Optical parallel processing
based on the photorefractive effect

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Abstract

In optical parallel processing data pages are modulated on a laser beam and elaborated in a page by page modus. Data pages can contain several millions of bits, thereby providing very high processing rates which have the potential of surpassing conventional electronics.

This thesis is devoted to the investigation and demonstration of high repetition rate optical parallel processing based on new photorefractive materials operated at visible wavelengths. The generation of high frame rate image sequences and the successive optical correlation of these images were investigated in detail. Optical correlation can be used for pattern recognition and object tracking and is therefore of wide interest for practical applications.

Two different kinds of optical correlators have been considered. The foundation for both these correlators is a holographic storage system which, based on pulsed laser read-out, is capable to generate a fast image sequence. With this system a read-out frame rate of 50’000 images/s has been demonstrated. This is to our knowledge the highest sustained frame rate demonstrated for a holographic storage system. Different storage materials namely KNbO₃, LiNbO₃, and LiTaO₃ were investigated and successfully used to store 100 images.

The first kind of correlator considered in this work is the Vander Lugt type correlator. It is based on multiple holograms acting as matching filters. These filters were implemented using photorefractive LiNbO₃ and LiTaO₃ crystals. In these materials the photorefractive effect provides a reversible storage of the holograms and a long storage time of several years, or even longer using an appropriate fixing method. These properties make the photorefractive effect one of the best methods for building a Vander Lugt type correlator. With our set-up a correlation rate of 10’000 images/s at a repetition rate of 100 comparing images/s was achieved. In our system a very accurate correlation output was demonstrated, enabling the realization of applications like key search in large databases, quality check systems, pattern and position recognition systems.

The second type of correlator considered in this work is the joint Fourier transform type correlator (JFT). In this arrangement the Fourier transformed of the two modulated input beams, write a hologram in real time in a nonlinear optical material. As nonlinear material we choose photorefractive Sn₂P₂S₆ crystals which was for the first time operated in the interband regime at wavelengths of 488, 518, and 532 nm. Under illumination with 40 ns laser pulses from a Q-switched diode pumped solid state laser at 532 nm, a fast writing time of 2 µs was observed already at a moderate fluence of few µJ/cm². With this system a correlation rate of 10’000 images/s has been demonstrated. This is the highest correlation rate reported for
a photorefractive JFT correlator at visible wavelength. Unlike for the Vander Lugt type correlator the JFT correlator based on interband gratings maintains the shift invariance between the input pattern. The system can be used for object tracking and pattern recognition in moving scenes.

LiNbO$_3$ and LiTaO$_3$ showed a strong self-focusing effect which is in principle undesirable for holographic storage, but can be exploited to produce self-trapped light beams (spatial solitons). The effect was investigated in detail in KNbO$_3$ leading to first experimental demonstration of spatial solitons in this material.
Zusammenfassung

In der optischen parallelen Datenverarbeitung wird die Information seitenweise auf einem Laserstrahl moduliert und verarbeitet. Die Datenseiten können mehrere Millionen Bits enthalten und ermöglichen somit sehr hohe Verarbeitungsraten, die herkömmliche elektronische Systeme potenziell übertreffen können.


Die zweite Art optischer Korrelatoren, welche im Rahmen dieser Arbeit untersucht wurde, ist der “joint Fourier transform correlator” (JFT). In dieser Anordnung schreiben die Fouriertransformierten der zwei Eingangsbilder ein Hologramm in Echtzeit in einem optisch nichtlinearen Material. Als nichtlineares Material wur-
Zusammenfassung


LiNbO₃ und LiTaO₃ wiesen einen starken Selbstfokussierungseffekt auf, welcher grundsätzlich für holographische Speicherung nicht von Vorteil ist, aber in der Erzeugung von selbstgeführten Strahlen (räumliche Solitonen) Verwendung findet. Dieser Effekt wurde ausführlich in KNbO₃ untersucht, was zur ersten experimentellen Beobachtung räumlicher Solitonen in diesem Material geführt hat.
Chapter 1

Introduction

The requirement for digital storage of information is growing at an enormous rate (it is expected to exceed $10^{20}$ bits in the year 2000). An important amount of resources will be needed to access, process, and elaborate all this information. Electronic calculators will reach their limits of physically possible operating rates and miniaturization in 10 to 20 years. They suffer from limitation due to their sequential nature and from poor scalability.

Since its proposal by van Heerden [1, 2] in 1963 holographic storage is promising to be the answer to this necessity. This could not be fully realized yet because of the lack of reliable, compact laser sources, high quality holographic materials, and efficient interfacing elements to electronics. The latest development in laser technology, spatial light modulator technology, and charge coupled device technology, allow for the first time an implementation of optical storage systems which can compete with conventional systems[3, 4]. Furthermore optical system can now be efficiently embedded in electronic systems[5, 6], presenting a synergy which will permit to exploit the best of both technologies.

The aim of this thesis is the investigation and demonstration of fast optical parallel processing applications based on the photorefractive effect. Specifically a holographic storage system capable of generating an image sequence of 50'000 images/s which is feeding a fast optical correlator is demonstrated (see Fig. 1.1). Correlation is the basic step in pattern recognition systems and is of fundamental importance for many applications as for example key search operations in very large databases or in object tracking systems.

1.1 Optical computing

The idea of using light to transmit information is one of the most fruitful of the entire mankind. At the beginning very rudimentary methods were used to encode the information on the light signal (for example covering a fire with a towel). The technology developed only slowly till the invention of the laser in 1960 by T. H. Maiman. Whereas nowadays information transmission over optical fiber is a well established standard technique with demonstrated transmission rates as high as 160 Gbit/s at a single wavelength.
Chapter 1. Introduction

Figure 1.1: Fast optical correlation: The image sequence produced by the fast read-out of an holographic memory is correlated with a second input image.

In principle much higher throughput can be obtained if the information is encoded not only temporally but also spatially in a light wave front. This can be achieved using spatial light modulators (SLM). SLMs with more than one million of single channels (pixels) are available. Unfortunately the frame rates of these systems are not yet as high as the 160 GHz demonstrated for a single channel, which would lead to potentially gigantic throughput of 160 Pbit/s. However the most attractive feature of such a massive parallel system is that it is possible to process information in a page-at-once mode. For example the Fourier transform of a data page modulated on a light beam, can be obtained very simply by the use of a lens. Basing on this principle efficient parallel algorithms for many problems like polynomial and matrix computation, sorting, and string matching are possible[7].

1.2 Holographic volume storage

In holographic storage information is stored inside the material volume in form of a hologram. The holograms are generated fixing the interference pattern between the reference and image wave pairs. Illuminating the crystal with one reference beam the corresponding image is reconstructed (see Fig. 1.2). The advantage of this technique compared to conventional storage system like DRAM chip or hard disk, is the possibility to exploit the entire volume and not only the surface of the storage
media. The potential storage density of holographic memories is as high as $10^{12}$ bit/cm$^3$. The demonstrated storage density in photorefractive materials is however lower by 2 orders of magnitude ($\approx 10^{10}$bit/cm$^3$). The main reason for the reduced density is the cross talk noise between the stored images. Cross talk noise arises from light scattered from the multiple stored holograms which is added to the light intensity of the actual reconstructed image. An important effort has been devoted to the investigation and reduction of cross talk noise. The optical scattering properties of the storage material are also important for the storage capacity as they are defining the maximum signal-to-noise ratio between light scattered by the hologram and light scattered by defects. A third source of storage capacity reduction is coming from the self-focusing of the data beam and reference beam, which can in certain unfavorable conditions dramatically reduce the image quality.

The second important advantage of holographic memories over conventional storage system is that the information, organized as data pages, can be read out in parallel. Using an appropriate multiplexing technique very high storage densities, and a fast read-out rate can be achieved. Both properties together make holographic storage an ideal source for parallel optical processing systems. The memory can therefore be considered as a programmable spatial light modulator. The need to integrate the storage element in the SLM is also indicated by the fact that the repetition rate of todays commercially available high resolution SLM is limited by the data transfer rate between memory and SLM device.

![Figure 1.2: Holographic storage: The holograms are written by fixing the interference pattern produced by the object/reference wave pairs. Readout is achieved illuminating with the reference beam corresponding to the image of interest.](image)
1.3 Optical correlation

Optical correlation builds the base element for pattern recognition and object tracking systems. The output of a correlation gives information about the similarity and the relative position of two input images. Mathematically the correlation of two patterns \( s_1(x, y) \) and \( s_2(x, y) \) is defined as

\[
s_1 \otimes s_2(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} s_1(x', y') s_2^*(x' - x, y' - y) \, dx' \, dy'.
\]  

(1.1)

All optical implementations of correlators are based on the correlation theorem

\[
s_1 \otimes s_2 = \mathcal{F}^{-1}[\mathcal{F}(s_1)\mathcal{F}^*(s_2)]
\]  

(1.2)

where \( \mathcal{F}(\cdots) \) denotes the Fourier transform and \( (\mathcal{F}^{-1}(\cdots)) \) the inverse Fourier transform. The correlation theorem reduces the evaluation of the integrations in Eq. (1.1) to a simple multiplication, if the input pattern are first Fourier transformed.

