ kHz and low light intensities (10 $\mu$W/cm$^2$ – 1 mW/cm$^2$). The experimental results for the two crystals are depicted in Fig. 2.3.

In his work, Wemple presented a polarization-potential model for describing the dispersion of the electro-optic coefficient [47]: In one dimension, the electro-optic coefficient can be calculated by a differentiation with respect to the induced polarization $P$:

$$r = \epsilon_0(\epsilon - 1) \cdot \frac{d}{dP} \left( \frac{1}{n^2} \right), \quad (2.9)$$

where $\epsilon$ is the low-frequency dielectric constant in the corresponding direction. Starting from the single oscillator Sellmeier equation for the dispersion of $n$

$$n^2 - 1 = \frac{S_0 \lambda_0}{1 - (\lambda_0/\lambda)^2} = \frac{E_d E_0}{E_0^2 - E^2}, \quad (2.10)$$

the electro-optic coefficient can be derived using Eq. (2.9) as

$$r_{\text{eff}}(\lambda) = \epsilon_0(\epsilon - 1) \left( 1 - \frac{1}{n(\lambda)^2} \right)^2 \frac{dE_0/dP}{E_d} \left[ (1 - K) + (1 + K) \left( \frac{\lambda_0}{\lambda} \right)^2 \right], \quad (2.11)$$

with the dispersion energy $E_d = S_0 h^2 c^2 / E_0$, the polarization potential $dE_0/dP$ and the dispersion parameter $K = d(\ln E_d)/d(\ln E_0)$. 

29
Electro-optical properties of LiTaO$_3$

Table 2.3: Parameters for describing the dispersion of the electro-optic coefficient. The given values are the fit parameters for calculating the electro-optic coefficient using Eq. (2.11) for $a$ and Eq. (2.12) for $b$.

<table>
<thead>
<tr>
<th>Coeff.</th>
<th>$K$</th>
<th>$dE_0/dP$ (eVm$^2$/As)</th>
<th>$K_1$</th>
<th>$K_2$</th>
<th>$dE_\epsilon/dP$ (eVm$^2$/As)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CLT - $r_{13}$</td>
<td>0.51</td>
<td>0.92</td>
<td>2.70</td>
<td>-2.03</td>
<td>3.54</td>
</tr>
<tr>
<td>SLT - $r_{13}$</td>
<td>0.56</td>
<td>0.80</td>
<td>1.53</td>
<td>-1.27</td>
<td>1.66</td>
</tr>
<tr>
<td>CLT - $r_{33}$</td>
<td>-0.31</td>
<td>1.54</td>
<td>2.01</td>
<td>-2.00</td>
<td>4.65</td>
</tr>
<tr>
<td>SLT - $r_{33}$</td>
<td>-0.39</td>
<td>1.40</td>
<td>1.18</td>
<td>-1.98</td>
<td>3.04</td>
</tr>
</tbody>
</table>

Comparing this model with the measured electro-optic coefficients, the polarization potential model (Eq. (2.11)) cannot describe the data at shorter wavelengths. This is because the single oscillator Sellmeier equation is a long wavelength approximation and cannot be applied near the absorption edge. For comparing with other materials, $K$ and $dE_0/dP$ that describe the data at wavelengths above 350 nm are listed in Table 2.3 and the slopes are plotted in Fig. 2.3 as dashed lines.

In order to better describe the data near the absorption edge, we extend here the derivation of the electro-optical coefficient with the simplified two oscillator Sellmeier equation (Eq. 2.6) where we considered additional contribution of the second oscillator in a constant $A = E_{dl}/E_\gamma$. Since we are interested in the description of electro-optic coefficients near the absorption edge, i.e. $E \gg E_{IR}$, we can neglect the infrared term. Using Eq. (2.9), we get

$$r_{eff}(\lambda) = \epsilon_0(\epsilon - 1) \left( 1 - \frac{1 + A}{n(\lambda)^2} \right)^2 \frac{dE_\epsilon/dP}{E_{dl}} \times \left[ (1 - K_1 - K_2) + (1 + K_1 + 2K_2) \left( \frac{\lambda_\epsilon}{\lambda} \right)^2 - K_2 \left( \frac{\lambda_\epsilon}{\lambda} \right)^4 \right]. \quad (2.12)$$

$K_1$ and $K_2$ are dispersion parameters, where

$$K_1 = \frac{d \ln E_{dl}}{d \ln E_\epsilon} \quad (2.13)$$

describes interband transition strength changes of the dominating oscillator induced by the polarization, and

$$K_2 = \frac{d \ln (E_{dl}/E_\gamma)}{d \ln E_\epsilon} \cdot R \quad (2.14)$$
describes polarization induced changes of the second oscillator. The quantity \( R = (E_x E_d) / (E_y E_d) \) was previously introduced by DiDomenico to describe the two oscillator model and was expected to be \( R \lesssim 0.5 \) for most of the inorganic crystals [52]. If we calculate this quantity from the Selmeier parameters (Table 2.2), we get values between 0.4 and 0.9, which is in good agreement with that approximation. Fig. 2.3 shows that the extended model (solid lines) well describes our data in the UV region. The parameters for the slopes are listed in Table 2.3. For wavelengths above 400 nm, the \( r_{33} \) value is in SLT by about 0.5 pm/V higher than in CLT, which is below the accuracy of our measurements. Below 350 nm it becomes slightly higher for CLT, which could be attributed to the shifted absorption edge, however, the difference is still within the experimental error.

An important quantity for the application of electro-optic materials is the half-wave-voltage which is defined as

\[
V_\pi = \frac{\lambda}{n^3 r L} = v_\pi \frac{d}{L},
\]

where \( d \) is the distance between the electrodes, \( L \) the length of the beam path in the crystal, \( n \) the refractive index, \( r \) the electro-optic coefficient and \( \lambda \) the wavelength of the light. It defines the applied voltage needed for rotating the beam polarization by 90°. A quantity which is independent of the crystal geometry (i.e. \( d \) and \( L \)) is the reduced half-wave-voltage \( v_\pi = \lambda / (n^3 r) \). This quantity is depicted in Fig. 2.4 for the SLT crystal as a function of the wavelength using Eq. (2.5) and Eq. (2.12) and the parameters from Table 2.2 and Table 2.3 for \( n(\lambda) \) and \( r(\lambda) \) respectively. The dispersion of \( v_\pi \) is similar in both investigated crystals but the absorption edge of SLT is shifted towards shorter wavelengths. At \( \lambda = 275 \text{ nm} \), the absorption in CLT is already 15 cm\(^{-1}\) while in SLT it is only about 1 cm\(^{-1}\). The reduced half-wave voltage at the deep UV wavelength \( \lambda = 275 \text{ nm} \) is about 300 V for the larger coefficient \( r_{33} \), and about 560 V for \( r_c = r_{33} - (n_o/n_e)^3 r_{13} \).

2.5 Conclusions

We have determined the dispersion of the unclamped electro-optic coefficients \( r_{13} \) and \( r_{33} \) in congruent and near-stoichiometric lithium tantalate down to \( \lambda = 275 \text{ nm} \). Electro-optic coefficients up to \( 52 \pm 1 \text{ pm/V} \) were measured at \( \lambda = 275 \text{ nm} \). The measured wavelengths cover the range from the deep UV absorption edge at 270 nm over nearly the whole visible spectrum to 633 nm. We have extended the polarization potential model from Wemple and DiDomenico to describe the dispersion of the coefficients in the UV. The theoretical curve describes very well the experimental data within the error of the measurements. In the investigated wavelength range the values for SLT and CLT
Figure 2.4: Absorption $\alpha_e$ in CLT and SLT for the extraordinary polarization and reduced half-wave voltage $v_\pi$ for the higher electro-optic coefficient $r_{33}$ and for the configuration employing $r_c = r_{33} - (n_o/n_e)^3 r_{13}$. Since $v_\pi$ is similar for both investigated crystals, only the one of SLT is plotted. For comparing the absorption, a vertical dotted line is plotted at $\lambda = 275$ nm.

differ by less than 3%, which is below the accuracy of our measurements. The reduced half-wave-voltage at 275 nm was found to be about 300 V for $r_{33}$ and about 560 V for the configuration employing $r_c = r_{33} - (n_o/n_e)^3 r_{13}$, for both crystal compositions.

In addition the refractive indices at UV wavelengths from 275 nm to 400 nm were determined and the Sellmeier coefficients for the UV and visible wavelengths reevaluated.
Chapter 3

Deep UV light-induced, fast reconfigurable and fixed waveguides in Mg doped LiTaO$_3$†

Dynamic waveguides are induced beneath the surface of magnesium doped near-stoichiometric lithium tantalate by deep UV light at $\lambda = 257$ nm using the interband photorefractive effect. The waveguides can be reconfigured in 10 ms at UV intensities of 100 mW/cm$^2$. We show the importance of the background illumination for the build-up of dynamic optical waveguides. We also present a new fixing process of the light-induced waveguide structures when the background light is absent. These quasi-fixed structures with dark decay times of several days are due to charges trapped in deep traps.

3.1 Introduction

Optical waveguides are one of the fundamental building blocks for integrated optical applications and devices such as modulators, filters, switches and couplers [56]. In nonlinear optical applications like harmonic generation or parametric mixing, guiding the light allows maintaining a high intensity over a much longer interaction length leading to larger conversion efficiencies as in bulk materials. There are many techniques for fabricating permanent waveguides including ion in-diffusion, ion exchange, ion implantation, fs laser ablation, photolithography and epitaxial thin film deposition. By using these techniques, a permanent change of the refractive index in the waveguide region is obtained. This precludes an easy reconfiguration of the waveguide structure. Recently, several techniques based on light-induced refractive index changes in photorefractive materials have been

†The results of this Chapter have been published in Optics Express 14, pp. 8278–8289 (2006).
developed [29,41,57,58]. One of the advantage of such optically induced waveguides is that they can be reconfigured solely by changing the light illumination. For applications such as optical switching, optical routing or dynamic optical interconnections, fast reconfigurable waveguides are required, which can be easily achieved with the light induced techniques.

A promising material for frequency doubling [59,60], electro-optics [61], long-persistent holographic storage [36] and interband photorefraction [33] is near-stoichiometric and/or Mg-doped LiTaO$_3$, that is transparent down to UV wavelengths near $\lambda = 260$ nm. The advantage of this material compared to the as-grown congruent LiTaO$_3$ for the above mentioned applications is the greatly reduced concentration of intrinsic trap levels that are responsible for the conventional photorefractive effect. [33,36,59–61] We demonstrate here the fabrication of light-induced waveguides in magnesium doped near-stoichiometric lithium tantalate (SLT:Mg). We first apply the light-induced technique that was previously used for KNbO$_3$ [41], where the waveguides were produced via interband photorefractive field screening in a biased crystal. The induced waveguides in SLT:Mg are reconfigurable in a fast way using controlling light at $\lambda = 257$ nm, with waveguide build-up times of 10 ms at UV intensities of 100 mW/cm$^2$.

Recently, in SLT:Mg, a quasi-fixing of holographic gratings near the crystal surface was observed using interband illumination [62, 63]. We investigate this quasi-fixing via interband photorefractive measurements in the transverse geometry. We show that this fixing is due to charges trapped in long-lifetime deep traps. We further demonstrate, that the same fixing effect can be used to obtain quasi-fixed waveguide structures in this material. Using such fixing mechanism, in routing and switching applications, an optical interconnection can be maintained without the need of any light illumination. When the connection is no longer required, it can be deleted by simply illuminating the crystal homogeneously, allowing another new connection to be established leading to a new waveguide structure.

3.2 Experimental

3.2.1 Growth and preparation of Mg-doped near-stoichiometric LiTaO$_3$

We used a magnesium doped near-stoichiometric LiTaO$_3$ crystal (SLT:Mg) grown by K. Kitamura et al. at the National Institute for Material Science, Tsukuba. The crystal growth axis was along $c$ using a lithium-rich melt (LiO$_2$:Ta$_2$O$_5$ = 58:42 as mole ratio) using the double crucible Czochralski method equipped with an automatic powder sup-
3.2 Experimental

MgO was added into this Li-rich melt. The grown crystal was poled by applying an electric field along the \( c \)-axis at a temperature higher than the Curie temperature \( T_c = 694.3 \pm 1 \)°C. The MgO concentration of the grown crystal was 1.0 mol% as determined by Ion coupled Plasma Mass Spectroscopy. The \( a \) and \( b \) surfaces and the edges of the crystal were polished from both sides. On the \( c \) surfaces, silver-paste electrodes were painted for applying the electric field. The crystal length along the \( a \)-axis (guiding direction) was 7.91 mm, and the distance between the electrodes (\( c \)-axis) was 9.0 mm.

![Figure 3.1: Simplified electric field and refractive index distribution in a photorefractive crystal for the generation of light induced waveguides by band-to-band excitation:](image)

- a) Unperturbed state with refractive index \( n_0 \).
- b) External field \( E_0 \) decreases the refractive index homogeneously by \( \Delta n \).
- c) UV-excited charges screen the external field and produce a waveguide.

### 3.2.2 Light induced waveguides

In photoconducting electro-optic crystals reconfigurable waveguides are produced by interband photorefraction [41] as explained in the following and as illustrated in Fig. 3.1. A homogeneous external electric field \( E \) that is applied to an electro-optic crystal induces a uniform change in the refractive index:

\[
\Delta n = -\frac{n^3}{2}r_{\text{eff}}E, \tag{3.1}
\]

where \( r_{\text{eff}} \) is the effective electro-optic coefficient for the chosen configuration and \( n \) the refractive index for the corresponding wavelength and polarization. If a small portion of
such a biased photorefractive crystal is nonuniformly illuminated, free charge carriers are produced in the bright regions, which screen the applied field due to charge transport and trapping in dark regions. As a result one gets a strongly reduced electric field in the bright regions, while in dark regions the field is basically unchanged. Via the electro-optic effect, this field distribution generates the refractive index profile, where the refractive index in the dark regions is lowered by $\Delta n$. Using a proper field direction and a suitable distribution of the illumination, light induced waveguide structures can be generated in this way. By changing the illumination the waveguide structure can be reconfigured.