In this work two different types of optical correlators are considered. The first type was proposed by Vander Lugt[17] and is based on a fixed matching filter. The filters are basically pre-recorded holograms. Using a thick holographic medium multiple filters can be recorded in the same material using appropriate reference waves. The correlation time for this kind of device is given by the addition of the setup time of the input SLM, the time needed by the light beam to reach the detector, and the response time of the detector. Whereas the number of correlations is given by the number of matching filters stored in the holographic medium. Providing fast SLM and detection systems very high correlation rates of more than \( 5 \cdot 10^5 \) correlations/s are possible.

The second type of correlator considered is the joint Fourier transform correlator (JFT) proposed by Weaver and Goodman[18]. In this correlator the matching filter is written in real time by the spatial Fourier transformed of the two input signals. The response time of the system is therefore mainly given by the build-up time of the hologram inside the dynamic holographic material. This type of correlator offers the advantage that the images to be compared do not need to be known in advance and the system is fully shift invariant if a thin holographic medium is used.

1.4 The photorefractive effect

Optical computing depends crucially on the availability of suitable nonlinear optical material. Requirements depend strongly on the kind of targeted application. For storage application for example a very long decay time of the written holograms is desired, whereas for a joint Fourier transform correlator a fast build-up and fast decay is required in order to achieve high correlation rates. Additional requirements are a controllable photo sensitivity and light absorption of the material. Due to its unique nature the photorefractive effect[19] can meet most of these requirements, by an appropriate material selection and a controlled doping treatment. The photorefractive effect is based on light induced charge transport in electro-optic materials.
Trapped charge carrier are excited in a mobile state where they can freely move until they are retrapped in an immobile state. In this process an internal space charge field is building up. Through the electro-optic effect this spatially modulated field will translate in a refractive index change, which is in turn detectable by optical diffraction measurements.

In photorefractive materials holographic grating build-up times ranging from few ns to several hours were observed with appropriate light sources. An additional interesting feature is the possibility to observe nonlinear effects already at a moderate light power of few µW.

1.5 Content of this thesis

The aim of this thesis is to demonstrate the potential of the photorefractive effect for optical parallel computing applications. For this purpose a detailed knowledge of the photorefractive effect is required. This is given in Chapter 2 where a general introduction to the photorefractive effect is given. A model which considers a single level of trapped electrons which can be excited in the conduction band is described. As characterization techniques, two wave mixing and the Z-scan technique are presented. The latter is a newly developed method which allows the determination of many photorefractive parameters also in very small samples. In the same chapter photoconductivity and photorefractive effect based on direct band-to-band photoexcitation are explained. Special attention is given to the case of pulsed illumination, for which an analytical solution can be found under the assumption that the diffusion process in the conduction and valence bands is much faster than the recombination of electrons and holes. The general case of interband photorefractive effect combined with a single trap level is finally addressed by numerical simulations of the time and space dependence of the photorefractive space charge field.

Chapter 3 is devoted to photorefractive holographic storage. In the first section the principles of photorefractive holographic storage are presented, including multiplexing techniques and the storage density issue. The influence of photorefractive self-focusing on the image quality is explicitly considered. The theory describing the effect can be found in appendix A, where experimental results for photorefractive self-focusing and spatial soliton formation in KNbO₃ are also reported. Chapter 3 continues with the photorefractive characterization of more than 20 different LiNbO₃, LiTaO₃, and KNbO₄ crystals and a photo-polymer (DuPont HRF 600) to be used as holographic storage material. The chapter terminates with the presentation of a holographic storage system based on a pulsed diode pumped laser. The system is used to store and successively read out 100 image in LiNbO₃, LiTaO₃, and KNbO₃ crystals. The pulsed operation enables the generation of a 50'000 frames/s image sequence, which is the highest reported for pulsed holographic systems to our knowledge.

In Chapter 4 optical correlation based on the Vander Lugt type correlator is considered. The theory for such a correlator is reported and the impact of using thick holographic grating explicitly addressed. In the following an implementation of a Vander Lugt type correlator based on the extension of the holographic storage
system described in Chapter 3 is presented. With this system high quality correlation and high correlation rates (10'000 frames/s) will be shown.

The joint Fourier transform (JFT) correlator is the topic of Chapter 5. The theory for the JFT correlator which considers the influence of thick holographic grating is reported first. An important part of the chapter deals with the first and complete characterization of Sn$_2$P$_2$S$_6$ crystals at a wavelength of 532 nm. The absorption, photoconductivity and photorefractive grating build-up in this material are reported as function of the temperature and under continuous wave and pulsed illumination. The experiment shows that Sn$_2$P$_2$S$_6$ is well suited for the implementation of a JFT correlator. The Chapter concludes with the demonstration of the correlation of a 10'000 frames/s image sequence. This is the highest correlation rate demonstrated for JFT correlator at visible wavelength.
Chapter 2

The photorefractive effect

In photorefractive materials the refractive index can be modified by illumination with light. There are many different effects which can lead to photorefraction as for example photochemical effects, photoinduced reorientation of molecules, Kerr effect at high intensity, or photoinduced thermal effects. In this chapter we will restrict the definition of the photorefractive effect to the light induced refractive index change due to charge transport in electro-optic materials. This definition has become common in literature. The photorefractive effect can show very high light sensitivity in appropriate material thus permitting the observation of such effects at very low light power (< μW). Photorefraction gives rise to many interesting effects as for example light induced wave-guiding, phase conjugation, beam amplification (optical transistor), and four wave mixing. Potentially interesting applications based on these effects are among others holographic storage, laser beam clean up, novelty filters, dynamical optical interconnects, reconfigurable waveguides and switches, and optical correlators.

In the following section the basic theory of photorefraction, starting from the band-transport model will be presented. The special case of direct band-to-band photo-excitation and photoexcitation using pulsed light sources will be considered in the following sections.

2.1 The photorefractive effect

The photorefractive effect was first observed by Askin et. al. in 1966 as beam distortion of a high power laser beam. It took however several more years before the effect was totally understood.

The photorefractive effect can be described considering the following three simple processes:

- Creation through photo-excitation of mobile charge carriers.
- Charge transport of the mobile charge carriers due to diffusion, electric forces, and photogalvanic effect.
- Recombination of the mobile charge carriers (electron or holes) into a trapped state.
Chapter 2. The photorefractive effect

Under inhomogeneous illumination, a space charge field will build up due to the charge redistribution of the second step. The space charge field affects the refractive index through the electro-optic effect, which finally produces a refractive index modulation. In the case of continuous illumination this three steps occur all simultaneously leading to a final steady state charge distribution whereas for short pulse illumination, the charge carriers are excited first, and then redistributed in the dark.

In the following sections the steps introduced above will be discussed in more detail.

2.1.1 Creation of mobile charge carriers

The conventional photorefractive effect is observed in dielectric or semiconductor material. The mobile state to which the charge carriers are excited is the quasi empty conduction band for electrons or the quasi full valence band for holes. Basically two different processes are possible namely the excitation of charge carriers trapped by defects situated at some level in the band gap or the direct excitation from the valence band to conduction band (interband excitation). The photon energy controls which of these processes can take place and play the major role. For photon energy above the band gap all excitation mechanisms are present simultaneously, whereas for a photon energy below the band gap energy, the interband excitation is excluded. Lowering the photon energy even more, it is possible to exclude the excitation of trapped charge carriers depending on their position in the band gap. This property can be used for the so called optical gated holography, where a homogeneous light beam at shorter wavelength is used to produce the population of an empty trap level accessible by a longer wavelength. This mechanism is especially interesting for holographic memory systems to avoid image degradation during read-out. In general many different kinds and levels of traps are present in the band gap. It is possible to introduce additional levels by intentionally doping the crystal during growth. The investigation of the optimum dopant in photorefractive materials like KNbO$_3$, LiNbO$_3$, LiTaO$_3$, Sr$_{0.6}$Ba$_{0.4}$Nb$_2$O$_6$, and BaTiO$_3$ has been a field of large activity in the last three decades. Besides the presence of the trapping levels also their population in thermal equilibrium is important for the photorefractive effect. In oxides this can be controlled changing the amount of oxygen vacancy in the crystal by an oxidation or reduction treatment.

2.1.2 Charge transport

After excitation the charge carriers are in a higher energetic state due to the Franck-Condon effect. They will thermalize within some ps, and eventually form a polaron. The charge transport is governed by electric forces, by diffusion and the photogalvanic effect. Electric forces are given by an external applied field or the internal space charge field produced by charge redistribution. Diffusion is produced

---

*The minimal photon energy needed for exciting a trapped charge carrier cannot be directly related to his position in the energy band scheme due to important contribution from the Franck-Condon shift.
2.1. The photorefractive effect

by the random walk of the charge carriers due to their thermal energy. After excitation the charge carrier can show an initial momentum in a preferential direction. This is called photogalvanic effect[33] and can be used to produce charge separation in an effective way. The effect is important in many materials as for example in LiNbO$_3$ or LiTaO$_3$.