By choosing light with photon energy larger than the band-gap energy of the material, charges can be excited directly from one band to the other, i.e. by interband excitation \[8\]. This process is much more effective in terms of use of the incident photons with respect to a conventional photorefractive effect, where the photoexcitation occurs from dopant or impurity energy levels within the material band gap. In LiTaO$_3$ interband excitation leads to three orders of magnitude faster response compared to conventional trap-level excitation \[33\]. Due to the strong light absorption at interband wavelengths ($\alpha_z = 690 \pm 40 \text{ cm}^{-1}$ at $\lambda = 257 \text{ nm}$ in SLT:Mg which was measured using a thin plate of thickness 100 $\mu$m \[64\]), the screened regions are just underneath the surface. Thus, the waveguides were probed directly beneath the surface, which was possible due to sharp polished crystal edges.

A schematic view of the experimental set-up to produce light induced waveguides is shown in Fig. 3.2. An electric field of about 5.5 kV/cm was applied along the $c$-axis of the crystal using silver painted electrodes. A mask with a slit width of 100 $\mu$m was homogeneously illuminated by 257 nm UV light polarized parallel to the $c$ axis of the crystal. The UV light was provided by an external cavity frequency doubling (WaveTrain, Spectra Physics) of the output of an Ar-ion laser (Innova 200 Coherent Inc.), producing continuous-wave laser radiation at $\lambda = 257 \text{ nm}$ with a maximum output power of about 200 mW. The slit was then imaged by the cylindrical lens $CL_2$ ($f = 75 \text{ mm}$) onto the $b$ surface of the crystal so that the imaged UV stripe had a width of about 20 $\mu$m along the $c$-axis and was aligned parallel to the $a$-axis. With a beamsplitter, part of the UV light was separated and after rotating the polarization to avoid interference and expanding the beam, used to homogeneously illuminate the crystal. This background light produced a homogeneous conductivity that permitted a better definition of the narrow stripe region where the external field was screened.

The probe beam from a HeNe laser (633 nm) polarized along the $c$-axis of the crystal was focused by a spherical lens $L_1$ ($f = 80 \text{ mm}$) to a waist of 30 $\mu$m at the entrance of the waveguide. The exit of the waveguide was imaged by a lens $L_2$ ($f = 30 \text{ mm}$) onto a charge-coupled device (CCD) camera and a photodiode. With the photodiode we
Figure 3.2: Top (a) and front (b) views of the experimental set-up for producing light induced waveguides (schematic). L₁ and L₂ are spherical lenses, CL₁ and CL₂ cylindrical lenses, BS beam splitters, M mirror, U the applied voltage.

measured the dynamics of the waveguide while the CCD camera recorded the distribution of the output light. A pinhole was put in front of the photodiode in order to detect the peak intensity \( I_0 \) in the center of the readout beam. This intensity increases if the beam is guided in the center. The magnitude of the detected intensity \( I_0 \) is a measure of the total power located in the center and is approximately inverse proportional to the waveguide width \( d \).

3.2.3 Holographic characterization

Light induced charge excitation and recombination processes in photorefractive materials, which are relevant for our light induced waveguide experiments, can be investigated through holographic diffraction experiments [8]. Two light beams that interfere in a photorefractive crystal produce a space charge electric field due to charge excitation in bright regions and recombination in darker regions. This electric field induces a refractive index grating via the electro-optic effect. In conventional photorefraction, charges are excited from mid band-gap levels. When the photon energy of the interfering beams is larger than the band-gap energy of the material, charges can be excited directly from one band to the other. This interband photorefractive effect produces a holographic grating which, in a simplified way, consists of two grating types [65]. Near the surface, where the writing intensity is higher, we obtain gratings for that band-to-band recombination is dominant. At greater depth, where the light intensity is smaller, recombination into traps dominates.
Interband photoreflective effects in SLT:Mg have been previously studied in longitudinal geometry, where the writing beam and the read-out beam enters the crystal from the same surface [33] and the diffracted beam is a mixture of diffraction at different grating compositions along the beam path. The depth structure of an interband photoreflective grating can be determined in the transverse readout geometry, for that the read-out beam is propagating parallel to the input surface of the recording beams. In this geometry, the read-out beam interacts over the whole crystal length with the same grating composition. Therefore the diffraction efficiency, which is defined as the ratio between diffracted and incident light intensities inside the crystal, reflects the grating composition at the depth of propagation and was determined by measuring the diffracted power after the crystal, taking into account Fresnel losses.

3.3 Results and discussion

3.3.1 Depth profile of the interband grating

Fig. 3.3 shows the result of a holographic measurement in SLT:Mg where the diffraction efficiency was measured for different depths of the readout beam. The grating was written with two UV beams at $\lambda_{UV} = 257$ nm and probed with a HeNe-beam. The crystal orientation is depicted on the right side of Fig. 3.3. The angle between the writing beams was $2\theta_{UV} = 16.4^\circ$ corresponding to a grating spacing of $\Lambda = \lambda_{UV}/(2 \sin \theta_{UV}) = 0.9 \, \mu m$. The grating was written with a total UV intensity of $I_{UV} = 250$ mW/cm² at the crystal surface. The regions of the two different gratings can be distinguished due to the fact, that in LiTaO₃ the trap-dominated grating and the band-dominated grating are mutually phase shifted by $\pi$ [33,63]. Therefore the diffracted beams from the two different gratings interfere destructively, which gives rise to the observed minimum of the efficiency profile (open squares in Fig. 3.3). The shaded areas in Fig. 3.3 show the dominant regions of the two gratings.

Quasi-fixing of holographic gratings at room temperature in SLT:Mg was recently reported [62,63]. We measured the depth profile of this quasi-fixed grating after switching off both writing beams at the same time. The filled circles in Fig. 3.3 show the measured diffraction efficiency as a function of the depth of the read-out beam. One can clearly see that the location of this fixed grating corresponds to the location of the trap dominated grating. This indicates that charges excited by the high photon energy UV light are in this region located in deep traps with a longer lifetime and an energy level higher than the photon energy of the readout beam. The lifetime in the dark was estimated to several days, while under permanent read-out with He-Ne light, the fixed grating decayed within
3.3 Results and discussion

3.3.1 Diffraction Efficiency

Figure 3.3: Left: Diffraction efficiency $\eta$ as a function of the depth of the readout beam $d$ beneath the illuminated surface measured during recording of the grating (open squares) and after blocking the UV writing beams (filled circles). The shaded regions indicate the interband dominated grating (light shading) and the trap dominated grating (dark shading) as explained in the text. One can clearly see, that the fixed grating is located at the depth of the trap dominated grating. Right: Set-up and crystal orientation for diffraction measurements in the transverse geometry.

10 hours. Homogeneous illumination with light at lower wavelengths ($\lambda \lesssim 360$ nm) could excite the charges from these deep traps and destroy the fixed structures. A fixing process was also observed in our waveguide experiments with SLT:Mg which we will discuss in section 3.3.4.

3.3.2 Waveguide profile

In the light-induced waveguide experiment (Fig. 3.2), we first measured the beam profile of the read-out beam at the output surface of the crystal. The images are depicted in Fig. 3.4 for the situations without (left) and with (right) UV stripe illumination. In both cases, an electric field of about 5.5 kV/cm was applied and the crystal was illuminated with a uniform background UV-light with an intensity of approximately 10 mW/cm$^2$. The UV stripe had an intensity of about 200 mW/cm$^2$. In absence of the UV stripe, the natural diffraction of the probe beam was observed, which expands to a full width half maximum (FWHM) of about 70 $\mu$m (left-hand side). On the right-hand side of Fig. 3.4, one can clearly distinguish, that the UV stripe produced a waveguide in which the red probe beam was guided. The FWHM of the intensity distribution of the guided waveguide mode was about 17 $\mu$m. The central beam profile along the orthogonal axis was not significantly affected by the UV stripe.
Light-induced waveguides in Mg-doped near-stoichiometric LiTaO$_3$

Figure 3.4: Top, CCD images of the intensity distribution of the He-Ne probe beam at the exit face of the biased crystal. Bottom, one dimensional beam profiles along the white dashed arrows. The left-side images correspond to the situation with UV illumination off, while for the right side images the UV illumination was on.

The refractive index change for the guided He-Ne light, induced by the UV stripe, can be calculated using Eq. (3.1). With the refractive indices $n_2 = 2.177$ and $n_3 = 2.175$, the electro-optic coefficients $r_{13} = 8 \text{ pm/V}$ and $r_{33} = 31 \text{ pm/V}$ [61], and the electric field $E_3 = 5.5 \text{ kV/cm}$, the induced refractive index changes are $\Delta n_2 = -2.3 \times 10^{-5}$ and $\Delta n_3 = -8.8 \times 10^{-5}$ for the two polarizations respectively.

We approximate our waveguide with a step-profile planar waveguide. The width of this waveguide corresponds to the width of the imaged UV stripe and was $d = 20 \mu$m along the c-axis. With our read-out beam, which is polarized in the c direction, we can excite only TM modes. For this simplified model, the total number of guided TM modes can be calculated with the cutoff condition for the N-th mode($N=1,2,3,...$) [66]

$$ (N - 1) \frac{\pi}{2} = k \frac{d n_2}{2 n_3} \sqrt{n_3^2 - \bar{n}_3^2} \equiv V, $$

where $n_2$ and $n_3$ are the refractive indices for light polarized in the b and c direction inside the waveguide (core). The parameters with an overbar belong to the quantities describing the cladding ($\bar{n}_i = n_i + \Delta n_i$). The symbol $d$ represents the waveguide width and $k$ the wavenumber of the probe beam in vacuum.

Applying our values into Eq. (3.2) we find, that the first two modes should be guided.

The intensity distribution inside the waveguide of the fundamental mode TM$_0$ of such a waveguide can be described by

40
\[ I(z) = I_0 \cos^2(\kappa z) \quad z \in [-d/2, d/2] \] (3.3)

where \( \kappa \) is the solution of the eigenvalue equation for TM modes [66]

\[
\tan \left( \frac{\kappa d}{2} \right) = \frac{n_2 n_3}{n_2 n_3} \sqrt{\left( \frac{2V}{k d} \right)^2 - 1}.
\] (3.4)

From Eq. (3.3) the full width half maximum of the intensity distribution can be calculated as

\[ W_{\text{FWHM}} = \frac{\pi}{2\kappa} \] (3.5)

Inserting our parameters in the above equations, the fundamental mode TM\(_0\) of our waveguide is expected to have a width of \( W_{\text{FWHM}} = 15 \mu\text{m} \), which is in excellent agreement with the measurement.

### 3.3.3 Waveguide dynamics

The build-up (\( \tau_b \)) and decay times (\( \tau_d \)) of the waveguide structures were determined by opening and closing the UV controlling light. During both processes, the crystal was illuminated with homogeneous background UV light. The times were determined by measuring the transmission of the probe beam through a small pinhole with diameter of about 100\(\mu\text{m} \) in the image plane of lens \( L_2 \) as shown in Fig. 3.2(a). The transmission through the pinhole was high only if the He-Ne beam was guided inside the crystal and the observed time constants in the transmission dynamics reflect those of the waveguide build-up and decay processes. The build-up of the waveguide structure is therefore characterized by the build-up of the monitored intensity, which can be described with a semi-heuristic double exponential function of the form

\[ I = I_1(1 - e^{-t/\tau_1}) + I_2(1 - e^{-t/\tau_2}). \] (3.6)

A similar expression describes the decay of the waveguide structure. The fast component \( I_1 \) carried about 80% of the intensity and was about one order of magnitude faster than the slow component \( I_2 \). Fig. 3.5(a) shows the fast component of the waveguide formation and decay processes as a function of the controlling UV-intensity.

The build-up time (\( \tau_b \)) is the time needed for screening the applied electric field, which is attained by moving charges out of the illuminated region and forming a space-charge field pointing against the initially applied field. In the first approximation the response scales with the dielectric response time.
\[
\tau_b = \frac{\varepsilon \varepsilon_0}{\sigma_{ph}} = \frac{\varepsilon \varepsilon_0}{\epsilon \mu_e n + \epsilon \mu_h p},
\]

(3.7)

where \(\varepsilon\) is the low-frequency dielectric constant, \(\varepsilon_0\) is the permittivity of free space, \(\sigma_{ph}\) is the photoconductivity induced by the UV illumination, defined by the mobilities \(\mu_e\) and \(\mu_h\) and the densities \(n\) and \(p\) of free electrons and holes respectively, and \(e\) is the elementary charge. Eq. (3.7) allows to estimate the induced photoconductivity with the measured build-up times. At UV intensities of 10 mW/cm\(^2\) we get a photoconductivity of about \(\sigma_{ph} = 10^{-8} (\Omega m)^{-1}\). This is the same order of magnitude as the photoconductivity measured in undoped near-stoichiometric LiTaO\(_3\) (\(\sigma_{ph} = 3 \times 10^{-8} (\Omega m)^{-1}\)) [33].

<table>
<thead>
<tr>
<th>UV Controlling Intensity (I_{UV}) (mW/cm(^2))</th>
<th>Time (ms)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>1000</td>
<td>1000</td>
</tr>
</tbody>
</table>

\[\tau_b \propto I_{UV}^{-0.5}\]

<table>
<thead>
<tr>
<th>UV Background Intensity (I_{BKG}) (mW/cm(^2))</th>
<th>Decay Time (ms)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>50</td>
<td>50</td>
</tr>
</tbody>
</table>

\[\tau_d \propto I_{BKG}^{-0.5}\]

**Figure 3.5:** a) Build-up times \((\tau_b)\) and decay times \((\tau_d)\) of the light induced waveguides as defined in Eq. (3.6) for different controlling UV-intensities at the crystal surface. During the decay the crystal was still illuminated with the background UV light of \(I_{BKG} = 7\) mW/cm\(^2\). b) Decay times for different background UV intensities for a fixed controlling light intensity \(I_{UV} = 100\) mW/cm\(^2\).

For higher writing intensities, more mobile charge carriers are excited which screen the applied electric field and the screening becomes faster. For band-to-band excitation, the charge densities \(n\) and \(p\) are equal and grow with the square root of the light intensity [8]. According to Eq. (3.7), the build-up times are then inverse proportional to the square root of the controlling UV light intensity \(\tau_b \propto (I_{UV})^{-0.5}\). Our measured build-up times show also this proportionality as shown in Fig. 3.5(a) by the solid line. Therefore, we conclude that the main process for inducing the mobile charge carriers is the interband photorefractive effect where the charges are excited directly from one band into the other by the illumination.