The key properties for describing the charge transport are the mobility and the carrier life time. In dielectric materials which exhibit a very small mobility, the standard method for mobility measurement based on the Hall effect cannot give accurate results. Better results are obtained by holographic time of flight measurements[57]. With this kind of measurements it is possible to determine the mobility and lifetime of the excited charge carriers in several decades of time, permitting to obtain the pure band mobility some ns after excitation and also the lowering of the observed mobility due to the presence of traps near the conduction band (shallow traps) observed in photorefractive experiments under continuous illumination[58]. The influence of shallow traps on the mobility and charge recombination can then be considered by introducing an effective mobility and an effective recombination rate for the time window of interest[59].

2.1.3 Retrapping of charge carriers

The final step in the photorefractive effect consist in the retrapping of the previously excited charge carriers. The retrapping can be radiative or non radiative for both recombinations from band to band and recombine into deep traps. In all these cases the recombination rate is proportional to the local number density of free charge carriers and the number density of empty traps.

2.1.4 Photorefractive index gratings

Irradiating a photorefractive material with an homogeneous light illumination the only effect observed is a change in the conductivity. 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 plane waves. Considering two plane waves with wavevectors $k_1$ and $k_2$ and amplitudes $A_1$ and $A_2$ respectively we obtain a sinusoidal intensity pattern of the form

$$I(x) = I_0 (1 + m \cos (K_g \cdot x))$$

where $K_g = k_1 - k_2$ is the grating vector, $x$ is the position vector, $m = 2 \frac{|A_1||A_2|}{|A_1|^2 + |A_2|^2}$ is the modulation depth, and $I_0 = \frac{1}{2} \sqrt{\varepsilon_0 / \mu_0 (|A_1|^2 + |A_2|^2)}$ the mean intensity. For such an intensity distribution the photorefractive effect will build-up a spatial periodic space charge field $E_{sc}$ with the same period. The direction of $E_{sc}$ is oriented parallel to the grating vector $K_g$ and we will denote in the following the amplitude of the first spatial Fourier component of the space charge field with $E_{sc}$. The exact amplitude of $E_{sc}$ and the dephasing between the intensity pattern and $E_{sc}$, which depends on the material properties, will be treated in detail in sections 2.2 and 2.3.
The presence of a periodic space charge field $E_{sc}$ in an electro-optic material generates a refractive index grating which can be investigated by diffraction experiments of a third plane wave, eventually at a different wavelength. For non centrosymmetric materials which exhibits the linear electro-optic effect (Pockels effect) the change of the dielectric permittivity tensor is defined as

$$\Delta \epsilon^{-1} = r^{eff}(K_g)E_{sc}. \quad (2.2)$$

where $\epsilon$ is the dielectric permittivity tensor in the material at light wavelength and $r^{eff}(K_g)$ is a matrix describing the electro-optic effect which considers the unclamped electro-optic tensor and the additional contributions from the piezo-electric effect.[60].

For the special case that the grating spacing vector $K_g$ is along the polar axis of the material, the amplitude of the spatially periodic refractive index change $\Delta n$ can be expressed as

$$\Delta n \approx n^3 r^{eff} E_{sc} \quad (2.3)$$

where $n$ is the refractive index seen by the beam, and $r^{eff}$ is the effective electro optic coefficient relevant for this geometry. In the following section the diffraction at index gratings is considered in detail.

**Beam diffraction at an holographic grating**

Diffraction gratings are one of the most powerful elements in modern optics. For a diffraction grating two regimes can be distinguished. In the so called Raman-Nath regime multiple diffraction orders appear. This is the case of thin gratings in which the two quantities

$$Q = \frac{2\pi \lambda d}{\Lambda_g^2} \quad (2.4)$$
$$\rho = \frac{\lambda^2}{\Lambda_g^2 n \Delta n} \quad (2.5)$$

are $Q < 1$ and $\rho < 10$. In Eq. (2.4) $\lambda$ is the wavelength of the light, $d$ is the interaction length between light and grating, and $\Lambda_g$ is the grating spacing which is related to the grating vector by $|K_g| = 2\pi / \Lambda_g$. The second regime is the so called Bragg diffraction where only one diffraction order appears. Bragg diffraction is observed for thick gratings ($Q \gg 1$ and $\rho \geq 10$). In this regime a full diffraction of the input beam is possible. We will focus on Bragg diffraction only because it is of major interest for photorefractive experiments in thick crystals.

The diffraction efficiency $\eta = I_{dif}/I_{in}$, where $I_{dif}$ and $I_{in}$ are the intensities of the diffracted beam and input beam respectively, was first calculated by Kogelnik[61] using the theory of coupled waves and recently expanded to the case of anisotropic materials by Montemezzani and Zgonik[62]. For an unslanted transmission grating (see Fig. 2.1) one obtains the following equation:
2.1. The photorefractive effect

Figure 2.1: Schematic representation of light diffraction at an unslanted holographic grating with grating period $\Lambda_g$. On the right-hand side the wave-vector momentum conservation for the incident and diffracted beam is depicted. The difference between the wave-vectors must correspond to the grating vector $K_g = 2\pi / \Lambda_g$.

\[
\eta = \frac{\sin^2 \nu^2 + \xi^2}{1 + \xi^2 / \nu^2} e^{-\alpha d} \quad \text{(2.6)}
\]

\[
\xi^2 = \frac{\Delta k^2}{4} \quad \text{(2.7)}
\]

\[
\nu^2 = \frac{k_0^2 A_r^2}{16 n_i n_{dif} g_{in} g_{dif} \cos \vartheta_{in} \cos \vartheta_{dif} d^2} \quad \text{(2.8)}
\]

\[
g_{in,dif} = \hat{e}_{in,dif} \hat{d}_{in,dif} \quad \text{(2.9)}
\]

where $\alpha$ is the linear intensity absorption of the material, $\Delta k$ represents the wave-vector mismatch from the Bragg condition, $g_{in,dif}$ are the projection cosines between electric ($\hat{e}_{in,dif}$) and dielectric displacement ($\hat{d}_{in,dif}$) unit beam of the incident and diffracted beam respectively, and $\vartheta_{in,dif}$ are the angles inside the material between the Poynting vectors of the incident and the diffracted wave and the normal of the entrance surface (see Fig. 2.1). The dependence of the diffraction efficiency on the beam incidence angle in the air $\vartheta'_\text{in}$ is shown for a 1 cm thick LiNbO$_3$ crystal in Fig. 2.1. The diffraction efficiency presents a strong central peak. For small values of $\nu^2 \ll 1$ the curve of Fig. 2.2 is proportional to $\nu^2 (\sin (\xi) / \xi)^2$. In this case $\sqrt{\eta}$ results simply proportional to $\nu$ and therefore proportional to the spatial field modulation $E_{sc}$. The position of the side minima and the zero crossing is determined by $\xi$. As can by noted from Fig. 2.2 the grating presents a very high angular selectivity. The width of the central peak is only about 0.02°, this is the fundamental property which holographic storage is based on. Another important property of Eq. (2.6) is that for large value of $\nu \approx 1$ diffraction efficiency as high as 100% can be expected if the

\[1\]The relation between the angle inside the material and in the air is given by Snell’s law

\[n_{in} \sin (\vartheta_{in}) = \sin (\vartheta'_{in})\]
absorption is negligible. This allows a very effective way to deviate light beams.

The diffraction properties of a given material can be described by the refractive index modulation $\Delta n$. In order to describe the time dependence of $\Delta n$ we can introduce the sensitivities $S_1$ and $S_2$ as figure of merit. Where $S_1$ is defined as

$$S_1 = \frac{1}{\alpha} \frac{\partial \Delta n}{\partial (I_0t)} \bigg|_{I_0t=0}$$

and is directly related to the light energy $I_0t$ needed to produce a certain $\sqrt{\eta}$ in thick sample ($d \gg 1/\alpha$). The second sensitivity

$$S_2 = \frac{\partial \Delta n}{\partial (I_0t)} \bigg|_{I_0t=0}$$

is related to the light energy needed to produce a certain $\Delta n$. The sensitivities $S_1$ and $S_2$ are very useful to compare the holographic properties among different material and effects leading to photorefraction.

In the next sections we will describe the charge transport models which drive the build-up of such holographic gratings.

### 2.2 The single level model

In this section we consider the simplest possible model for the photorefractive effect. In this model a single level of trapping sites is introduced between the valence and
2.2. The single level model

The conduction band (see Fig. 2.3). The traps are acting as electron donors and the bound electron can be excited in the conduction band by a photon with an appropriate energy. In a real crystals it is a common case that more complicated level structures are present. The model described here gives, however, a good picture of the kind of properties which can be expected from the photorefractive effect. This model was presented by Kuhktarev, Vinetskii, et. al.\[63\] and will be referred in the following as the Kuhktarev-Vinetskii model. The model can be described by the following set of equations:

\[
\frac{\partial n}{\partial t} = (s_e I + \beta_e)(N_D - N_D^+) - \gamma_e n N_D^+ + \frac{1}{e} \nabla J_e, \quad (2.13)
\]

\[
\frac{\partial N_D^+}{\partial t} = (s_e I + \beta_e)(N_D - N_D^+) - \gamma_e n N_D^+, \quad (2.14)
\]

\[
J_e = e n \mu_\epsilon E + k_B T \mu_\epsilon \nabla n + e s_e I (N_D - N_D^+) L_{ph}, \quad (2.15)
\]

\[
\nabla (c_0 \epsilon_{eff} E_{sc}) = e (N_D^+ - n - N_A), \quad (2.16)
\]

where \(n\) is the free electron concentration in the conduction band, \(N_D\) is the donor concentration, \(N_D^+\) is the concentration of ionized donors, \(N_A\) is the concentration of ionized donor in the dark for \(\beta_e = 0\), \(J_e\) is the electron current density vector, \(E_{sc}\) is the space charge field vector, \(I\) is the space dependent light intensity, \(s_e\) is the electronic photoionization constant, \(\beta_e\) the electronic dark generation rate, \(\gamma_e\) is the electron recombination constant, \(\mu_\epsilon\) is the electron mobility tensor, \(L_{ph}\) is the photogalvanic transport length vector, \(\epsilon_{eff}\) is the vacuum dielectric permittivity.
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is the effective dielectric permittivity tensor, \(\varepsilon\) is the unit charge, \(k_B\) the Boltzmann constant, and \(T\) the absolute temperature.

The first two equations are the rate equations for the free electron concentration and the ionized donors concentration. The third equation is the definition of the electric current in the conduction band, and has the form of the continuity equation when introduced in the first equation. The last equation is the Gauss equation which relates the charge density with the electric field \(E_{sc}\).

An algebraic solution of the Kuhktarev-Vinetskii equation exists only for some special cases. In the next section, the solution for homogenous illumination is presented, whereas in the following section 2.2.2 the solution for sinusoidal illumination, which is important for most applications, is considered.

2.2.1 Solution for homogenous illumination

Under illumination with homogeneous light intensity \(I_0\), all spatial derivatives in Eqs. (2.13)-(2.16) can be neglected, and the main effect of the illumination is a change of free electron concentration \(n_0\) and of the ionized donors \(N^+\). Solving the equations for the case of moderate light intensity \(n \ll N_D - N_A\) one obtains the following exponential form

\[
n_0(t) = \frac{N_D - N_A}{\gamma_e N_A} \left( \beta_e + s_e I_0 \left[ 1 - e^{-\gamma_e N_A t} \right] \right).
\]

(2.17)

Also the decay in the dark can be described with an exponential form with the same time constant \(1/(\gamma_e N_A)\). The change of free carrier concentration can be tested by measuring the conductivity of the crystal which is related to \(n_0\) by the relation

\[
\sigma_{tot} = e\mu_e n_0.
\]

(2.18)

Photoconductivity combined with optical absorption and beam coupling experiments (see section 2.2.3) give a powerful method for the characterization of photorefractive materials.

2.2.2 Solution for sinusoidal illumination

A solution of Eqs. (2.13)-(2.16) can be found in the case where the material is illuminated with an intensity distribution as described by Eq. (2.1).

The stationary amplitude of the first Fourier-component of the resulting space charge field \(E_{sc}\) is then given by

\[
E_{sc} = -im \frac{E_q^* (E_D - iE_0)}{E_q^* + E_D - iE_0}.
\]

(2.19)

For a better physical interpretation of \(E_{sc}\) we have introduced the trap limited field \(E_q^*\) and the diffusion field \(E_D\). The first is defined as

\[
E_q^* = \frac{e}{\varepsilon_{eff} K_g N^*_{eff}}
\]

(2.20)
2.2. The single level model

and gives a limit for the maximum $E_{sc}$ that can be generated with the available traps. In Eq. (2.20) $\epsilon_{\text{eff}}$ is the effective dielectric constant for the chosen configuration,[64], and $N_{\text{eff}}^*$ is the effective trap density defined as

$$N_{\text{eff}}^* = \frac{N_D^+ (N_D - N_D^0)}{N_D} \tag{2.21}$$

where $N_D^0$ is the spatial average of ionized donors with illumination switched on. The diffusion limited field is given as

$$E_D = \frac{k_B T}{e} K_g \tag{2.22}$$

It corresponds to the field amplitude of a sinusoidal electric field that exactly counteracts the effect of the charge diffusion process.

In the case no electric field is applied Eq. (2.19) can be simplified to

$$E_{sc} = \frac{-\text{i} m}{E_q^* + E_D^*} \tag{2.23}$$

An important conclusion can be deduced from Eq. (2.23) 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. Based on this property a method for the experimental determination of $N_{\text{eff}}^*$ will be presented in section 2.2.3.

The build-up time of the space charge field in the case no electric field is applied can be described by the single exponential

$$E_{sc}(t) = E_{sc}(t \to \infty) \left(1 - e^{-t/\tau_{pr}}\right). \tag{2.24}$$

The time constant $\tau_{pr}$ of the photorefractive build-up is given by

$$\tau_{pr} = \frac{\tau_{Di}}{1 + 2\pi L_D / \Lambda_g} \tag{2.25}$$

The first constant $\tau_{Di}$ in this equation is the Maxwell dielectric relaxation time constant, and represents the time which is needed for the space charge field to relax by a given conductivity $\sigma_{\text{tot}}$. This is described by

$$\tau_{Di} = \frac{\epsilon_0 \epsilon_{\text{eff}}}{\sigma_{\text{tot}}} = \frac{\epsilon_0 \epsilon_{\text{eff}}}{\epsilon k e n_0}. \tag{2.26}$$

The other entities in Eq. (2.25) are the grating spacing $\Lambda_g$ and the diffusion length $L_D$ which gives the average length that an excited electron will diffuse before it is retrapped and the Debye screening length $\Lambda_{Db}$. The diffusion length is given by

$$L_D = \sqrt{\frac{k_B T \mu_e}{e \gamma_e N_A}} \tag{2.27}$$

and the Debye length is defined as

$$\Lambda_{Db} = 2\pi \sqrt{\frac{\epsilon_0 \epsilon_{\text{eff}} k_B T}{e^2 N_{\text{eff}}}} \tag{2.28}$$
and describes the penetration depth of the electric screening field around a defect in a conducting material. As can be seen from Eq. (2.25) the build-up time in photorefractive materials depend essentially on the dielectric relaxation time. In the single level model the relaxation time is inversely proportional to the mean intensity $I_0$. This allows to reach short response times by a simple increase of the light intensity. It is noteworthy that in contrast to the build-up time the stationary amplitude of the space charge field is independent on the light intensity. This is true as long as the light intensity $I_0$ is larger than the dark intensity $I_D$. The dark intensity is the equivalent light intensity which would produce a conductivity change equal to the conductivity in the dark.

In the following sections two simple experiments useful for the characterization of photorefractive material are presented. The first one is based on photorefractive coupling of two laser beam and allows the determination of the effective trap density $N_{\text{eff}}^*$. The second experiment (Z-scan) is a single beam technique in which the photorefractive lens produced by the beam itself is characterized. With this method the dark intensity $I_D$, the refractive index change $\Delta n$, and the photogalvanic field $E_{\text{ph}}$ can be determined.

### 2.2.3 Two beam coupling

Two beam coupling is a simple experimental technique, to evaluate the photorefractive properties of a material. The experiment allows to probe the magnitude and the build-up time of the space charge field $E_{\text{sc}}$. From the beam coupling measurement, the effective trap density $N_{\text{eff}}^*$ (see Eq. (2.21)), which is mainly responsible for grating spacing dependence of the space charge field, can be determined.

In two beam coupling experiments, two coherent light beams, the signal beam with the intensity $I_{\text{sig}}$ and the pump beam with intensity $I_{\text{pmp}}$, interact in the photorefractive media. The nonlinear interaction between the two waves may result in energy or phase transfer between the beams.

The interference between the signal and pump beams produces an intensity pattern in the crystal, and a space charge field described by Eq. (2.23) builds up. If diffusion is the dominant charge transport mechanism for the photoexcited charge carriers, a refractive index grating is built up, which is exactly $\pi/2$ out of phase with respect to the intensity grating. The pump beam is diffracted off the grating and the diffracted wave is either exactly in phase with the signal or out of phase. Therefore, the signal is either amplified or depleted. For $I_{\text{pmp}} \gg I_{\text{sig}}$ (undepleted pump approximation) the measured energy transfer can be characterized by the exponential gain $\Gamma$ which is defined as

$$\Gamma = \frac{1}{d} \ln \left( \frac{I_{\text{sig}}^{\text{on}}}{I_{\text{sig}}^{\text{off}}} \right)$$

(2.29)

where $I_{\text{sig}}^{\text{off}}$ and $I_{\text{sig}}^{\text{on}}$ are the intensities of the transmitted signal beam before and after turning on the pump beam and $d$ is the length of the path of the signal beam in the crystal along its energy propagation direction [62].