The build-up time at UV intensities of 100 mW/cm\(^2\) was about 10 ms. This is one order of magnitude slower than the build-up of light-induced waveguides in KNbO\(_3\) at
3.3 Results and discussion

\[ \lambda = 364 \text{ nm} \] for the same intensity. The absorption constant of KNbO\(_3\) at that wavelength is \( \alpha_z = 540 \text{ cm}^{-1} \), which is comparable to the absorption constant \( \alpha_z = 690 \text{ cm}^{-1} \) in SLT:Mg at 257 nm \[41\].

In SLT:Mg the decay times are approximately constant for all UV intensities of the controlling light in contrast to KNbO\(_3\) where an increase of the decay times was observed \[41\]. The decay time \( \tau_d \) depends only on the background intensity. Fig. 3.5(b) shows the fast decay times for different background intensities for controlling intensity of about 100 mW/cm\(^2\). We found that the decay time is inverse proportional to the square root of the background intensity (\( \tau_d \propto I_{BKG}^{-0.5} \)). This time dependence can be explained by the fact that the waveguide structures are deleted by the uniform background UV light and that the main process again is the interband photorefractive effect.

3.3.4 Quasi-fixing of waveguides and double waveguides

An interesting effect was observed without illuminating the crystal with the UV background light, as illustrated in Fig. 3.6. On the top of Fig. 3.6 the signal monitored by the photodiode is shown. We distinguish six processes (a) to (f) that are indicated with dashed arrows in the top of Fig. 3.6, and schematically presented with the refractive index/electric field profiles on the bottom of Fig. 3.6. These 6 processes are the following:

(a) **Initial state:** After applying an electric field of about 5.5 kV/cm. The refractive index decreased uniformly over the whole crystal according to Eq. (3.1).

(b) **Waveguide build-up and quasi-fixing:** When illuminating the crystal with the UV stripe, the applied electric field is screened by the photoexcited charge carriers, changing the refractive index back to its original value in the illuminated region and a waveguide was formed as theoretically expected. If we blocked the UV controlling light, the waveguides remained partially stable. We attribute this behaviour to the same mechanism responsible for the formation of the quasi-fixed holographic grating discussed in section 3.3.1. Beyond the edges of the UV stripe, the UV light intensity is weak, therefore the charges can reside in deep traps. The evolution of the quasi-fixed waveguide under continuous readout is shown in Fig. 3.7(a). The fixed structure was stable in the dark over several days, while it decayed under permanent readout with He-Ne light within 10 hours, similar to the decay of the quasi-fixed photorefractive gratings. The fixed structures could be deleted by homogeneous illumination with lower wavelength light (\( \lambda \lesssim 360 \text{ nm} \)). This explains why this fixing was not observed when using the background light with wavelength \( \lambda = 257 \text{ nm} \).

(c) **Widening of the structure:** Provided that the crystal is illuminated with the controlling UV stripe further on and the background UV illumination is still absent, the waveguide can widen until roughly the point where the photoconductivity induced by the
Light-induced waveguides in Mg-doped near-stoichiometric LiTaO$_3$

Figure 3.6: Top: Peak intensity of the read-out beam at the crystal output surface as a function of time showing the dynamics of waveguide formation in SLT:Mg when the background UV illumination was absent. Bottom: Schematic illustration of the processes (a)-(f) responsible for the observed dynamics. The solid lines represent the change of index profile $n$, the dashed lines the electric field $E$. The thin lines indicate $n = n_0$ and $E = 0$, which are the initial values. The dotted arrows point towards the direction of the electric field, while the solid arrows indicate the enlargement direction of the structure. Detailed explanations of the processes (a) to (f) are given in the text.
3.4 Conclusions

controlling beam equals the dark conductivity of the material. If the dark conductivity is very weak, as is the case for LiTaO$_3$ ($\sigma_{\text{dark}} = 1.5 \times 10^{-15}\Omega^{-1}\text{cm}^{-1}$) [33], already a weak stray light can contribute to a strong widening of the photoinduced waveguide structure. Due to this widening, the probe beam was not guided anymore, which is indicated by a decrease of the monitored signal at the photodiode. Since the widening was induced by week stray light, this process was much slower than the build-up of the waveguide. In the case of the presence of background illumination, the widening is avoided due to the artificial increase of the homogeneous background conductivity.

(d) Quasi-fixed space-charge field: When the UV controlling light is blocked, the index profile does not change, it remains quasi-fixed due to charges trapped in deep traps as explained before at stage (b). These charges generate a space charge field that screens the applied field $E_0$. Therefore, by switching off the applied electric field, this space charge field remains and the refractive index profile increases by $\Delta n$ over the whole crystal. The increase of the signal in Fig. 3.6 after blocking the UV controlling light can be explained by an UV induced small reversible change of the absorption constant at visible wavelengths in photorefractive crystals, known as light induced absorption [33,36].

(e) Double waveguide: By again illuminating with the UV stripe, mobile charges are produced which screen the remained(fixed) electric field. Following the space charge field, also the refractive index decreases to its original value $n_0$ in the screened regions. This screening is fast in the center, forming a double waveguide (Fig. 3.7(b), and slower in adjacent regions due to the similar process as described at stage (c). The build-up and decay times of this double waveguide with no external electric field were approximately the same as the build-up and decay of the initial waveguide.

(f) Screening of the fixed structure: After some time, the fixed space-charge electric field was completely screened and the refractive index changed back to the initial value $n_0$.

3.4 Conclusions

For the first time, to the best of our knowledge, light induced waveguides by interband photorefraction were produced in magnesium doped near-stoichiometric lithium tantalate. Guiding of the light was demonstrated for a 633 nm He-Ne probe beam with recording times of 10 ms at UV intensities of 100 mW/cm$^2$. The influence of the background illumination on the fast reconfigurable waveguides was demonstrated and a new fixing of the waveguide structures has been discovered: When the background UV illumination was absent, the waveguide structures remained fixed as soon as the controlling UV beam was blocked.
Figure 3.7: a) In absence of the UV background illumination, we obtained a quasi fixed waveguide after switching off the UV controlling light just after the build-up at stage (b) of Fig. 7. The figure shows the evolution of the quasi-fixed waveguide under continuous readout. b) Beam profile of the probe light exiting the double waveguide that we obtained when switching off the UV controlling light after the fast screening at stage (e) of Fig. 7.

In SLT:Mg we also demonstrated fixing of holographic gratings, which we further investigated by means of interband photorefractive measurements in the transverse readout geometry. We have shown, that the fixed structures were due to charges trapped in long-lifetime deep traps. The dark decay times were determined to be several days, while under permanent strong readout at 633 nm the structures decayed within 10 hours. The fixed grating and waveguiding structures could be deleted by illuminating the crystal homogeneously with light at wavelengths below 360 nm, in particular by the homogeneous UV background illumination.
Chapter 4

Light deflection and modulation through dynamic evolution of photoinduced waveguides†

Light induced waveguides produced by lateral illumination of a photorefractive crystal show a complex dynamic evolution upon removal of the sustaining applied electric field. Using this effect, deflection and modulation of the guided light is realized by taking advantage of the screening and counter-screening of the space charge distribution. The spot separation upon deflection can exceed 10 times the original waveguide width. Numerical simulations of the refractive index evolution and beam propagation show a good agreement with the observations.

4.1 Introduction

Waveguides can be induced dynamically in a photoconductive electro-optic material by lateral illumination of the sample surface with a properly shaped control illumination in association with an applied electric field [41,67], or, in alternative, by taking advantage of the photogalvanic response of the sample [68]. In the former case, the charge redistribution induced by the control illumination leads to the screening of the applied field, which can result in a locally increased refractive index in the illuminated region. The physical mechanism is similar to the one at work for waveguide formation by longitudinally propagating photorefractive bright spatial soliton beams [69]. Past investigations have shown that the steady-state shape of the waveguide reflects essentially the one of

†The results of this Chapter have been published in Optics Express 16(21), pp.16646-16658 (2008) by G. Montemezzani as first author.
Light deflection through photoinduced waveguides

the light laterally illuminating the crystal. This is true in the case where the homogeneous conductivity of the crystal approaches the photoconductivity produced by the lateral control light. If this is not true, the waveguide shape can dynamically broaden beyond the width associated with the illumination. This situation is typically found in the absence of a homogeneous background radiation in low-conductivity materials. An example is LiTaO$_3$, for which an interesting waveguide-splitting phenomenon was recently discovered (see Chapter 3). After the broadening of the waveguide, the removal of the applied field leads to the splitting of the guided light into two lobes outside the area originally occupied by the primary guide. This effect was qualitative explained in terms of a re-screening of a space-charge distribution created by the initial screening of the applied field (double screening) [67].

One of the most attractive features of the photorefractive nonlinearity is the possibility for optical steering or modulation of a light beam. Light deflection and modulation via the photorefractive effect has been demonstrated in the past by different techniques, most of which rely on dynamic holography [70–74]. Variable light deflection was obtained by isotropic [70] or anisotropic diffraction [71] at a photorefractive Bragg grating tuned by a change of the recording wavelength. In alternative, a self Bragg matched configuration based on a double color pumped oscillator was also demonstrated [72]. Deflection of the reflected light was observed in sillenite crystals as a result of space-charge waves [73]. A photorefractive all-optical deflection switch was proposed in Ref. [74] by simultaneous interference of two pairs of waves and control of their relative phases (duplex two-wave mixing). Non-holographic beam deflection was demonstrated in He$^+$-implanted strontium barium niobate planar waveguides through the influence of a self-focused pump beam on a weaker probe beam [75]. In this case the effect was attributed to a combination of thermal and photorefractive effect. The lateral motion at constant velocity of a self-focused beam across a photorefractive crystal was also shown to give rise to strong beam deflection as a result of the inertia of the refractive index change dynamics [76].

In this work we study the above mentioned waveguide splitting process in detail using the two low conductivity photorefractive crystals Sr$_x$Ba$_{1−x}$Nb$_2$O$_6$ (SBN) and LiTaO$_3$. Upon removal of the electric field used to sustain the generation of the dynamic photoinduced waveguide, the light originally propagating in the waveguide is expelled from this region. We show that the double screening phenomenon mentioned above leads to a novel type of dynamic light deflection that may be controlled by adjusting different parameters. Among these the homogeneous background conductivity and the value of the electric field used during the waveguide recording step are the most important. Together with the shape of the lateral illumination, the background conductivity defines the initial conditions for the splitting process (the initial refractive index profile). The applied field
controls the refractive index contrast and, together with the photorefractive response time, has a direct influence on the speed at which the light "expulsion" takes place. The experimental observations are qualitatively confirmed by numerical simulations of the beam propagation based on a simplified model for the dynamic evolution of the refractive index distribution. This model can explain the exceptionally large light "expulsion" distance observed in SBN. For this crystal the waveguide recording light is only weakly absorbed and the light reflected from the back surface of the crystal contributes in the definition of the shape of the refractive index distribution. Finally, it is shown that, using a periodically modulated applied electric field, a periodic switching of the output positions of the guided beam can be obtained under constant optical illumination. This new deflection-type switching is electrically controlled and requires the shaped control light merely as a kind of catalyst.

4.2 Experimental technique

Light induced waveguides were formed using a set-up similar to the one given in Chapter 3 except for the absence of the homogeneous background illumination. Fig. 4.1 gives a simplified scheme of the set-up. The control illumination defines the region and shape of the primary waveguide through the combination of the mask and the imaging optics. The probe wave guided in this guide is observed by the CCD camera at the output face of the crystal. The experiments presented in this work were performed with the

Figure 4.1: Simplified scheme of the experimental set-up. L1, L2: spherical lenses; CL1, CL2: cylindrical lenses; ND: neutral density filter. A voltage $U$ is applied to the photorefractive crystal.
electro-optic crystal strontium barium niobate (Sr$_{x}$Ba$_{1-x}$Nb$_2$O$_6$, x=0.61) weakly doped with 0.02 mol% Ce, and, for comparison, with near stoichiometric LiTaO$_3$ with a Mg doping of 0.7 mol%. The dimensions of the crystals were $a \times b \times c = 10 \times 5 \times 5 \text{ mm}^3$ and $a \times b \times c = 10 \times 4 \times 3 \text{ mm}^3$, for SBN and LiTaO$_3$, respectively. The controlling light defining the waveguide shape was produced by transmission through a straight slit imaged onto the crystal and was at the wavelength $\lambda = 514 \text{ nm}$ for SBN and at $\lambda = 257 \text{ nm}$ for LiTaO$_3$. The former wavelength is moderately absorbed in SBN (with absorption constant $\alpha = 0.26 \text{ cm}^{-1}$), while the latter is strongly absorbed in LiTaO$_3$ ($\alpha \approx 690 \text{ cm}^{-1}$ [64]). An electric field was applied for both materials along the $c$-axis of the crystal in order to induce the waveguides in combination with the lateral control illumination. The direction of the field was such as to reduce the refractive index, what is generally known as the focusing case. The probe light was in both cases at $\lambda = 633 \text{ nm}$ and was polarized parallel to the crystal $c$-axis. Its propagation direction was along the longest direction for both crystals and its intensity was low, such that this wave was not influencing the waveguide formation. We concentrate in this work exclusively on the behavior observed upon removal or modulation of the electric field after a light induced waveguide was already formed by the process described in Refs. [41, 67] and reached steady-state. The applied electric field is therefore the only quantity that is varied during our measurements, i.e., we kept constant illumination conditions of the controlling light and constant coupling conditions for the red probe wave.

**Figure 4.2:** Waveguide splitting and relaxation upon removal of the applied electric field ($E_0 = 4 \text{ kV/cm}$) in the SBN crystal. The initial photoinduced waveguide was recorded to steady state using 514 nm light. The local intensity was 125 mW/cm$^2$ in the 25 µm wide imaged slit on the crystal lateral surface. The lateral illumination is maintained after removal of the applied field. The width of the imaged area is 410 µm.
4.3 Beam splitting and deflection upon field removal

Figure 4.2 shows an example of the observed dynamics of the guided light exiting the SBN nonlinear crystal (back surface of the crystal imaged to a CCD camera). After the applied electric field is removed the light splits into two lobes. The corresponding deflection angle varies with time and reaches a maximum before relaxing towards the center. In its final stage the condition for natural diffraction is reached (absence of a waveguide). In this particular case the width of the one-dimensionally-guided wave before removal of the electric field was 16 $\mu$m (full-width at half maximum (FWHM) at the output crystal surface).