The exponential gain $\Gamma$ can be calculated starting from the wave equation and the undepleted pump approximation. The general result for anisotropic crystals (see
Ref. [62]) is given by

\[ \Gamma = \frac{2\pi}{\lambda} n_{\text{pmp}}^2 n_{\text{sig}} g_{\text{pmp}} a_r r^{\text{eff}} \text{Im} (E_{sc}) \]  

(2.30)

where \( \lambda \) is the vacuum wavelength, \( n_{\text{pmp}} \) and \( n_{\text{sig}} \) are the refractive index seen by the pump and signal beam respectively, \( a_r \) is the polarization projection factor for anisotropic absorption (see Ref.[62]) and is given by \( a_r = e_{\text{sig}} \cdot e_{\text{pmp}} \) only in the case of isotropic absorption. The factor \( g_{\text{pmp}} \) gives the correction for the anisotropic refractive index and is defined by \( g_{\text{pmp}} = d_{\text{pmp}} \cdot e_{\text{pmp}} \) and \( r^{\text{eff}} \) is the scalar electro-optic (EO) coefficient valid in the given photorefractive configuration and can be calculated in the following way[65]

\[ r^{\text{eff}} = d_{\text{pmp}} r^{\text{eff}} d_{\text{sig}} \]  

(2.31)

where \( d_{\text{sig}} \) and \( d_{\text{pmp}} \) are the directions of the dielectric displacement, \( e_{\text{sig}} \) and \( e_{\text{pmp}} \) are the directions of the electric field, and \( r^{\text{eff}} \) is an effective electro-optic matrix which takes into account all elastic deformations induced by the space-charge field and depends on the grating vector direction[65].

**Transmission geometry**

In the co-propagating or transmission geometry, as presented in Fig. 2.1 a), the signal and the pump beam enter the crystal from the same face. The crystal is normally cut along the crystallographic axes so that for symmetric incidence the refractive indices \( n_{\text{sig}} = n_{\text{pmp}} \) are the same, no matter how the beams are polarized. The gain \( \Gamma \) is usually measured for different grating spacing \( \Lambda_g \) varying the angle \( 2\theta \) between the signal and pump beams. The experimental values may then be described with the theoretical expression

\[ \Gamma = \frac{2\pi}{\lambda} n_{\text{pmp}}^3 r^{\text{eff}} \frac{E_s^* E_D}{E_q^* + E_D} e_{\text{sig}} \cdot e_{\text{pmp}}. \]  

(2.32)

In the case when the two beams are polarized perpendicular to the incidence plane (s-polarization) the only dependence on \( \Lambda_g \) is given by the term

\[ \Gamma(\Lambda_g) \propto \frac{E_q^* E_D}{E_q^* + E_D}, \]  

(2.33)

which presents a maximum for a certain grating spacing \( \Lambda_g' \), where \( E_D \) is equal to \( E_q^* \) (see Fig. 2.5). In the case that the experimentally accessible range of \( \Lambda_g \) covers \( \Lambda_g' \), \( N_{\text{eff}}^* \) can be determined even without a complicated fit. For an usual sample with two parallel polished surfaces, the minimum reachable value of \( \Lambda_g \) is \( \Lambda_g^{\text{min}} = \lambda/2 \). Especially with highly doped crystals, when \( N_{\text{eff}}^* \) is expected to become very large, the maximum of \( \Gamma \) moves towards smaller \( \Lambda_g \). If the position of the maximum \( \Lambda_g' \) is below \( \Lambda_g^{\text{min}} \), the fitting function is only weakly dependent on \( N_{\text{eff}}^* \) in the measured region. This can be seen by considering that for \( \Lambda_g > \Lambda_g' \) the diffusion field \( E_D \) is the limiting field in Eqs. (2.32) and (2.33), and \( E_D \) is independent on \( N_{\text{eff}}^* \).
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Figure 2.4: Two beam coupling geometry in a) co-propagating geometry and b) counter-propagating geometry. The signal beam $I_{\text{sig}}$ and the pump beam $I_{\text{pmp}}$ write a grating denoted by the grating vector $K_g$ and $\theta$ is the half angle between pump and signal beam in a) and the incidence angle of pump and signal beam in b).

Figure 2.5: Grating spacing dependence of the space charge field $E_{sc}$. For small grating spacing $E_{sc}$ is proportional to the charge limited field $E^*_q$ whereas for large grating spacing $E_{sc}$ is proportional to the diffusion field $E_D$. 
2.2. The single level model

In the case when the polarizations of the two beams are in the plane of incidence, the determination of large \( N_{\text{eff}}^* \) is even more problematic. At smaller \( \Lambda_g \) that is increasing the angle \( \theta \), the scalar product between the polarizations of the two beams \( \mathbf{e}_{\text{sig}} \cdot \mathbf{e}_{\text{prop}} \) decreases, additionally reducing the sensitivity in the critical region close to \( \Lambda_g' \).

It is noteworthy that \( \Lambda_g' \) can also be increased by choosing a geometry with a high effective dielectric constant \( e_{\text{eff}} \). In a polar material this is normally the case, when the grating vector is perpendicular to the polar axis. The effective trap density can then be determined measuring two beam coupling through absorption grating. The method is described in ref. [66]. However there are crystals in which even with this method a precise determination of the trap density is not possible.

Reflection geometry

An additional way to overcome these problems is to increase the accessible grating spacings range by the use of reflection geometry, that is with the signal beam and the pump beam aligned anti parallel as seen in Fig. 2.1 b). The minimum \( \Lambda_g \) is given in this case by \( \Lambda_g^{\min} = \frac{\lambda}{2n} \) which is the smallest possible grating spacing (approx. 0.11 \( \mu \)m for KNbO\(_3\) at \( \lambda = 488 \) nm). At this grating spacing \( E_D \) is in the order of 15 kV/cm\(^{-1}\) at room temperature, and generally larger than \( E_q^* \). This means that \( E_q^* \) is the limiting field in Eqs. (2.32) and (2.33) and a measurement of the gain \( \Gamma \) is more directly related to \( E_q^* \) and therefore to \( N_{\text{eff}}^* \).

The equation for two beam coupling in this configuration can be rewritten to give the effective trap density as

\[
N_{\text{eff}}^* = \frac{4\pi e_0 e_{\text{eff}} n_{\text{sig}}}{e} \frac{E_D \Gamma}{2\pi n_{\text{sig}}^3 v_{\text{eff}} E_D - \Gamma \lambda} \tag{2.34}
\]

and by making the approximation for the case of \( E_q^* \ll E_D \) one obtains

\[
N_{\text{eff}}^* \approx \frac{2e_0}{e} \frac{1}{n_{\text{sig}}^2 v_{\text{eff}} \Gamma} \tag{2.35}
\]

The counter-propagating geometry has the additional advantage that the diameter of the signal and pump beam can be kept small permitting the measurement of two beam coupling at higher intensities, e.g. few W/cm\(^2\), already with mW power laser. This is not the case in the co-propagating geometry, where the spot width must be increased to achieve good overlap of the two beams.

Influence of electron-hole competition

When only one type of charge carriers and photosensitive centers in the crystal are dominant, the theory of two beam coupling presented so far is exactly valid. If more charge carrier types and centers are involved, Eq. (2.23) has to be modified. The case in which two charge carriers of opposite charge are participating in the build-up of the photorefractive space charge field is called electron-hole competition and was theoretically treated in Refs.[67, 68]. Two different simplified models were proposed in Ref. [68]: Model I in which the simultaneous electron and hole transport is treated...
and both charge carriers originate from a single common set of recombination centers and Model 2 in which the electrons and holes have their own recombination centers set. Implication of both models on two beam coupling are now discussed.

When electrons and holes have a single common set of recombination centers the resultant space charge field \( E_{dc} \) is modified by introducing a correction \( \zeta_1(K_g) \) as follows

\[
\text{Im}(E_{dc}) = m \frac{E_D E_q^*}{E_D + E_q^*} \zeta_1(K_g)
\]

where

\[
\zeta_1(K_g) = \frac{1 - \frac{\sigma_h}{\sigma_e} + \frac{E_D}{E_q} \left( \frac{\Gamma_{eh}}{\Gamma_{ie}} - \frac{\Gamma_{eh}}{\Gamma_{ie}} \right)}{1 + \frac{\sigma_h}{\sigma_e} + \frac{E_D}{E_q} \left( \frac{\Gamma_{eh}}{\Gamma_{ie}} + \frac{\Gamma_{eh}}{\Gamma_{ie}} \right)}.
\]

here \( \sigma_e \) and \( \sigma_h \) are the electron and hole conductivities, \( \Gamma_{dih} \) is the dielectric relaxation rate for holes and \( \Gamma_{ih} \) and \( \Gamma_{ie} \) are the sum of the photoproduction and ion recombination rates appropriate for holes and electrons, respectively[68].

All entities on the right-hand side of Eq. (2.37) are defined as positive, it is then easily seen that the limit \(-1 \leq \zeta_1(K_g) \leq 1\) must hold. The correction factor \( \zeta_1(K_g) = 1 \) in the case of pure electrons conduction and \( \zeta_1(K_g) = -1 \) in the case of pure holes conduction. The correction \( \zeta_1(K_g) \) can vary strongly as a function of the grating vector and even a change of sign can be predicted. In the case of large grating spacing, that means \( E_q^* \gg E_D \Gamma_{dih}(1/\Gamma_{ih} + 1/\Gamma_{ie}) \), the correction \( \zeta_1(K_g) \) can be approximated with a constant factor

\[
\zeta_1(K_g) = \hat{\sigma} = \frac{\sigma_e - \sigma_h}{\sigma_e + \sigma_h}.
\]

A verification of this behavior can be found for instance in Ref. [69].

In the second model electrons and holes have their own independent sets of recombination centers. Also in this case \( E_{dc} \) can be modified introducing a correction \( \zeta_2(K_g) \) which is of the form

\[
\zeta_2 = \frac{N_{eh}^*}{N_{eh} + N_{ec}^*},
\]

where \( N_{eh}^* \) and \( N_{ec}^* \) are the effective trap densities for holes and electrons respectively. The correction term still lies within the interval \(-1 \leq \zeta_2 \leq 1\) and \( \zeta_2 = 1 \) in the case of pure hole conduction and \( \zeta_2 = -1 \) in the case of pure electron conduction. Similarly as in the first model at large grating spacing, in this model the influence of the electron-hole competition consists of a constant multiplicative term.

Which model is adequate for a given crystal, can be only verified by measurements. Model 1 has more free parameters and is used when it is not possible to fit the results with model 2. The measurements at short grating spacings are then determined by the dielectric relaxation rate for holes and the photoproduction and ion recombination rates. In the case of model 2 the trap density can be determined by two measurements. At large grating spacing \( (E_D \ll E_q^*) \) the space charge field is given by \( E_{dc} = iE_D \zeta_2 \) and \( \zeta_2 \) can be directly measured, and at small grating spacing the effective trap density is determined through the already known \( \zeta_2 \).
Experimental methods

It is a common situation encountered in photorefractive crystals that several active centers are present. Sometimes, also using model 1, it is not possible to obtain a good agreement between two beam coupling theory and experiment. A theoretical model which considers more then two recombination centers is presented in Ref. [70]. However, in this case the number of recombination centers should also be investigated by additional experimental techniques such as EPR, induced absorption and transparency (see Ref. [71]) if meaningful results are to be obtained.

The determination of the trap density and the correction factor for electron-hole competition in highly doped crystals is only possible considering experiments in both geometries, counter- and co-propagating. Measurements in co-propagating only geometry leads to a set of redundant fit parameters for model 1 (2.37) as reported in Ref. [67].

The obvious consequence of electron-hole competition for the results of the two beam coupling experiment in counter-propagating geometry is that the measured effective trap density has to be considered as a lower bound for the real trap density. If we measure a small \( N_{\text{eff}}^* \), the true trap density or densities might be anyway bigger, however the strong electron-hole competition makes the crystal less attractive for practical applications.

Apart from electron-hole competition there are also other non-ideal experimental conditions which can diminish the measured gain \( \Gamma \). Partial poling of the crystal, badly polished surfaces of the crystal, defects in the crystal, photorefractive fanning and reflection of pump and signal beam on the crystal surfaces, all these problems should be avoided.

Partial poling of the crystal, bad polished surface of the crystal, defects in the crystal can be avoided by appropriate growth and preparation techniques, and are usually not a problem in our KNbO₃ crystals. Fanning can be reduced by choosing the polarization and the propagation direction of the beams. In order to avoid reflections it can be useful to put the sample in an index matching liquid or use Brewster angle incidence of the beams.

Note that neglecting these effects would not cause any error in the determination of \( N_{\text{eff}}^* \) when the experimental results cover the range of maximum \( \Gamma \), but would lead to an incorrect interpretation as the electron-hole competition. These effects should be minimized especially when the effective trap density is determined solely by the counter-propagating method.

Example: Mn doped KNbO₃

As a demonstration for the advantages of the counter-propagating two beam coupling method, we characterized a KNbO₃ crystal doped with Mn by measuring the trap density \( N_{\text{eff}}^* \) using both co- and counter-propagating geometries.

We first performed experiments in the co-propagating geometry (see Fig. 2.6). The grating spacing vector was chosen parallel to the polar axis, and the pump and signal beam were generated by the 488 nm line of an Ar⁺ ion laser. The polarizations were taken perpendicular to the plane of incidence. The pump beam was expanded and produced a homogeneous illumination of 1 W/cm². The signal
Figure 2.6: Two beam coupling measurements in a KNbO3 crystal doped with Mn. The left-hand side of the graph shows the gain $\Gamma$ in the counter-propagating geometry for grating spacings $\Lambda_g$ from 0.1 to 0.2 $\mu$m and the right-hand side in the co-propagating geometry for grating spacing from 0.4 to 1.5 $\mu$m. The theoretical curve are plotted for following different effective trap densities: dash-dotted line $N_{\text{eff}}^* = 3.6 \cdot 10^{22}$ m$^{-3}$, solid line $N_{\text{eff}}^* = 6.5 \cdot 10^{22}$ m$^{-3}$, and dashed line $N_{\text{eff}}^* = 12 \cdot 10^{22}$ m$^{-3}$. The gains measured in the co-propagating geometry are well fitted by all of the three theoretical curves. It is the measurement in the counter-propagating geometry which permit a precise determination of the trap density.

beam had a 400 times smaller intensity in order to comply with the undepleted pump approximation. It had a smaller diameter as the pump to guarantee a total overlap of the two beams in the crystal. The intensity of the signal beam was then measured with pump beam turned on and off for different angles $2\theta$. From this data and Eq. (2.29) the gain was determined. The photorefractive space charge field $E_{sc}$ was calculated using the effective EO coefficient and the effective dielectric coefficient from Ref. [65]. The field $E_{sc}$ was fitted using a constant electron-hole competition factor $\zeta_2$ and the results are plotted in the right-hand side of Fig. 2.6. The best fits of the two parameters $N_{\text{eff}}^*$ and $\zeta_2$ are $N_{\text{eff}}^* = (3.6^{+10}_{-7}) \cdot 10^{22}$ m$^{-3}$ and $\zeta_2 = 0.63 \pm 0.05$. But as shown in Fig. 2.6, we can still get a reasonable fit of the measured points by choosing a value of $N_{\text{eff}}^*$ which is 4 times bigger than the value of the best fit. The additional point $\Gamma$ measured in this geometry, denoted by a triangle in the left-hand side of Fig. 2.6, is plotted in a separate graph because the effective EO coefficient and the effective dielectric constant are different for both geometries. For KNbO3 in the counter-propagating geometry the effective dielectric coefficient...
2.2. The single level model

is $c_{\text{eff}} = 796$ that is 23 times larger than in the co-propagating case ($c_{\text{eff}} = 34$), which also helps increasing the precision for the determination of the trap density as was mentioned before. The determination of $N^*_{\text{eff}}$ using all data points (including the point measured in the counter-propagating geometry) gives the following result: $N^*_{\text{eff}} = (6.5 \pm 0.4) \times 10^{22} \text{ m}^{-3}$ and $\zeta_2 = 0.58 \pm 0.01$. Comparing with the previous result we see that the additional point leads to a higher trap density and a lower error. This can also be seen considering the theoretical curves plotted in the left-hand side of Fig. 2.6 which strongly deviate from the measured point when different trap densities are considered.

When we calculate the effective trap density $N^*_{\text{eff}}$ considering only the point measured in the counter-propagating geometry without taking into account electron-hole competition we obtain the following result: $N^*_{\text{eff}} = (4.2 \pm 0.4) \times 10^{22} \text{ m}^{-3}$. The trap density $N^*_{\text{eff}}$ measured in the counter-propagating geometry only, which has to be considered as a lower bound, is still bigger than the trap density measured in co-propagating geometry, but within its accuracy range.

The counter-propagating geometry is therefore adequate as a quick characterization method in which the quality of a crystal for its capabilities of build up strong photorefractive gratings can be estimated by a one point experiment. This can be of interest if a large number of crystals has to be characterized.

2.2.4 Z-scan measurement

When a single light beam propagates in a photorefractive material the inhomogeneous intensity profile produces an inhomogeneous change in refractive index. The profile of the refractive index change is similar to a graded index lens and can focus (or defocus) the beam itself. This effect is called self-focusing and was first observed in photorefractive materials by Ashkin [25] et. al. in 1966. If the light induced lens exactly compensates the diffraction of the beam, we obtain an invariant intensity profile along the beam propagation direction. If this is achieved one can speak of a spatial soliton (see Appendix A). For thin crystals the photorefractive effect builds up a weak light induced lens which will not change the intensity profile inside the crystal but only act on the phase of the outcoming beam. The characterization of the light induced lens allows to determine the photoconductive and photogalvanic properties of the investigated crystal. Z-scan experiments[72] are a standard technique for characterization of third-order nonlinearity and can be used for the measurement of photorefractive light induced lenses. In this kind of experiments the investigated platelet sample is moved through the waist of a Gaussian beam along the propagation direction. The far field beam width is then determined for each position of the waist with respect to the sample. Compared to conventional electric photoconductivity measurements, the photorefractive Z-scan measurements presents the advantage of a much higher precision for material with very low conductivities and that parasitic effect like screen of the applied field at the electrodes does not affect the measurements. In addition the method works well also for very small samples, which is not the case for wave-mixing experiments.
Photorefractive nonlinearity

When a photorefractive material is illuminated inhomogeneously an internal space charge field \( E_{sc} \) builds up due to a redistribution of photo excitable free charge carriers (see section 2.1). This space charge field produces a spatial variation of the refractive index through the electro-optic effect. The field \( E_{sc} \) can be calculated starting from the Kuhktarev-Vinetskii Eqs. (2.13)-(2.16) which assumes a single impurity level in the band gap and a single charge carrier species. Making the assumptions that the space charge field is due only to trapped charge carriers, that the beam width is constant in the propagation direction \( z \), and that the beam width is much larger than the Debye screening length (2.28), the following general equation is obtained (see Appendix A and Ref. [73])

\[
\nabla \cdot \mathbf{E}_{sc} + \nabla \cdot \ln(I + I_D) \cdot \mathbf{E}_{sc} = -(E_0 - E_{ph}) \frac{\partial}{\partial x} \ln(I + I_D) + \frac{k_B T}{e} \frac{\nabla^2 I}{I + I_D}, \tag{2.40}
\]

where \( T \) is the absolute temperature, \( k_B \) is the Boltzmann constant, \( \nabla \cdot = (\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, 0) \) denotes the projection of the gradient in the \( x - y \) plane, which is normal to the beam propagation \( z \), and the \( x \) coordinate is parallel to the homogeneous externally applied electric field. The \( x \)-axis also coincides with the crystal polar axis and in this case the photogalvanic field \( E_{ph} \) (see Ref. [19]) is also parallel to \( x \). The intensity of the beam is \( I \) and \( I_D \) is the equivalent dark intensity. \( E_{ph} \) and \( I_D \) are the fundamental parameters for self-focusing and soliton observation and are specific to each crystal.