Figure 4.3 shows the position of the "center of mass" of the two lobes as a function of time. All measurements were taken after a waveguide was recorded during the same time and at the same intensity of the controlling light. The only difference between the three curves is the value of the electric field $E_0$ applied during waveguide recording. The initial fast displacement speed becomes gradually slower before inverting its direction; the system relaxes then towards the conditions were no waveguides longer exist. The maximum beam displacement is larger when the field $E_0$ was larger.

![Figure 4.3: Position of the "center of mass" of the expelled light lobes as a function of time. The initial photoinduced waveguides were recorded in SBN during 10 seconds to steady-state. The local intensity was 375 mW/cm$^2$ in the 25 $\mu$m wide imaged slit. The curves differ by the field $E_0$ applied during waveguide recording and removed at time $t = 0$. Red solid line: $E_0 =$ 4 kV/cm, blue dotted line: $E_0 =$ 2.4 kV/cm, green dashed line: $E_0 =$ 1.6 kV/cm.](image)

In the case of SBN shown here, the total maximum separation between the two lobes is about 235 $\mu$m for a field of $E_0 =$ 4 kV/cm. Note that this displacement largely exceeds the width of the original projected waveguide (25 $\mu$m), as well as the width of the
Light deflection through photoinduced waveguides

naturally diffracting probe wave at the output of the crystal in absence of a waveguide (FWHM ≈ 57 µm). Similar experiments performed in LiTaO₃ give generally smaller displacements of the light spots. As an example, Fig. 4.4 shows the near field image of the output surface of the LiTaO₃ crystal for the moment of maximum lobe distance. At the brighter spot position the separation is ≈ 92 µm, which is only slightly wider than the width of the naturally diffracted probe beam. It can be noticed that in this case the observed spots are slightly oblique. The spot distance increases to ≈ 105 µm closer to the surface illuminated by the UV control light and diminishes to ≈ 77 µm by moving down by about 70 µm. The reason for the appearance of these oblique spots will be discussed later.

We observe the exceptionally large light "expulsion" distance in SBN even though the guided light is still relatively well confined at the moment where the field is removed (see Fig. 4.2). This is different than the situation generally encountered in LiTaO₃, where, in absence of a background illumination, the probe light is no longer well guided at steady-state prior to field removal as explained in Chapter 3. A more complex refractive index structure seems therefore to be responsible for our observations in SBN. We will discuss these aspects in detail in the next section.

Figure 4.4: Snapshot of waveguide splitting observed at the output surface of a LiTaO₃ crystal for the moment of maximum lobe separation. The electric field applied during recording of the primary waveguide was 5.5 kV/cm and the primary waveguide width was 20 µm. The UV control illumination comes from the top.

4.4 Modeling of beam propagation

4.4.1 SrₓBa₁₋ₓNb₂O₆

In order to elucidate better the above anomalous waveguide splitting behavior, we have performed numerical simulations of the probe beam propagation by the beam propagation
4.4 Modeling of beam propagation

In our model the one-dimensional refractive index profile is assumed to be invariant along the propagation direction $z$, but varies in time. Just after the removal of the electric field, the refractive index profile is assumed to possess the distribution

$$\Delta n(x) = \Delta n_0 \frac{I(x)}{I(x) + I_D}, \quad (4.1)$$

which applies to the case where the conventional photorefractive effect is at work. Here $\Delta n_0$ is the refractive index change amplitude proportional to the applied electric field according to $\Delta n_0 = n^3 r E_0 / 2$, where $n$ is the average refractive index, $r$ is the active effective electro-optic coefficient and $E_0$ is the field applied during waveguide recording. The quantity $I(x)$ is the spatially dependent intensity of the control light generating the waveguide. $I_D$ is the dark intensity [78], that is the equivalent intensity producing the same density of mobile carriers than the one giving rise to the homogeneous (average) conductivity of the crystal. Note that the dark intensity can be related to a combination of the dark conductivity and the photoconductivity due to a homogeneous background irradiation of the crystal, if the latter exists (see Ref. [79] for a detailed discussion). In the following the quantities $I(x)$ and $I_D$ will be normalized in such a way that $I = 1$ corresponds to the intensity of the controlling light on the crystal in the center of a perfectly imaged slit. Expression (4.1) corresponds to the refractive index distribution due to steady-state screening of the applied field after formation of the primary waveguide, shifted up by the value $\Delta n_0$ as a result of the removal of the field $E_0$.

It was shown earlier, that a sufficiently high internal space-charge field opposite to the spontaneous polarization can induce ferroelectric domain reversal leading to the fixing of spatial soliton waveguides in SBN:75 crystals [80]. In our SBN:61 crystal, this kind of waveguide fixing was not observed for the range of parameters and for the experimental procedure used in our investigations. Therefore we can safely neglect the effect of domain reversal in the description of the dynamic evolution of our photoinduced waveguides. In this framework, the relaxation of the refractive index distribution upon removal of the applied electric field can be represented in the simplest approximation as

$$\Delta n(x, t) = \Delta n_0 \frac{I(x)}{I(x) + I_D} \exp \left[-t \left(I(x) + I_D\right)\right], \quad (4.2)$$

Here the dimensionless time $t$ has been normalized to the value of the photorefractive response time $\tau_{PR}$ for a normalized intensity equal to 1; $\tau_{PR}$ depends principally on the conductivity and on the dielectric properties of the crystal [81]. The physical model underlying Eqs. (4.1) and (4.2) completely neglects all charge carrier diffusion effects. It assumes a fully local response dynamics, for which the local relaxation time is inversely proportional to the local total intensity [82]. Despite for the simplifying assumptions,
this model maintains the most essential physical features and, as seen below, has the merit to reproduce the experimental observations in a satisfactory way.

A crucial aspect for the implementation of the model calculations is the choice of the light intensity distribution $I(x)$. In a first step we have performed several simulations by associating $I(x)$ solely with the control illumination directly incident on the region of the photoinduced waveguide (the green light propagating from bottom-left to top-right in Fig. 4.1). In the case of SBN we were never able to reproduce the observed behavior sufficiently well using a physically sound set of parameters. It appears therefore evident that the directly incident wave alone cannot lead to the strong repulsion observed. Our choice is therefore to consider an incoherent superposition of two components, $I(x) = I_1(x) + I_2(x)$. The first, $I_1(x)$, is the control light directly incident from outside. This is the wave discussed above that is intended to produce the narrow primary waveguide. We take the simplifying assumption that the slit defining the waveguide is perfectly imaged at some plane inside the crystal and that the waveguide is being probed very close to this plane, in the Fresnel diffraction regime of the imaged slit. The normalized intensity $I_1(x)$ can then be expressed as

$$ I_1(x) = \frac{1}{2} \left( [C(X_2) - C(X_1)]^2 + [S(X_2) - S(X_1)]^2 \right), \quad (4.3) $$

where $C(X)$ and $S(X)$ are the Fresnel cosine and sine integrals, respectively. The integration limits are $X_2 = \sqrt{2/(Xd_1)}(x+a)$ and $X_1 = \sqrt{2/(Xd_1)}(x-a)$, where $2a$ is the width of the imaged slit on the crystal, $X' = \lambda/n$ is the light wavelength in the medium, and $d_1$ is the distance between the waveguide probing depth and the depth of ideal imaging. The second component, $I_2(x)$, consists of the same light after propagating inside the crystal to the other lateral surface and being reflected back towards the region of the waveguide. In most cases this reflected light is already in the Fraunhofer diffraction regime. The normalized intensity $I_2(x)$ can be calculated as well using Eq. (4.3), by replacing the distance $d_1$ with the total path distance $d_2$ from the ideal plane of perfect imaging to the back lateral surface, and back to the waveguide. In addition, the intensity $I_2(x)$ of the back reflected light has to be further normalized by a factor

$$ K = \left( \frac{1-n}{1+n} \right)^2 \exp(-\alpha d_2), \quad (4.4) $$

that takes into account the Fresnel reflection and absorption of the controlling light (absorption constant = $\alpha$).

The light distribution $I(x) = I_1(x) + I_2(x)$ is depicted in Fig. 4.5(a) for a set of parameters proper to our crystal sample and experiments. The corresponding initial refractive index distributions according to Eq. (4.1) are shown in the same figure for three values of the
4.4 Modeling of beam propagation

Figure 4.5: (a) Light intensity $I = I_1 + I_2$ for the parameters $2a = 25 \mu m$, $d_1 = 10 \mu m$, $d_2 = 9.5 \text{ mm}$, $n = 2.33$, $\alpha = 0.26 \text{ cm}^{-1}$; (b) Refractive index distribution just after removal of the applied field for $I_D = 0.005$ (red solid curve), $I_D = 0.1$ (blue dotted curve), and $I_D = 0.0001$ (green dashed curve).

normalized dark intensity $I_D$. The value $I_D = 0.005$ is consistent with the observed ratio between the formation time and the dark lifetime of the photoinduced structures in our crystal. The refractive index distribution of Fig. 4.5(b) shows that in this case the initial primary waveguide sits on a wider plateau of increased refractive index induced by the back reflected control light. As seen below, the refractive index contrast between the central primary guide and the plateau is nevertheless sufficient to guide the probe wave. For larger values of $I_D$ (blue dotted line in Fig. 4.5(b)) one obtains a stronger and better confined initial central waveguide. In fact, if $I_D$ exceeds the intensity level of the back reflected control light (which is about 3% at its maximum in our case), the plateau can be almost completely eliminated. This is the concept used earlier for recording photoinduced waveguides with background illumination [41,67], which experimentally leads to a much less dramatic dynamics upon field removal. On the other hand, for small values of $I_D$ (green dashed line in Fig. 4.5(b)), the formation of the central initial waveguide over the plateau is prevented and the probe light would no longer be guided before the removal of the applied field.

With the knowledge of the intensities $I_1(x)$ and $I_2(x)$ and of the initial distribution $\Delta n(x,t=0)$, we can calculate the evolution of the refractive index profile of the waveguide using Eq. (4.2). This $z$-independent profile can then be used to calculate the propagation of the red probe wave at any time by the beam propagation method [23,77]. As an example, Fig. 4.6 shows the calculated dynamics for SBN upon removal of the applied field. The temporal evolution of the probe beam propagation is calculated for a set of parameters corresponding to those used experimentally. The maximum refractive index change $\Delta n_0 = 3.5 \times 10^{-4}$ used in the simulation can be induced by a field
Figure 4.6: Simulated time evolution of the waveguide splitting phenomenon after removing the sustaining applied electric field at the time \( t = 0 \). The left-hand side shows the propagation of the probe beam in the SBN crystal containing the splitted photoinduced waveguide with the given color scale for its intensity. The top-right diagram shows the evolution of the refractive index profile according to Eq. (4.2). The bottom-right diagram shows the profile of the probe beam intensity on the output surface of the crystal. Parameters: \( \Delta n_0 = 3.5 \times 10^{-4} \), \( I_D = 0.005 \), input waist of probe beam = 18 \( \mu m \). All other parameters as in Fig. 4.5. The time is normalized to the photorefractive response time. The movie can be downloaded at “http://www.opticsinfobase.org/oe/viewmedia.cfm?uri=oe-16-21-16646&seq=2”.

\[ E_0 = 2.4 \text{ kV/cm} \] (corresponding to the intermediate field in Fig. 4.3) via the electro-optic coefficient \( r_{333} = 235 \text{ pm/V} \) of SBN (\( x = 0.61 \)) \[83\].

Figure 4.6 shows the temporal evolution of the probe wave propagation, of the refractive index profile and of the probe wave output intensity. At the time \( t = 0 \) the refractive index profile corresponds to the one given by the solid line in Fig. 4.5(b), the index contrast between the central region and the plateau is enough to guide the probe wave, in agreement with the experiments. The following simulated time evolution shows that the probe light is rapidly expelled from the initial central waveguide, it reaches the maximum distance for a time of the order of 5 times the photorefractive response time \( \tau_{PR} \), before slowly relaxing back to a straight propagation. The calculated dynamics of the waveguide splitting process is in good qualitative agreement with the observed one (see Fig. 4.2 and Fig. 4.3). The simulated spot separation distances are consistent with those observed experimentally. We recall that the simulations are calculated under simplified ideal conditions. The strong effect of the border of the refractive index change profile associated to the central lobe of the diffracting back reflected control light
is a consequence of this simplification. In the experimental situation, any scattering stray light, as well as multiple reflections, contribute to smooth the refractive index contrast at the border of the central lobe.

Our observations of Fig. 4.3 show that the probe light splits faster and further apart if the field that is applied during the primary waveguide recording is larger. Figure 4.7 shows a snapshot of the initial splitting \( t = 0.3 \) of the probe wave for three values of \( \Delta n_0 \) corresponding to the three values of the field in Fig. 4.3. The stronger initial spatial splitting for the higher fields can be easily recognized, in agreement with the experimental observations. It has to be noted, that for materials with short diffusion lengths, such as our SBN crystal, the photorefractive response time is a decreasing function of the electric field [81]. Therefore, the same normalized time corresponds to an earlier ”real” time for the case of a large field than for the case of a low field. This effect contributes to accelerate further the dynamics for the larger fields with respect to what appears from Fig. 4.7.

![Figure 4.7: Initial splitting of the waveguide at the normalized time \( t = 0.3 \) for three values of \( \Delta n_0 \).](image)

- (a) \( \Delta n_0 = 6 \times 10^{-4} \) \( (E_0 \approx 4 \text{ kV/cm}) \);
- (b) \( \Delta n_0 = 3.5 \times 10^{-4} \) \( (E_0 \approx 2.4 \text{ kV/cm}) \);
- \( \Delta n_0 = 2.4 \times 10^{-4} \) \( (E_0 \approx 1.6 \text{ kV/cm}) \).

The other parameters are as in Fig. 4.6.