The internal homogeneous field \( E_0 \) is produced by an external voltage \( V_{ext} \) applied in the direction of the \( +x \)-axes by

\[
E_0 = \xi V_{ext} / d \tag{2.41}
\]

where \( d \) is the electrode spacing and \( \xi \leq 1 \) is the reduction factor for the screening effect at the electrodes[74]. The photovoltaic pseudo field produced by the photogalvanic effect is defined as

\[
E_{ph} = \frac{\gamma N_A L_{ph}}{\mu}, \tag{2.42}
\]

where \( \mu \) is the mobility and \( \gamma \) the recombination rate of the excited charge carrier, \( N_A \) is the density of the ionized donors in the dark and \( L_{ph} \) is the photogalvanic drift length. The sign of \( E_{ph} \) is positive for a photogalvanic current pointing in the \( -x \) direction.

The dark intensity \( I_D \) is defined as the light intensity at the primary wavelength \( \lambda \) which produces a change in conductivity equal to the conductivity in the dark. \( I_D \) can be increased by an incoherent homogeneous illumination \( I_0 \) at another wavelength \( \lambda_0 \)

\[
I_D = \frac{\beta}{s(\lambda)} + \frac{s(\lambda_0)}{s(\lambda)} I_0, \tag{2.43}
\]

where \( \beta \) is the dark generation rate and \( s(\lambda) \) the photoionization constant of the charge carriers upon illumination with light of wavelength \( \lambda \).
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Note that in Eq. (2.10) the \( x \) and \( y \)-axis are not equivalent. This means that the cylindrical symmetry of the propagating beam is broken, therefore we may observe a different focusing behavior in the two transversal directions [75]. The differential Eq. (2.10) can be simplified considering the special case of light intensity distribution which does not depend on the \( y \) direction. Experimentally this situation is easily achieved using cylindrical focusing. We then obtain

\[
E_{sc} = -(E_D - E_{ph}) \frac{I}{I + I_D} + \frac{k_B T}{e} \frac{\partial I}{\partial x}.
\]

The last term of Eq. (2.44) is the diffusion term and is due to the thermal diffusion of the excited charge carriers. This term can be often neglected: for a Gaussian beam profile of 10 \( \mu \)m width and the ratio \( I/I_D < 100 \), where \( I \) is the maximum intensity of the beam we obtain at room temperature a field smaller than 100 V/cm. The diffusion field scales inversely proportional to the beam width and logarithmically in \( I/I_D \). Because it is antisymmetric with respect to the \( x \) axis, its main effect is a lateral bending of the beam.

By neglecting diffusion, the phase change \( \Delta \Phi_{NL} \) produced by the photorefractive space-charge field through the electro-optic effect is given by

\[
\Delta \Phi_{NL} = \Delta \Phi_0 \frac{I/I_D}{1 + I/I_D},
\]

where \( \Delta \Phi_0 \) is

\[
\Delta \Phi_0 = -\frac{\pi l}{\lambda} n^3 r^{eff} (E_D - E_{ph})
\]

and \( \lambda \) is the wavelength of light, \( n \) is the refractive index, \( l \) is the length of the optical path in the crystal, and \( r^{eff} \) is the effective electro-optic coefficient. For the 1-dimensional case, the \( r^{eff} \) defined in Ref. [65] is valid. This can be verified considering the Fourier expansion of the soliton profile in the transversal direction when the same \( r^{eff} \) would apply for all the grating vectors in the series because they have the same direction. In a general two dimensional case the situation is more complex and still necessitates further studies. For \( I/I_D \ll 1 \) Eq. (2.45) can be approximated by

\[
\Delta \Phi_{NL} = \Delta \Phi_0 \frac{I}{I_D}
\]

which gives a Kerr type nonlinearity, that is the model used in previous works on photorefractive Z-scan experiments [76, 77, 78]. At higher ratios \( I/I_D \) the phase shift saturates, therefore we use the term saturable Kerr type nonlinearity for Eq. (2.45).

Theory of Z-scan

The Z-scan measurement is a single beam technique (see Fig. 2.8). The beam waist of a Gaussian beam with waist width \( w_0 \) is moved along the beam propagation axis, through the investigated material. For each position of the beam waist, the beam width is determined in the far field [72]. This is usually achieved by an aperture in
Chapter 2. The photorefractive effect

front of the detector. In Z-scan measurements one usually assumes that the intensity profile in the medium is constant along the propagation direction and the nonlinear interaction produces only a phase change of the beam. This assumption is justified if the following two conditions are satisfied: (i) The path of the beam in the crystal must be small compared to Rayleigh length $z_0 = \pi w_0^2 / \lambda$ of the Gaussian beam, and (ii) the beam deflection produced by the maximum nonlinear phase change $\Delta\Phi_{\text{max}}$ must be small compared to the beam width. This can be quantitatively formulated as

$$l/n \ll z_0,$$  \hspace{1cm} (2.48)

and

$$\Delta\Phi_{\text{max}} \ll k_n w_0^2 / l,$$  \hspace{1cm} (2.49)

where $l$ is the thickness of the medium, and $k_n = 2\pi n / \lambda$ is the wave-vector in the medium. We first notice that by solving the linear paraxial wave equation

$$-q \psi_t + -q \psi_z \frac{1}{2} \frac{k_n^2}{\psi} = 0$$

for a beam focused only in the $x$-direction we obtain the following form for the electric field wave amplitude $\psi$ normalized to its maximum value $\psi_0$

$$\psi(x, z) = \psi_0 \sqrt{\frac{w_0}{w(z)}} \exp \left[ -\frac{x^2}{w^2(z)} - i \left( \frac{k_n x^2}{2 R(z)} + \delta(z) \right) \right]$$ \hspace{1cm} (2.50)

with the following definitions

$$\delta(z) = -\frac{1}{2} \arctan \left( \frac{w(z)}{w_0} \right),$$ \hspace{1cm} (2.51)

$$w(z) = w_0 \sqrt{1 + \left( \frac{z}{z_0} \right)^2},$$ \hspace{1cm} (2.52)

and

$$R(z) = z \left( 1 + \left( \frac{z}{z_0} \right)^2 \right).$$ \hspace{1cm} (2.53)

Under the assumptions, stated above, the beam exiting the crystal at position $z_{\text{Out}}$ can be described as

$$\psi_{\text{Out}}(x) = \psi(x, z_{\text{Out}}) \exp \left( i \Delta\Phi_{\text{NL}}(x) \right),$$ \hspace{1cm} (2.54)

where phase change $\Delta\Phi_{\text{NL}}(x)$ is given by Eq. (2.45). Outside the nonlinear medium, the beam propagates in free space to the detector. Under the assumption that the path length $R_0$ from the crystal to the detector aperture is much longer than the transversal extension of the beam, we can calculate the Fraunhofer diffraction to find the amplitude at the aperture in front of the detector

$$\tilde{\psi}_{\text{Out}}(x_a) = \frac{1}{i \lambda R_0} \int dx \psi_{\text{Out}}(x) e^{-ik_n x_a x / R_0}.$$ \hspace{1cm} (2.55)
The Z-scan transmission $T$ defined as the normalized light power falling through the aperture is given by

$$T = \frac{P(\Delta n)}{P(\Delta n = 0)} = \int_{\text{aperture}} dx_a |\tilde{\psi}_{\text{out}}(x_a)|^2 / P(0). \quad (2.56)$$