### 4.4.2 LiTaO₃

In the case of LiTaO₃ the control light is strongly absorbed and induces direct band-to-band phototransitions. For such interband processes the quadratic recombination of carriers leads to a photoconductivity which increases only with the square root of the light intensity [15]. This has an effect on the screening of the applied electric field. By assuming that the square root dependence is valid over the whole range of relevant
intensities, Eqs. (4.1) and (4.2) have to be modified, and Eq. (4.2) is replaced by

$$\Delta n(x,t) = \Delta n_0 \frac{\sqrt{I(x)}}{\sqrt{I(x)} + \sqrt{I_D}} \exp \left[ -t \left( \sqrt{I(x)} + \sqrt{I_D} \right) \right].$$

(4.5)

Here the same intensity normalization as above is used and the role of $I_D$ is the same as in the case of SBN. Due to the large absorption constant ($\approx 690 \text{ cm}^{-1}$ [64]), the back reflected control wave $I_2(x)$ does not play any role in the case of LiTaO$_3$. Nevertheless, in the absence of a background illumination a significant widening of the primary waveguide during recording (with field on) is observed, as was discussed in [67]. This widening is due to control light surrounding the ideal imaged slit defining the waveguide (stray light). A combination of diffraction effects (described in first approximation by the function $I_1(x)$ of Eq. (4.3)) and of scattering effects due to bulk defects or surface imperfections (more difficult to describe mathematically) is expected to contribute to the stray light producing the widening. Even though the intensity level of the scattering might be very small, it can lead to quite strong widening because of the very low levels of the dark intensity $I_D$ proper of LiTaO$_3$ under UV illumination. The quantity $I_D$ can be estimated by considering the depth until which a holographic grating can be recorded by the UV light. This depth corresponds roughly to the position where $I/I_D = 1$ and is of the order of 250 $\mu$m [67] for crystals similar to the one used in our measurements. Using the above absorption constant we can then estimate a ratio of the order of $I/I_D = 10^5$ at the depth of 80-90 $\mu$m of our waveguides. The main reason for the very small value of $I_D$ is the high quantum efficiency connected with the interband phototransitions. Below, we will discuss some simulations of the probe beam propagation in LiTaO$_3$ by considering the refractive index evolution given by Eq. (4.5). For simplicity we neglect the effect of scattering and limit the description of the stray light to the diffractive component by using the expression (4.3) for the control light intensity, always with a non vanishing value for the distance $d_1$.

Figure 4.8(a) shows the calculated propagation of the probe wave in the stationary state just before the applied field is removed. By considering the electro-optic response of LiTaO$_3$ [61], the refractive index amplitude $\Delta n_0$ used for the simulations corresponds to an applied field of 5.5 kV/cm. The wave is only badly guided due to the fact that the waveguide has suffered a significant widening, as seen by the refractive index distribution in Fig. 4.8(c), which applies to the same situation. Fig. 4.8(b) shows the same propagation but for a normalized time $t = 5$ after removing the applied electric field, which corresponds roughly to the moment of maximum separation. Figure 4.8(d) shows the refractive index distribution at the same time. As in the case of SBN, we observe the splitting of the propagating wave into two lobes. However, the calculated separation distance between the lobes is smaller than in SBN, which is in agreement with the
Figure 4.8: Model calculations for the case of LiTaO$_3$ using Eqs. (4.3) and (4.5) with the parameters $\Delta n_0 = 8.8 \times 10^{-5}$, $2a = 20 \mu$m, $n = 2.72$ and a probe beam input waist of 15 $\mu$m. (a) Probe beam propagation in the primary waveguide widened to steady-state at $t = 0$ for $I_D = 10^{-5}$ and $d_1 = 45 \mu$m. (b) Same as (a) but for the time $t = 5$ after splitting of the waveguide. (c) and (d) Refractive index profiles for case (a) and (b), respectively. (e) Output probe beam intensity profile at three depths for $t = 5$. The central diagram corresponds to the case (b). The upper diagram to a position 35 $\mu$m closer to the surface ($I_D = 8.9 \times 10^{-7}$, $d_1 = 10 \mu$m). The lower diagram to a position 35 $\mu$m deeper ($I_D = 1.1 \times 10^{-4}$, $d_1 = 80 \mu$m). The dashed green line is a guide for the eye evidencing the relation with the spot obliquity of Fig. 4.4.

Observations in the two crystals (compare Fig. 4.2 and Fig. 4.4) and with the model simulations for SBN (Fig. 4.6).

As discussed earlier, the near field experimental observations in LiTaO$_3$ (Fig. 4.4) show that the two lobes are slightly tilted with a larger separation closer to the entrance surface of the control light. Due to the large absorption, a change in depth is connected to a change of the normalized dark intensity $I_D$. We discuss the lobe tilt with the help of Fig. 4.8(e), where we have calculated the output intensity profile of the probe beam for three different values of $I_D$, corresponding to three depths mutually separated by 35 $\mu$m. Together with $I_D$, we have changed also the value of the imaging mismatch $d_1$, increasing it from 10 $\mu$m for the top diagram in Fig. 4.8(e), to 45 $\mu$m for the middle one, and to 80 $\mu$m for the bottom one. In general, larger values of $d_1$ lead to wider intensity distributions that may lead to bigger spot separations. However, our simulations have shown that, for the range of intensity-to-dark-intensity ratio in which we operate experimentally, the
change of the parameter $d_1$ has only a very small influence on the spot obliquity with respect to the influence of the normalized $I_D$. Indeed the simulations of Fig. 4.8(e) correctly predict a larger spot separation for the top diagram (see dashed green helping line) that corresponds to the layer closer to the surface. This larger separation is due to the stronger widening occurring for the larger values of the ratio $I/I_D$. The calculations predict a decrease of the separation by about 25-30 $\mu$m for a depth increase of 70 $\mu$m, which is in quite good agreement with the observations of Fig. 4.4, the absolute value of the spot separation is matched within roughly 20% between experiments and simulations. It is worth noticing that, if we perform the simulations using Eq. (4.2) instead as with Eq. (4.5), we obtain a much stronger dependence of the spot separation on the normalized value of $I_D$. The spot obliquity predicted by Eq. (4.2) is nearly doubled, what prevents to match the experimental data. This proves the necessity to take into account the square root dependence of the photoconductivity on the light intensity in the model.

4.5 Modulation of the transmitted light by waveguide anti-waveguide alternation

Since the removal of the electric field prevents the probe light to remain guided in its original photo-induced waveguide, we can use this effect for a novel light modulation technique. The application of a periodic high-voltage to the crystal leads to a waveguide and an anti-waveguide appearing cyclically at the place illuminated by the shaped control

![Figure 4.9: Modulation of the output position of the probe beam under a periodic electric field. A triangularly shaped electric field with amplitude 2 kV/cm and frequency 1 Hz is applied to the crystals. The resulting waveguide-antiwaveguide alternation results in the periodically varying output position of the probe wave. The two frames shown above are separated by half a period, the corresponding displacement between the two positions is about 90 $\mu$m. SBN crystal, controlling wave intensity = 0.4 W/cm². The width of the imaged area in the multimedia file is 480 $\mu$m. The movie can be downloaded at “http://www.opticsinfobase.org/oe/viewmedia.cfm?uri=oe-16-21-16646&seq=3”.

60
4.5 Waveguide anti-waveguide alternation

light. The modulation occurs through the coupling of the spatial position of the output beam to the electric field, without changing any of the illumination conditions. Figure 4.9 shows an example of this modulation as obtained in SBN under a periodic applied field of triangular shape. Note that in the experiment of Fig. 4.9 we have deliberately slightly misaligned the incidence direction of the probe wave with respect to the axis of the waveguide. In this way, upon removal of the field, the light is expelled only on one side of the waveguide. In the example given in Fig. 4.9, when considering the conditions separated by half a voltage cycle, the output position of the probe beam shifts by roughly 90 $\mu$m. When repeating the same kind of experiments with LiTaO$_3$ similar results are obtained, however the observed shift is of the order of 40 $\mu$m in this case.

![Figure 4.10: Waveguide modulation dynamics for LiTaO$_3$ as measured by transmission of the probe wave through a pinhole (red thick line). The modulated applied field is shown by the thin blue line. a) 0.3 Hz modulation, b) 5 Hz, c) 10 Hz.](image)

In order to better characterize the dynamics of the effect, we have inserted a small pinhole in the image plane of the crystal’s output surface and detected the transmission through this pinhole by a photodiode. Figure 4.10 shows an example of this time dependence for the case of LiTaO$_3$ and three different frequencies of the applied field. The diameter of the measuring pinhole was one half the nominal width of the photoinduced waveguide. The intensity of the incident UV controlling light at the surface of the crystal was 1.1 W/cm$^2$. Considering that the waveguide was probed at an average depth of $\approx 90 \mu$m, the local UV intensity in the probed region was $\approx 2$ mW/cm$^2$. The dynamics of Fig. 4.10 is following the particular form of the voltage delivered by the high voltage power supply being used. Nevertheless, at the two higher frequencies, a slight delay between the maximum of the applied voltage and the maximum of the transmitted intensity can be recognized. In these conditions, the field can lead to an increase of the waveguide strength even at the beginning of its decreasing half period. When compared to the period $T$ of the applied field, at the above intensity the resulting retardation between the red and blue curve is roughly $0.15 \times T$ for $T = 100$ ms, $0.04 \times T$ for $T = 335$ ms, while it becomes
nearly irrelevant at lower frequencies. As can be expected due to a slower dynamics, this retardation increases if the controlling light intensity is reduced. For instance, for a 40 times lower intensity we get a phase retardation of $0.35 \times T$ at $T = 100$ ms. The maximum modulation amplitude of the transmitted probe light is found for the above experimental conditions in LiTaO$_3$ when the applied field period is in the range of 200–350 ms.

For similar experiments performed using SBN we obtain essentially the same kind of results as those depicted in Fig. 4.10. Here the optimum modulation amplitude is found for an applied electric field period of the order of 1 second (incident controlling light intensity = 90 mW/cm$^2$). At this intensity the retardation discussed above goes from $0.2 \times T$ at $T = 400$ ms down to $0.002 \times T$ at $T = 10$ s. It should be noted that in SBN, due to the much lower absorption of the controlling light, the observed dynamics is much less dependent on the probing depth below the surface as in the case of LiTaO$_3$.

4.6 Conclusions

The evolution of light induced one-dimensional waveguides when the sustaining electric field is switched off leads to a novel kind of dynamic light deflection. We have studied this effect in detail using SBN and LiTaO$_3$ crystals. Simulations of the beam propagation in the split waveguide on the base of a simple model lead to a good agreement with the experimental observations. In the case of SBN the spot separation can exceed the original waveguide width by 10 times. It is found that the control light back reflected from the second lateral surface is responsible for the additional waveguide widening, which leads to this large deflection distance. In LiTaO$_3$ the primary waveguide widening is due solely to stray light and gives smaller deflection distances. In this material the strong absorption constant leads to oblique spots after waveguide splitting which have been successfully modeled. Application of a modulated electric field leads to the modulation of the transmitted probe light as a result of a waveguide anti-waveguide alternation. This novel modulation method has been demonstrated for low frequencies of the applied field. Faster speeds are expected by increasing the local intensity of the controlling light. The use of materials with higher photosensitivity, such as Sn$_2$P$_2$S$_6$ or reduced KNbO$_3$, is also expected to lead to faster modulation speeds in a regime that uses the conventional photorefractive effect, which allows to obtain a waveguide dynamics being virtually independent from the observation depth.
Chapter 5

Fast dynamic waveguides and waveguide arrays in photorefractive Sn$_2$P$_2$S$_6$ induced by visible light$^\dagger$

We report on dynamic waveguides and waveguide arrays induced beneath the surface of electro-optic Sn$_2$P$_2$S$_6$ crystals by visible light at 514 nm. The waveguide structures are generated by interband photoexcitation and drift or diffusion charge transport mechanism. These structures are probed nondestructively in the transverse direction with a beam at a longer wavelength. We measured the fastest formation of light induced waveguides in the visible up to now. The recording times are below 200 µs for intensities above 0.1 W/cm$^2$. By interfering two light beams, dynamic waveguide arrays are generated with waveguide spacings of 7µm. If an electric field is applied to the crystal, these arrays can be spatially shifted by 1.5 µm for an applied field of $E_0 = 1$ kV/cm.

5.1 Introduction

Waveguides are the basic elements in integrated optical applications [56]. Many of these applications like optical switching, routing or dynamic optical interconnections require fast switching between waveguide channels. This is normally done electro-optically by applying complicated electrode configurations on permanently structured waveguides [75, 84]. Several techniques based on light-induced refractive index changes have been recently proposed [20, 41, 57, 58, 67, 85]. The most interesting feature of these waveguides is that they can be dynamically reconfigured, and thus signals can be routed by solely changing the light illumination.

$^\dagger$The results of this Chapter are in peer review (2008).
Periodic dielectric structures such as photonic crystals or photonic lattices have recently gained a lot of interest due to their exciting features like controlling and manipulating the propagation and manage the diffraction of optical beams [20, 29, 86]. In photorefractive crystals, a periodic modulation of the refractive index can be induced dynamically by interfering two or more light beams inside the crystal. Beam steering in such a 1 dimensional optically induced lattice has recently been demonstrated using a third controlling beam [87]. For investigations of light propagation in periodic lattices, strontium-barium-niobate (SBN) is the most extensively employed material due to its high electro-optic activity ($r_{33} = 235 \text{ pm/V}$ for SBN:60, $r_{33} = 1340 \text{ pm/V}$ for SBN:75 at $\lambda = 0.5 \mu\text{m}$) [21]. However, the photorefractive response times of SBN in the visible are in the order of a few seconds [21], which makes this material not suitable for applications, where short waveguide formation times are required.

In this work, we investigate the potential of tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$) for dynamic waveguide applications. Sn$_2$P$_2$S$_6$ is a semiconducting ferroelectric material with interesting optical and nonlinear optical properties: high photorefractive efficiency in the infrared up to the telecommunication wavelength $1.55 \mu\text{m}$ [2–4] and a large electro-optic coefficient ($r_{111} = 174 \text{ pm/V}$ at 633 nm [5]). Furthermore, the photorefractive response of Sn$_2$P$_2$S$_6$ in the near-infrared is very fast, more than two orders of magnitude faster than in any other photorefractive ferroelectric crystal as e.g. Rh-doped BaTiO$_3$ [88]. Recently, photorefractive self-focusing at 1.06 $\mu\text{m}$ was demonstrated in bulk Te doped Sn$_2$P$_2$S$_6$ crystals with 15 ms response time at peak intensities of 160 W/cm$^2$ [89].

The photorefractive response time can be decreased, if light with photon energy larger than the band gap of the material is used [15]. In this so-called interband photorefractive effect, refractive index structures can be generated by charge redistribution between the bands, which in general provides 2–3 orders of magnitude faster response than the conventional effect.

So far, light induced waveguides were demonstrated in KNbO$_3$ [41] and Mg doped LiTaO$_3$ [67] by interband photorefraction with controlling light at ultraviolet (UV) wavelengths. However, there are some drawbacks of using UV light such as availability of laser sources, need of special optical elements and coatings to mention a few. Sn$_2$P$_2$S$_6$ has a band gap energy of $E = 2.3 \text{ eV}$, which is lower than in conventional photorefractive crystals and enables interband photorefraction already in the visible at $\lambda = 514 \text{ nm}$ [6].

In this work we show that fast reconfigurable waveguides and waveguide arrays can be induced beneath the surface of Sn$_2$P$_2$S$_6$ crystals by using band-to-band excitations. The waveguiding structures are written beneath the surface in regions illuminated by 514 nm light by drift or diffusion of charge carriers, dominated by hole charge transport. They are probed nondestructively in transverse direction.
5.2 Light induced waveguides

The experiments were performed in a 6.8 mm long Sn$_2$P$_2$S$_6$ crystal oriented as shown in Fig. 5.1. The sample was nominally pure, to minimize the possibility of deep level trapping [15]. The use of interband light provides a faster effect, but on the other hand also a higher absorption for the controlling light. The absorption in Sn$_2$P$_2$S$_6$ is $\alpha = 490 \, \text{cm}^{-1}$ at the controlling wavelength of $\lambda_{\text{CL}} = 514 \, \text{nm}$ [6]. Therefore, the waveguide reaches a depth of only a few ten micrometers below the surface and the crystal needs sharp edges for in- and out-coupling of the guided light. Since the structures are written between the bands, readout at sub band-gap wavelengths as e.g. red or telecommunication wavelengths, does not disturb the waveguide structures.

The illumination of the crystal is shown in Fig. 5.1(a). The controlling light (Argon Ion Laser @ 514 nm) homogeneously illuminated a mask that was imaged onto the crystal z-surface by appropriate optics (not shown in the figure). The probe beam (HeNe @ 633 nm) traveled along the crystalline y-direction and was focused onto the input face of the crystal by a spherical lens ($f = 40 \, \text{mm}$) to a diameter of $2w_0 = 22 \, \mu\text{m}$. An out-coupling lens imaged the output face onto a CCD-camera. In order to excite an eigenmode, the readout beam was polarized along the dielectric 3-axis at an angle of $\psi = 43^\circ$ with respect to the x-axis according to the orientation of the indicatrix in Sn$_2$P$_2$S$_6$ [4, 5]. The controlling light was polarized in x-direction and an electric field was applied along x as well. For this configuration, we get an effective electro-optic coefficient of $r_{\text{eff}} = r_{111} \sin^2 \psi + r_{331} \cos^2 \psi + r_{131} \sin 2\psi = 183\, \text{pm/V}$ at 633 nm using

![Figure 5.1](image_url)

**Figure 5.1:** a) Arrangement for recording light induced waveguide structures. b) Simplified electric field (dashed red) and refractive index (solid green) distribution in a photorefractive crystal during the formation of the waveguides. Explanation is given in the text.
Light-induced waveguides and waveguide arrays in \( \text{Sn}_2\text{P}_2\text{S}_6 \)

**Figure 5.2:** a) CCD-images of the output face of a 6.8 mm long SPS crystal without (left) and with (right) a photoinduced waveguide. b) Build-up times \( \tau_b \) of the light induced waveguide as a function of the controlling light intensity.

The coordinate system as defined in [4]. Uniform background illumination at 514 nm produced a homogeneous conductivity that is needed for a better confinement of the waveguides [67].

The basic process for inducing a step index profile in photorefractive crystals is schematically shown in Fig. 5.1(b). i) is the unperturbed state with a uniform refractive index \( n_0 \). In a first step (ii) an electric field \( E_0 \) is applied, which homogeneously decreases the refractive index via the electro-optic effect to a value of \( n' = n_0 - \frac{1}{2}n_0^3r_{\text{eff}}E_0 \). Finally, the controlling light is switched on (iii) and electrons are excited to the conduction band. Free charges, electrons in the conduction band and holes in the valence band, drift and screen the applied electric field in the illuminated region. This results in an electric field pattern that is correlated to the pattern of the controlling light. Thus, a refractive index structure is produced, which has its maximum in the illuminated regions. For the simple case of a slit mask, we get a 1D planar waveguide [41,67].

Fig. 5.2(a) shows the output of a 6.8 mm long pure \( \text{Sn}_2\text{P}_2\text{S}_6 \) crystal. A straight slit was imaged onto the crystal \( z \)-surface to a width of 17 \( \mu \text{m} \) in \( x \)-direction. Compared to previous experiments in \( \text{KNO}_3 \) or \( \text{LiTaO}_3 \), much smaller fields are required due to the large electro-optic coefficient of \( \text{Sn}_2\text{P}_2\text{S}_6 \) (Table 5.1). In our experiments, the applied electric field was \( E = 900 \text{ V/cm} \), which resulted in a refractive index change of \( \Delta n = 2.3 \times 10^{-4} \) for the HeNe probe Beam. The profile of the guided light perfectly matches a \( \cos^2 \)-function, for the first waveguide-mode, with a FWHM of 12 \( \mu \text{m} \), which is in good agreement with the expected FWHM of 11 \( \mu \text{m} \) for the given index profile.
5.3 Light induced waveguide arrays

Table 5.1: Parameters for the recording of interband light induced waveguides in LiTaO$_3$ [67], KNbO$_3$ [41] and Sn$_2$P$_2$S$_6$[this work]

<table>
<thead>
<tr>
<th>Material</th>
<th>$\lambda_{CL}$</th>
<th>$n_0$</th>
<th>$r_{eff}$</th>
<th>$\alpha$</th>
<th>$E_0$</th>
<th>$\Delta n$</th>
<th>$\tau_b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiTaO$_3$</td>
<td>257 nm</td>
<td>2.18</td>
<td>31 pm/V</td>
<td>690 cm$^{-1}$</td>
<td>5.5 kV/cm</td>
<td>0.88×10$^{-4}$</td>
<td>16 ms</td>
</tr>
<tr>
<td>KNbO$_3$</td>
<td>364 nm</td>
<td>2.17</td>
<td>55 pm/V</td>
<td>550 cm$^{-1}$</td>
<td>4.8 kV/cm</td>
<td>1.34×10$^{-4}$</td>
<td>0.4 ms</td>
</tr>
<tr>
<td>Sn$_2$P$_2$S$_6$</td>
<td>514 nm</td>
<td>3.1</td>
<td>183 pm/V</td>
<td>490 cm$^{-1}$</td>
<td>0.9 kV/cm</td>
<td>2.5×10$^{-4}$</td>
<td>0.2 ms</td>
</tr>
</tbody>
</table>

$\lambda_{CL}$: Recording wavelength, $n_0$: Refractive index, $r_{eff}$: Electro-optic coefficient, $\alpha$: Absorption @ $\lambda_{CL}$, $E_0$: Electric field, $\Delta n$: refractive index change, $\tau_b$: Build-up Time at $I_{CL}$=0.1W/cm$^2$. $n_0$, $r_{eff}$, $\Delta n$ and $\tau_b$ are values for readout at 633 nm in the respective configuration.

The build-up times $\tau_b$ of light induced waveguides in Sn$_2$P$_2$S$_6$ as a function of the controlling light intensity are shown in Fig. 5.2(b). The response is very fast, with $\tau_b < 200 \mu s$ for intensities above 0.1 W/cm$^2$. As listed in Table 5.1, this is two times faster than the build-up times observed in KNbO$_3$ [41] and more than one order of magnitude faster than the build-up times measured in LiTaO$_3$ [67] for recording with the same intensity but in the UV. The build-up times were determined by recording the temporal evolution of the peak intensity of the output light. This was measured using a photodiode and a 100 $\mu$m pinhole in the image plane of the out-coupling lens. The square-root intensity dependence of the build-up times confirms the interband nature of the structure formation [15].

5.3 Light induced waveguide arrays

We further demonstrated optically induced waveguide arrays in Sn$_2$P$_2$S$_6$ at the interband wavelength $\lambda_{CL} = 514$ nm. For this we used a crystal with a length of 15 mm along the propagation direction ($y$). The array was induced by interfering two light beams that generated an interband photorefractive grating beneath the $z$-surface of the crystal. Such a structure represents a waveguide array with a waveguide spacing equal to the grating spacing $\Lambda$. In our set-up we had $\Lambda \approx 7$ $\mu$m which yielded a modulation depth in the $x$-direction of $\Delta n = 1.0 \times 10^{-4}$ for the readout light at $\lambda \approx 633$ nm, as determined by Bragg diffraction measurements [6].

Different than in the single waveguide experiments described above, the array structures could be created only by diffusion of charge carriers without the background illumination. If an additional electric field was applied to the crystal, a combination of diffusion and drift was responsible for the formation of the waveguide arrays.

We probed the array in transverse direction since the structures are written in a layer a few ten micrometers beneath the surface, similar to the light induced waveguide
Light-induced waveguides and waveguide arrays in Sn$_2$P$_2$S$_6$

experiments in the previous section. The light was coupled into the array with a 20x microscope objective. A cylindrical lens ($f = 200$ mm) was placed into the path of the probe beam, to minimize diffraction in the $z$-direction. The size of the read-out beam in the $x$ direction was $2w_x = 12\mu$m, so that the input-beam covered about two waveguides. The output after 15 mm propagation was again monitored by the CCD-camera and is shown in Fig. 5.3(a). Without the array, the beam diffracted to $2w_x = 330\mu$m. With the array, the input beam was distributed among several waveguides, as shown in Fig. 5.3(a). The build-up time of these waveguides was $\tau_b = 225\mu$s for controlling light intensity of 0.1 W/cm$^2$.

![Figure 5.3: a) Output of a 15 mm long pure Sn$_2$P$_2$S$_6$ crystal without (left) and with (right) the induced photorefractive waveguides. b) Measured profiles along the $x$-direction without an external applied electric field (red open squares) and with an applied electric field of 1kV/cm (blue solid circles). The lines are drawn for the guidance to the eyes.

An additional electric field $E_0 = 1$ kV/cm applied in the $+x$-direction of the crystal shifted the whole waveguide array by an amount of $\Delta x \approx -1.52\mu$m = $-0.21\Lambda$ (Fig. 5.3(b)), $\Lambda = 7.25\mu$m being the waveguide spacing. This can be explained by the fact, that the refractive index grating is phase shifted with respect to the light illumination. This phase shift strongly depends on the applied electric field and on the type of dominant charge carriers ($e^-$ or $h^+$). Measuring this phase shift allows us to give an estimation for the free carrier density $n_0$ responsible for the generation of the photorefractive grating, as well as for the direct recombination constant $\gamma_{dir}$ describing electron-hole recombination between the bands. For this we use the free carrier model from Ref. 15, which describes the space charge field $E_{sc}$ in the trap-free approximation valid for high light intensities. The expression for $E_{sc}$ can be further simplified for large grating spacing.
5.3 Light induced waveguide arrays

\( \Lambda \gtrsim 1 \mu m \) and an applied electric field smaller than the recombination field of the dominant charge carriers, i.e. the average internal electric field in which the charges drift for an average distance \( K^{-1} = \Lambda/2\pi \) before recombination. In our configuration this is true for \( E_0 \ll 20 \text{ kV/cm} \) considering the results of the Bragg diffraction measurements [6]. We furthermore consider only one type of charge carriers. This is eligible if one of the mobilities is much larger than the other one, i.e. \( \mu_e \gg \mu_h \) or \( \mu_h \gg \mu_e \) for electron or hole dominated charge transport respectively. This simplification yields the following relation for the space charge field:

\[
E_{sc} \approx \text{im} \frac{E_{qf}(\mp E_D + iE_0)}{2E_{qf} \mp iE_0} \tag{5.1}
\]

where \( E_D = Kk_BT/e \) is the diffusion field and \( E_{qf} = \frac{e}{\epsilon_0\epsilon} n_0 \) is the maximum electric field that can be created by free charge carriers; \( m \) grating modulation depth, \( k_B \) Boltzmann constant, \( T \) absolute temperature, \( e \) elementary charge, \( \epsilon_0 \) vacuum permittivity and \( \epsilon = 230 \) [4] the dielectric constant of Sn\(_2\)P\(_2\)S\(_6\). The upper signs in Eq. (5.1) are for electron and the lower signs for hole dominated charge transport respectively. The phase shift \( \phi \) between the waveguide array and the interference pattern can then be calculated from Eq. (5.1) and is given by:

\[
\tan \phi \approx \pm \frac{2E_{qf}E_D + E_0^2}{2E_{qf}E_0 - E_DE_0}. \tag{5.2}
\]

Taking into account the negative sign of the electro-optic effect (\( \Delta n \propto -E \)), positive and negative values of \( \phi \) correspond to dominant electron and hole charge transport respectively. The spatial shift of the array in the negative \( x \) direction after applying an electric field to the array implies a reduced negative phase shift in our experiment. This proves that the hole mobility in our Sn\(_2\)P\(_2\)S\(_6\) crystal is larger than the electron mobility, in agreement with two-wave mixing experiments at this wavelength [6].

Without applied electric field, a maximum shift of \( \phi_0 = -\frac{\pi}{2} \) is reached. After applying an electric field of \( E_0 = 1 \text{ kV/cm} \), the array is phase shifted by \( \phi_E = 1.32 \text{ rad} \). This corresponds to a phase shift of \( \phi = \phi_E + \phi_0 = -0.25 \text{ rad} \) between the array and the light fringes. Using the above definitions of \( E_D \) and \( E_{qf} \), the charge density can be estimated from Eq. (5.2) to \( n_0 \approx 2 \cdot 10^{16} \text{ cm}^{-3} \) for a writing light intensity of \( I_0 = 100 \text{ mW/cm}^2 \). Assuming a quantum efficiency close to 1, we can give an estimation for the direct recombination constant of Sn\(_2\)P\(_2\)S\(_6\): \( \gamma_{\text{dir}} = \alpha I_0/(\hbar
u n_0^2) \approx 4 \cdot 10^{-13} \text{ cm}^3\text{s}^{-1} \), with absorption constant \( \alpha \) and photon energy \( \hbar \nu \).

By applying a modulating electric field, it may be possible to modulate such arrays in real-time. The arrays may also be shifted by a modulated phase-difference between the two writing beams, which can be realized with an additional electro-optic modulator or a piezo-controlled mirror in the path of one of the writing beams, as used for moving photorefractive gratings [3].
5.4 Conclusions

We have demonstrated for the first time to our knowledge waveguides and waveguide arrays induced by band-to-band excitation at visible wavelengths. In the electro-optic material Sn$_2$P$_2$S$_6$ we measured the fastest build-up of light induced waveguide structures ($\tau = 200 \, \mu$s at $I_0 = 0.1 \, \text{W/cm}^2$) reported up to now. This is more than four orders of magnitude faster than for previously studied waveguides induced at visible wavelengths, which were produced by the conventional photorefractive effect [21]. The presented technique allows the generation of different straight and bent dynamic waveguide structures by using external masks or a spatial light modulator. Due to the fast response of Sn$_2$P$_2$S$_6$, these structures can be reconfigured in a sub-millisecond time-scale.
Chapter 6

Main conclusions and outlook

In this thesis we have studied the generation of reconfigurable waveguide structures for dynamic optical switching. The structures have been induced by light in photorefractive materials, namely magnesium doped near-stoichiometric lithium tantalate (Mg:SLT) and tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$, SPS). The first one is a very promising material for photorefraction at ultraviolet wavelengths, which will become more and more interesting as light sources at shorter wavelengths become more common. The latter allows for interband photorefraction in the visible, where many laser sources are presently available, and no special optical elements are needed. The presented technique allows the generation of different straight and bent dynamic waveguide structures by using external masks or a spatial light modulator.

Straight waveguides have been produced at the UV wavelength $\lambda_{UV} = 257$ nm in Mg:SLT with recording times of 10 ms at UV intensities of 0.1 W/cm$^2$. We have found a new phenomenon of fixing of the waveguide structures in absence of homogeneous background illumination. Further research of this fixing effect by investigating holographic gratings in the UV has shown that the fixed structures are due to charges trapped in long-lifetime deep traps. These structures can be read out nondestructively in the visible.

The evolution of light induced one-dimensional waveguides when the sustaining electric field is switched off, leads to a novel kind of dynamic light deflection. We have studied this effect in detail using Sr$_x$Ba$_{1-x}$Nb$_2$O$_6$ (SBN) and Mg:SLT crystals. In the case of SBN the spot separation can exceed the original waveguide width by as much as ten times, as a consequence of light back reflected from the second lateral surface. In Mg:SLT the primary waveguide widening is solely due to stray light and gives smaller deflection distances.

Special attention has been given to the optical and electro-optical properties of LiTaO$_3$ in the UV wavelength range. The dispersion of the unclamped electro-optic coefficients $r_{13}$ and $r_{33}$ in congruent and near-stoichiometric lithium tantalate down to $\lambda = 275$ nm
as well as the refractive indices at UV wavelengths from 275 nm to 400 nm have been
determined. Electro-optic coefficients up to $52 \pm 1$ pm/V and EO figures of merit up to
$n^3r = 915$ pm/V have been measured at $\lambda = 275$ nm. We have extended the polarization
potential model to describe the dispersion of the coefficients near the absorption edge.
The theoretical curve describes very well the experimental data within the error of the
measurements.

Straight waveguides as well as waveguide arrays have been induced in SPS at the
visible wavelength $\lambda_{\text{vis}} = 514$ nm. We have achieved the fastest build-up of light induced
waveguide structures ($\tau = 200 \mu$s at $I = 0.1$ W/cm$^2$) in the visible up to present. This
is more than four orders of magnitude faster than for previously studied waveguides induced at visible wavelengths, which were produced by the conventional photorefractive effect. Due to the fast response of Sn$_2$P$_2$S$_6$, these structures can be reconfigured in a
sub-millisecond time-scale. It has been shown, that an electric field alters the waveguide array in a constructive sense by increasing the refractive index modulation and inducing a spatial shift of the array.

The waveguide structures presented in this thesis are only confined in one direction, representing planar waveguides. However, for the realization of reconfigurable optical interconnects using light induced waveguides it is important to confine the guided light in two dimensions. Possibilities to realize two-dimensional confinement are

- Using thin films of a few $\mu$m thickness grown on a suitable substrate.

- External illumination from two sides of the crystal combined with an externally applied electric field may induce a channel waveguide in the bulk of the crystal. Steering of the wave guided in such a channel is possible if the external illumination is changed dynamically, for instance by using two electrically addressed spatial light modulators. Since these waveguides should be created in the bulk of the crystalline sample, in order to avoid an excessive absorption of the controlling light, the conventional photorefractive effect instead of the interband photorefractive effect has to be chosen. This, however, at the cost of a somewhat slower response.

- Also ion implantation may be used to create a waveguiding layer. The lateral confinement is achieved by irradiation of a crystal with low-dose MeV ions. This leads to the formation of a partially amorphized layer of decreased refractive index buried a few microns below the surface defining a barrier waveguide. By using such a barrier waveguide in combination with the presented light induced waveguides, it should be possible to create a dynamic channel waveguide, i.e., confinement in two dimensions.
Appendix A

Materials

A.1 Lithium Tantalate (LiTaO$_3$)

Lithium tantalate is widely used for acousto-optics, electro-optics, integrated optics, non-linear optics and holographic data storage [90, 91]. Compared to its isomorph lithium niobate, lithium tantalate has a higher melting temperature, a lower Curie temperature and a wider band gap. Due to its wide band-gap (4.6 eV), lithium tantalate (LiTaO$_3$) is a promising material for short-wavelength holographic data storage and dynamic holography. Congruent lithium tantalate crystals (CLT) can be grown by the traditional Czochralski method [48]. Such crystals have a nonstoichiometric nature ([Li$_{1-5x}$Ta$_x$V$_{Li,4z}$]TaO$_3$) where lithium vacancies V$_{Li}$ are compensated by antiside defects Ta$_{Li}$.$^+$. The concentration of this intrinsic defects is in the order of 1%, which can significantly influence the photorefractive properties such as the charge transport parameters or the photoexcitation cross-section.

The renewed interest in this material is driven mostly by the successful growth of near-stoichiometric crystals (SLT), i.e. crystals with a composition very close to stoichiometry [49, 92, 93]. Compared to congruently grown LiTaO$_3$, near-stoichiometric LiTaO$_3$ shows significant reduction of optical damage in the visible and an enhancement of the photosensitivity and of the diffusion-type photorefractive effect in the near ultraviolet [32, 34, 94]. Furthermore, the absorption edge is shifted from about 272 nm for congruent LiTaO$_3$ to about 260 nm, where the edge is defined at the absorption of 100 cm$^{-1}$ [50]. It has been shown that for near stoichiometric LiTaO$_3$, the typical photorefractive build-up and decay time constants are decreased by a factor of five compared to the congruent crystal [32]. Additionally, a distinct reduction of the saturated space-charge field is observed, which is due to a larger increase in the photoconductivity relative to the photogalvanic effect [34, 94].

For lithium niobate and lithium tantalate it was found that doping with magnesium...
(Mg) also reduces the optical damage in the visible [95, 96]. It has been shown, that the concentration of intrinsic (the remaining non-stoichiometric) defects decreases as Mg ions enter the Li sites and compensate the defects. In the UV, doping with magnesium enhances photorefrraction and leads to a higher two-wave mixing gain and a faster response [97]. Furthermore, it has been demonstrated, that the response at the interband wavelength $\lambda_{UV} = 257$ nm is considerably faster as compared to the undoped SLT.

Below the Curie temperature, the point group symmetry of lithium tantalate crystals is trigonal $3m$. The high temperature paraelectric phase is non-polar and has the point symmetry $\bar{3}m$. The phase transition exploits the displacement of Li and Ta ions due to elastic forces, which generates small dipoles and therefore results in a macroscopic spontaneous polarization $P_s$. At room temperature, lithium tantalate is uniaxial with the optical axis and spontaneous polarization along the crystallographic $c$-axis.

![Figure A.1: Absorption spectra of different LiTaO$_3$ crystals investigated in this thesis. $\alpha_a$ and $\alpha_c$ indicate the absorption constant for light polarized parallel to the dielectric $a$ and $c$ axis of the crystal respectively. For the stoichiometric and Mg doped crystals, the absorption constants are approximately equal in the depicted wavelength range.](image)

The Curie temperature $T_C$ strongly depends on the degree of stoichiometry of the grown crystals, which is given by the Li/(Ta+Li) atomic ratio. For congruent crystals the Curie temperature is $T_C = 601 \pm 1°C$, and for sintered powders of exact stoichiometric composition it is $T_C = 692 \pm 1°C$ [49].

Optical absorption spectra have been measured using a Perkin-Elmer spectrophotometer. The effects of multiple Fresnel reflections have been taken into account using the Sellmeier equation from Chapter 2. Figure A.1 depicts the absorption constant of the investigated CLT and SLT crystals as well as the spectrum of the SLT:Mg crystal in the
A.2 Tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$)

Table A.1: Physical properties of congruent and stoichiometric LiTaO$_3$. Optical properties are given for the wavelength $\lambda = 632.8$ nm.

<table>
<thead>
<tr>
<th>Property</th>
<th>CLT</th>
<th>SLT</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>UV absorption edge</td>
<td>$\lambda_{\text{Edge}}$</td>
<td>275 nm</td>
<td>260 nm</td>
</tr>
<tr>
<td>Spontaneous polarization</td>
<td>$P_S$</td>
<td>60 $\mu$C/cm$^2$</td>
<td>55 $\mu$C/cm$^2$</td>
</tr>
<tr>
<td>Coercive field</td>
<td>$E_c$</td>
<td>200 kV/cm</td>
<td>17 kV/cm</td>
</tr>
<tr>
<td>Curie temperature</td>
<td>$T_C$</td>
<td>601$^\circ$C</td>
<td>692$^\circ$C</td>
</tr>
<tr>
<td>Main refractive indices</td>
<td>$n_0$</td>
<td>2.1765</td>
<td>2.1775</td>
</tr>
<tr>
<td></td>
<td>$n_e$</td>
<td>2.1807</td>
<td>2.1742</td>
</tr>
<tr>
<td>Electro-optic coefficient</td>
<td>$r_{113}$</td>
<td>8 pm/V</td>
<td>9 pm/V</td>
</tr>
<tr>
<td></td>
<td>$r_{333}$</td>
<td>31 pm/V</td>
<td>31 pm/V</td>
</tr>
<tr>
<td>Piezoelectric coefficient</td>
<td>$d_{113}$</td>
<td>-3 pC/N</td>
<td></td>
</tr>
<tr>
<td>Dielectric constant</td>
<td>$\varepsilon_{33}$</td>
<td>43</td>
<td></td>
</tr>
<tr>
<td>Heat capacity</td>
<td>$c_p$</td>
<td>424 J/(kg K)</td>
<td></td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>$\lambda$</td>
<td>4.6 W/ (m K)</td>
<td></td>
</tr>
<tr>
<td>Pyroelectric coefficient</td>
<td>$p_3$</td>
<td>$4 \cdot 10^{-5}$ C/(m$^2$ K)</td>
<td></td>
</tr>
</tbody>
</table>

UV wavelength range. The absorption edge of the SLT crystals extends approximately 10 nm further towards the UV as compared with the congruent crystal.

To conclude this section on lithium tantalate Table A.1 lists the physical properties of CLT and SLT relevant for this thesis.

A.2 Tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$)

Tin hypothiodiphosphate is a very interesting material for photorefractive, electro-optical and nonlinear optical applications in the visible and infrared wavelength range. In the photorefractive field Sn$_2$P$_2$S$_6$ is characterized by a large beam coupling gain in the wavelength range 0.5...1.5$\mu$m [3, 6] and a relatively fast response, two orders of magnitude faster than BaTiO$_3$ doped with rhodium, which is the usual photorefractive material in the near infrared [88]. In electro-optics Sn$_2$P$_2$S$_6$ is useful because of its large electro-optical coefficients. For example the diagonal electro-optical coefficient $r_{111} = 166$ pm/V at $\lambda = 1313$ nm [5], which leads to a half-wave voltage 15-20 times smaller than in the standard material LiNbO$_3$. A good introduction to Sn$_2$P$_2$S$_6$ is given in Ref. 4. In this section we will summarize the properties relevant for this thesis.

Sn$_2$P$_2$S$_6$ single crystals can be produced by the conventional vapor transport tech-
nique [99,100] using iodine as a transporter: Stoichiometric amounts of the constituting elements and the transport gas are sealed into quartz ampoules where they react at 650°C - 700°C. The ampoule is placed in the hot spot of a horizontal temperature gradient, while the crystal grows in the cold end by chemical transport.

Sn$_2$P$_2$S$_6$ is a ferroelectric semiconductor with monoclinic point group symmetry $m$ at room temperature. The crystal undergoes a second-order phase transition to the centrosymmetric paraelectric phase (point group 2/$m$) at the Curie temperature $T_C = 338$ K [100].

The coordinate system chosen to describe the physical tensor symmetry is a right handed Cartesian $x$, $y$, $z$ system. The $y$-axis is perpendicular to the symmetry plane ($y \parallel b$), the $z$-axis is chosen parallel to the crystallographic $c$-axis and the $x$-axis perpendicular to $y$ and $z$. Due to the monoclinic symmetry, the optical indicatrix (dielectric coordinate system) is rotated with respect to the Cartesian coordinate system by an angle $\alpha$ as depicted in Fig. A.2. The angle $\alpha$ depends on temperature and on the wavelength. For $\lambda = 632.8$ nm and room temperature on gets an angle of $\alpha = 43.3 \pm 0.4^\circ$ [4].

Ferroelectricity in Sn$_2$P$_2$S$_6$ is given by movement of Sn atoms in a rigid P$_2$S$_6$ framework [101]. In crystals of class $m$ the direction of the spontaneous polarisation $P_S$ is not predictable, it must just lie in the (010) plane. $P_S$ happens to lie between $+x$ and $+z$, 15° from $+x$ [4]. The value for $P_S$ was found to be 15 $\mu$C/cm$^2$ at room temperature and the coercitive field is 750 V/cm [100].

As-grown Sn$_2$P$_2$S$_6$ crystals are usually poly-domain. For electro-optical, photorefractive, nonlinear optical or piezoelectric applications crystals need to be poled. This is
done by heating the sample well over the phase transition temperature $T_C = 66 \pm 2 \degree C$ and slowly cooling it down to room temperature under an applied electric field larger than the coercive field. During heating of a poled sample, $P_S$ decreases up to the critical temperature, where it goes to zero following the square root dependence typical for second order phase transitions [102].

We conclude this section on Sn$_2$P$_2$S$_6$ with an overview of its physical properties in Table A.2 (from Ref. 4).

Table A.2: Basic physical properties of Sn$_2$P$_2$S$_6$ at room temperature. The tensor elements are in the Cartesian coordinate system: (1, 2, 3)≡(x, y, z). For details and other tensor elements please refer to the given references.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spontaneous polarization $P_s$</td>
<td>15 $\mu$C/cm$^2$</td>
<td>[100, 103, 104]</td>
</tr>
<tr>
<td>Transparency range</td>
<td>530 nm – 8000 nm</td>
<td>[105]</td>
</tr>
<tr>
<td>Main refractive indices$^a$</td>
<td>$n_1 = 3.0256$</td>
<td>[106]</td>
</tr>
<tr>
<td></td>
<td>$n_2 = 2.9309$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$n_3 = 3.0982$</td>
<td></td>
</tr>
<tr>
<td>Dielectric constant $\epsilon_{11}$</td>
<td>230 – 300</td>
<td>[107]</td>
</tr>
<tr>
<td>Electro-optic coefficient$^a$</td>
<td>$r^T_{111} = 174$ pm/V</td>
<td>[5]</td>
</tr>
<tr>
<td></td>
<td>$r^T_{221} = 92$ pm/V</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$r^T_{331} = 140$ pm/V</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$r^T_{131} = 25$ pm/V</td>
<td></td>
</tr>
<tr>
<td>Piezoelectric coefficient $d_{111}$</td>
<td>244 pC/N</td>
<td>[108]</td>
</tr>
<tr>
<td>Density $\rho$</td>
<td>3.54·10$^2$ kg/m$^3$</td>
<td>[109]</td>
</tr>
<tr>
<td>Elastic constant $C_{1111}$</td>
<td>4.2 · 10$^{10}$ N/m$^2$</td>
<td>[110]</td>
</tr>
<tr>
<td>Pyroelectric coefficient $p_1$</td>
<td>7 · 10$^{-4}$ C/(m$^2$ K)</td>
<td>[111]</td>
</tr>
<tr>
<td>Coercive field $E_c$</td>
<td>7.5 · 10$^4$ V/m</td>
<td>[100]</td>
</tr>
<tr>
<td>Heat capacity $C_p$</td>
<td>240 J/(mol K)</td>
<td>[112]</td>
</tr>
<tr>
<td>Thermal conductivity $\lambda_1$</td>
<td>0.5 J/(s m K)</td>
<td>[113]</td>
</tr>
<tr>
<td>Nonlinear optical susceptibility$^b$</td>
<td>$\chi^{(2)}_{111} = 24$ pm/V</td>
<td>[114]</td>
</tr>
<tr>
<td></td>
<td>$\chi^{(3)}_{1111} = 17 \cdot 10^{-20}$ m$^2$/V$^2$</td>
<td></td>
</tr>
</tbody>
</table>

$^a$at 632.8 nm
$^b$at 1907 nm fundamental wavelength
Appendix B

Ion implantation in \( \text{Sn}_2\text{P}_2\text{S}_6 \)

The light induced waveguide structures investigated in this thesis were confined in depth only due to the absorption of the writing light. They reached a depth in the order of 100 \( \mu \text{m} \), which is much larger than the light wavelength and were therefore considered as planar optical waveguides with confinement only in transverse direction. In Chapter 6 we mentioned, that ion-implantation offers a possibility to induce an optical barrier just a few micrometers below the surface and to confine the light induced structures in depth. For this purpose ion-implantation was investigated in pure and Te-doped \( \text{Sn}_2\text{P}_2\text{S}_6 \) crystals using different ions.

This chapter first gives a short overview about ion implantation in optoelectronic crystals, explains the barrier coupling method for characterizing the optical barrier in ion implanted waveguides and shows the results obtained with \( \text{H}^+ \), \( \text{He}^+ \) and \( \text{O}^+ \) ions implanted in \( \text{Sn}_2\text{P}_2\text{S}_6 \).

B.1 Ion Implantation

Ion implantation is a universal technique for producing waveguides in a wide range of materials \[115–117\]. High energy ions loose their energy through collisions with electrons and nuclei in the target material. In most inorganic crystals, the penetrating ions loose their energy mainly by interaction with the target electrons, which has little effect on the refractive indices and the crystal structure \[115\]. However, at the end of the ion track, nuclear collisions become the dominating energy loss process, producing a small region of high lattice damage. This leads to a decrease in physical density and hence to a reduced refractive index. In this way a small region of lowered refractive index is created at the end of the ion track, the index barrier, surrounded by regions of higher refractive index.

The ionic energy deposition can be estimated using SRIM simulation (Stopping and Range of Ions in Matter, www.srim.org). The results of such a simulation is shown
in Fig. B.1 for the case of 1 MeV H\(^+\) ions implanted into Sn\(_2\)P\(_2\)S\(_6\). Using the model described in ref. [117] the refractive index profile can be calculated using the electronic \((G_{el}(z))\) and nuclear \((G_{n}(z))\) energy losses.

Pure and Te doped Sn\(_2\)P\(_2\)S\(_6\) crystals have been irradiated in a TANDEM accelerator on the polished z- or y-face by different ions listed in Table B.1. The samples were tilted by 7\(^\circ\) to avoid channeling along the crystallographic axes. After implantation, the edges of the crystals have been polished to enable light coupling and waveguide modes have been measured using the barrier-coupling method [118] as described in the next section. The prism-coupling technique cannot be applied because the refractive indices of Sn\(_2\)P\(_2\)S\(_6\) are too high \((n \sim 3)\) for standard rutile prisms \((n \sim 2.6)\) and the sample surface can be easily damaged by mechanical contact.

Table B.1: Ions used for implantation of Sn\(_2\)P\(_2\)S\(_6\) crystals. The barrier depth \(d\) corresponds to the peak position of the nuclear energy loss \((Gn(x))\) and has been estimated by SRIM simulations.

<table>
<thead>
<tr>
<th>Ion</th>
<th>Energy (MeV)</th>
<th>d (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H(^+)</td>
<td>1</td>
<td>13</td>
</tr>
<tr>
<td>He(^+)</td>
<td>2</td>
<td>6</td>
</tr>
<tr>
<td>O(^+)</td>
<td>10</td>
<td>5.5</td>
</tr>
</tbody>
</table>
B.2 Barrier coupling method

The barrier coupling method allows a contactless determination of the effective indices of the modes of ion implanted waveguides [118]. The principle of this method is illustrated in Fig. B.2(a). A light beam entering the bulk of the crystal from a lateral surface with an external angle $\theta$ will have a propagation constant along the waveguide propagation axis given by Snell’s law:

$$\beta := k_z = k_0 \sqrt{n^2 - \sin^2 \theta} \quad (B.1)$$

where $k_0 = 2\pi/\lambda$ is the vacuum wave vector and $n$ the refractive index of the bulk crystal for the incoming light beam. Because of the finite size of the barrier, light will not be totally reflected and a fraction of it will tunnel inside the waveguide region. However, the in-coupled light will only excite a mode when the propagation constant matches the effective index $N_m$ of the $m^{th}$ waveguide mode ($\beta = k_0 N_m$).

Figure B.2(b) shows the simple setup for the barrier coupling method. The crystal is placed on a rotation stage and the reflected signal is monitored for different incident angles of the beam. Whenever a waveguide mode is excited, the reflectivity drops to a minimum. An example of such a mode measurement is shown in Fig. B.3 for He$^+$ doped Sn$_2$P$_2$S$_6$. Fresnel reflections at the input and output surfaces have been taken into account. The reflectivity clearly shows several dips where the matching condition for exciting a mode are fulfilled. From the estimated positions of these minima ($\theta_m$) the effective indices $N_m$ have been calculated using Eq. (B.1) and are plotted in the inset of Fig. B.3 against the mode number $m$.

We approximated the ion-implanted waveguide with an asymmetric step-index slab waveguide consisting of air, Sn$_2$P$_2$S$_6$ core of thickness $d$ and refractive index $n_{\text{Bulk}}$ and an infinite barrier with refractive index $n_{\text{Bulk}} - \Delta n$. Depending on the configuration
Ion implantation in Sn$_2$P$_2$S$_6$

Figure B.3: Reflectivity as a function of the incident angle for a Te doped Sn$_2$P$_2$S$_6$ crystal implanted with He$^+$-ions measured at $\lambda = 633$ nm. The inset shows the measured effective indices (open circles) and the calculated modes of the best-fit index profile which have been connected for clarity and are shown by the solid line.

$n_{\text{Bulk}} = n_1 = 3.0256$ for $z$-implanted samples and $n_{\text{Bulk}} = n_x = 3.063$ for $y$-implanted samples at $\lambda = 633$ nm respectively. The calculated modes of such a slab waveguide have then been compared with the measured modes. In this way, the two free parameters, $d$ and $\Delta n$, have been estimated from the best-fit step-index profile and are listed in Table B.2. The obtained barrier depths are about 10% smaller compared with the ones from SRIM simulation (Table B.1). This could be on the one hand due to the accuracy of the SRIM routine, on the other hand the step-index approximation gives a reason for the smaller values: The step is not located at the position of the refractive index minimum (values in Table B.1), but in the slope of the index peak nearer to the surface. The implantation parameters like ion energy or fluence have been adjusted quite accurately, except for the crystals SPS44 and SPS45 where the ion energy has been 1.95MeV instead of 2MeV.

Coupling experiments have been performed in two different configurations: For the samples implanted along the $z$ axis, the in-coupling has been in $y$ direction and the polarization of the in-coupling beam was in the $x$-direction. For the samples implanted along the $y$-axis of the crystal, the propagation direction was along $z$ and the beam was polarized along the dielectric $x_1$-axis to excite hybrid $n_1$-modes [117]. In this way, Eigenmodes of the system have been excited in both configurations. The beam has been launched into the waveguide by end-fire coupling using light at $\lambda = 633$ nm. The sample implanted with $O^+$-ions has been damaged so heavily, that polishing the edges was impossible and thus coupling experiments have not been feasible. In the other samples
at least a small portion of the beam (about 1%) has been coupled into the guiding layer.

Photorefractive properties of ion implanted waveguides in Sn$_2$P$_2$S$_6$ have been measured by two-beam coupling experiments. The results are summarized in Ref. [119]. Figure B.4 shows the amplification of the in-coupled signal beam by a stronger pump beam. In the undoped crystal an increase of the effective number of traps $N_{\text{eff}}$ was observed, resulting in a higher two-beam coupling gain $\Gamma$. At $\lambda = 633$ nm $\Gamma$ increased from 3.5 cm$^{-1}$ to 6.8 cm$^{-1}$. In Te-doped Sn$_2$P$_2$S$_6$ no increase of $N_{\text{eff}}$ could be observed in the waveguiding region compared to the bulk crystal, which may be attributed to the high trap concentration in doped samples as compared to the ones created by implantation. At $\lambda = 1.55$ $\mu$m in Te-doped Sn$_2$P$_2$S$_6$ a similar photorefractive response was observed as in the bulk material. This is especially interesting since at that wavelength a high intensity is needed to reach the saturated two-beam coupling region, making waveguides the ideal choice, even though the dark conductivity in the waveguide is about three times higher as in the bulk.

**Table B.2:** Different Sn$_2$P$_2$S$_6$ crystals used for ion-implantation.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Ion</th>
<th>Implantation direction</th>
<th>Fluence ($10^{15}$ Ions/cm$^2$)</th>
<th>d ($\mu$m)</th>
<th>$\Delta n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SPS29d</td>
<td>He$^+$</td>
<td>z</td>
<td>0.25</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>SPS39b:Te</td>
<td>He$^+$</td>
<td>z</td>
<td>0.4</td>
<td>5.65</td>
<td>0.062</td>
</tr>
<tr>
<td>SPS40</td>
<td>H$^+$</td>
<td>y</td>
<td>13.5</td>
<td>9.60</td>
<td>0.055</td>
</tr>
<tr>
<td>SPS41</td>
<td>O$^+$</td>
<td>y</td>
<td>0.04</td>
<td>4.95</td>
<td>0.085</td>
</tr>
<tr>
<td>SPS44:Te</td>
<td>He$^+$</td>
<td>y</td>
<td>0.25</td>
<td>5.52</td>
<td>0.043</td>
</tr>
<tr>
<td>SPS45:Te</td>
<td>He$^+$</td>
<td>y</td>
<td>0.6</td>
<td>5.52</td>
<td>0.090</td>
</tr>
</tbody>
</table>

Figure B.4: Two-wave mixing experiment in an ion-implanted waveguide: CCD camera image of the coupled signal beam at $\lambda = 633$ nm in pure Sn$_2$P$_2$S$_6$. Left: signal beam alone, right: amplification of the signal beam with the pump beam switched on.
ION IMPLANTATION IN Sn$_2$P$_2$S$_6$
Bibliography


List of publications


I would like to thank all those people, without whom the successful completion of this thesis would not have been possible

- *Prof. Dr. Peter Günter*, for giving me the opportunity to collaborate in the Nonlinear Optics Laboratory,

- *Prof. Dr. Germano Montemezzani*, for being the co-examiner of this thesis and for his competent discussions and advisements,

- *Dr. Mojca Jazbinšek*, for her competent and friendly support of my thesis,

- *Roger Mosiman*, for making our office a nice working environment,

- *Jaroslav Jajfler* for expert crystal preparation,

- All other members of the NLO Group, for their kindness and friendship,

- *Dr. Max Döbeli* for sharing his expertise on ion-implantation,

- *Paul Studerus* and *Hans-Peter Hächler* for their help in electronic questions.

- My parents *Peider* and *Margrit* for their enduring love, support and encouragement.

- *Sibylle* for her love and patience.
Curriculum Vitae

Flurin Juvalta, citizen of Zuoz (GR), born September 15, 1979 in Zürich (Switzerland)

2008 PhD thesis on “Light-induced waveguides, waveguide arrays and switches in photorefractive LiTaO$_3$ and Sn$_2$P$_2$S$_6$”

2004–2008 Research and teaching assistant at the Nonlinear Optics Laboratory led by Prof. Dr. P. Günter, ETH Zürich

2004 Graduation with a diploma in experimental physics on “Nonvolatile holographic gratings in the surface region of Mg doped LiTaO$_3$ induced by deep UV light”

1999–2004 Study of physics at the Swiss Federal Institute of Technology (ETH, Zürich) Specialization in electro- and nonlinear optics,

1994–1999 High school specializing in mathematics and scientific studies in Zürich

1986–1994 Primary and secondary schools in Männedorf (ZH, Switzerland)