We insert the photorefractive nonlinearity described by Eq. (2.15) in Eqs. (2.55)-(2.56) and obtain a complete equation describing the 1-dimensional photorefractive Z-scan experiment. An analytical solution for the case of Kerr-type nonlinearity ($I/I_D \ll 1$) and small phase changes ($\Delta \Phi_0 \ll 1$) can be found based on the Gaussian decomposition method [79] which consists in the decomposition of the complex electric field at the exit plane of the crystal in a sum of Gaussian beams. For small phase changes given with the parameter $\Delta \Phi_{\text{NL}} \ll 1$, the Z-scan transmission $T$ can be approximated by the first two terms of the Gaussian decomposition. For a slit which is much smaller than the beam width at the aperture plane, one obtains

$$T = 1 + \Delta \Phi_{\text{max}}^2 \frac{1}{\sqrt{(1 + x^2)(9 + x^2)} - \Delta \Phi_{\text{max}} \text{sign}(x) \sqrt{2\sqrt{(1 + x^2)(9 + x^2)} - 2x^2 - 6}} \quad (2.57)$$

where $x = z/z_0$ is the normalized position and $\Delta \Phi_{\text{max}} = \Delta \Phi_0 I/I_D$ is the maximum phase change. A calculation for the general case of elliptical beam is presented in Ref. [75]. From Eq. (2.57) the difference in position $\Delta z/z_0$ between minimum and maximum of the transmission $T$ and the maximum difference in transmission $\Delta T_{\text{max}}$ can be calculated. We obtain $\Delta z/z_0 = 2.25$ and $\Delta T_{\text{max}} = 0.44|\Delta \Phi_{\text{max}}|$. When we compare these results with the 2 transverse dimensional case (2D) of Ref. [72] we see that the distance between maximum and minimum in the 1D case is larger ($\Delta z/z_0 = 1.7$ in the 2D case) and also the change in transmission is slightly larger ($\Delta T_{\text{max}} = 0.41|\Delta \Phi_{\text{max}}|$ in 2D case). Comparing Eq. (2.57) with the numerical calculation we find that they are both equivalent for $I/I_D < 0.1$. The condition of small phase change is thus fulfilled in most practical cases.

In a general case the equation can be evaluated through numerical integration. The results for the Z-scan transmission $T$ through an aperture much smaller than the beam width are presented in Fig. 2.7 for different values of $I/I_D$ with constant $\Delta \Phi_0 = 1$. The shape of the Z-scan curves strongly depends on the ratio $I/I_D$. For $\Delta \Phi_0 = 1$ the curves do not show the inversion symmetry with respect to $z = 0$ which is present in small phase calculation presented above. A large (de)focusing effect is desirable both in Z-scan measurements and in soliton production, therefore we were studying the dependence of the transmission variations on different beam intensities. Maximal effects are observed for the ratio $I/I_D \approx 6$ which is approximately twice the optimal value for soliton observation [80, 81]. For $I/I_D \ll 0.1$ we are in the Kerr regime (see Eq. (2.47)) and as expected a linear dependence of $\Delta T_{\text{max}}/\Delta \Phi_0$ is observed. For $I/I_D > 100$ the nonlinearity is saturated and only small phase changes can be produced in the center of the beam. In this intensity range the Z-scan transmission (see Fig. 2.7) shows an additional maximum and minimum.
Figure 2.7: Numerical calculation of the Z-scan transmission $T$ as a function of the normalized displacement $z/z_0$ of the sample relative to the beam waist for different values of the beam to dark-intensity ratio $\hat{I}/I_D$ and for a maximum phase shift $\Delta \Phi_0 = 1$.

Experiments in Fe doped KNbO$_3$

The set-up for the Z-scan experiment is schematically shown in Fig. 2.8. A beam of a 5 mW He-Ne laser is focused by a cylindrical lens with a focal length $f = 100$ mm to produce a beam with a beam waist parameter $w_0 = 30 \mu$m and the corresponding Rayleigh length $z_0 = 4.4$ mm. An iron doped KNbO$_3$ crystal (100 weight ppm iron in the melt) is moved along the beam propagation direction $z$ by means of a translation stage. The crystal dimensions are $(a \times b \times c) = 4.08 \text{ mm} \times 1.69 \text{ mm} \times 5.49 \text{ mm}$. The external field $E_{\text{ext}} = V_{\text{ext}}/d$ and beam polarization were parallel to the crystallographic $c$ axis. After traversing the crystal the central part of the beam is selected in the far field using an aperture and collected on a detector by a lens. The distance between the crystal and the detector aperture, which has to be much larger than the Rayleigh length, was chosen to be $R_0 = 1$ m. As aperture we used a slit which was placed parallel to the axis of the cylindrical lens. In order to maximize the relative change in intensity on the detector during the scan we have chosen a slit width of $s = 1$ mm which is much smaller than the beam width at the aperture plane.

In our set-up we can also use incoherent background illumination. A multi-line argon ion laser ($\lambda_0 \approx 488 - 514$ nm) and a beam splitter (see Fig. 2.8) are used to illuminate the crystal as homogeneously as possible. The input beam was chopped and the signal detected with a digital lock-in amplifier. This allowed us eliminate the undesired signal produced by the scattering of the Ar$^+$ laser beam.

In Fig. 2.9 we present the experimental result for a Z-scan measurement without
2.2. The single level model

The single level model

cylindrical lens beam splitter photodiode aperture

Figure 2.8: Scheme of the photorefractive Z-scan experiment. The KNbO₃ crystal is moved through the focus of 1-D Gaussian beam with intensity \( I \), produced by a cylindrical lens. The central part of the beam is collected on a photodiode by the spherical lens after traversing the aperture. The crystal can be illuminated by an additional homogeneous light beam \( I_0 \) using the beam splitter.

1.15
1.10
1.05
1.00
0.95
0.90
0.85

Figure 2.9: Z-scan transmission \( T \) measurement for a beam intensity \( I = 10 \text{ W/cm}^2 \) with no external field (\( E_{\text{ext}} = 0 \)) and without additional homogeneous illumination (\( I_0 = 0 \)). The measurements are denoted by points and the solid line is the theoretical curve with the photovoltaic field parameter \( E_{\text{ph}} = 1.3 \text{ kV/cm} \) and the dark intensity \( I_D = 2 \text{ W/cm}^2 \).
Figure 2.10: Maximum light transmission difference $\Delta T_{\text{max}}$ by the Z-scan measurement technique for different beam intensities $I$, with no external field ($E_{\text{ext}} = 0$), and no additional illumination ($I_0 = 0$). The measurements are denoted by points and the solid line is the theoretical curve with the photovoltaic field parameter $E_{\text{ph}} = 1.4$ kV/cm and the dark intensity $I_D = 2.5$ W/cm$^2$. The dashed line is the theoretical curve with the same parameters as above, however, neglecting the saturation of the nonlinearity.

the external field and a maximum intensity in the focal plane of $I = 10$ W/cm$^2$. For each position the crystal was first illuminated with homogeneous incoherent blue light with an intensity of 1 W/cm$^2$, in order to erase eventual rests of the space-charge modulations. After 30 s in the dark, the red beam was turned on and the intensity measured when the steady state was reached. The measurements indicate that the crystal without an external field produces a defocusing nonlinearity due to photogalvanic effect. The solid line in Fig. 2.9 is the result of a fit with a saturable Kerr-like nonlinearity (Eqs. (2.15)-(2.16)) with the following parameters: $I_D = (2 \pm 1)$ W/cm$^2$ and $E_{\text{ph}} = (1.3 \pm 0.1)$ kV/cm. This corresponds to a maximum refractive index change of $\Delta n_0 = \frac{1}{2} \frac{\Delta T_{\text{max}}}{2\pi} = 4.0 \times 10^{-5}$. These values are in good agreement with the ones calculated from photoconductivity measurements and measurements of the internal electric field by determination of the refractive index change produced through the electro-optic effect.

A further comparison with theory is presented in Fig. 2.10 where the maximum variation of the transmission $\Delta T_{\text{max}}$ is plotted against the beam intensity $I$. From a fit we obtain $I_D = (2.5 \pm 0.5)$ W/cm$^2$ and $E_{\text{ph}} = (1.4 \pm 0.1)$ kV/cm. The error of the dark intensity $I_D$ is reduced to 50% of the error determined by considering a single Z-scan experiment. The shape of the theoretical curve in Fig. 2.10 is a consequence of the saturation of the photorefractive nonlinearity. Without saturation the theoretical
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Figure 2.11: Maximum difference $\Delta T_{\text{max}}$ of the Z-scan transmission for different external fields $E_{\text{ext}}$. A beam intensity $I = 10$ W/cm$^2$, and no additional illumination ($I_0 = 0$) were used for these measurements which are denoted by points. The solid line is the theoretical curve with the photovoltaic field $E_{\text{ph}} = 1.4$ kV/cm and the screening factor $\xi = 0.4$.

curve would be a straight line (see dashed curve) and a simultaneous determination of $I/I_D$ and $E_{\text{ph}}$ would not be possible. The precision in the determination of $I_D$ can be increased considering also experimental points for $I/I_D > 3$, using the fact that $\Delta T_{\text{max}}$ presents a maximum. A stronger laser would be needed to reach that region.

In addition to varying the beam intensity $I$ it is also interesting to increase the dark intensity $I_D$ by illuminating the crystal with an incoherent homogeneous light beam. Note that no additional photovoltaic field is produced when $I_0$ is homogeneous throughout the crystal. We were measuring the Z-scan transmission for different levels of the homogeneous illuminations $I_0$ in the range of $I_0 = 0$ to $I_0 = 1.14$ W/cm$^2$ and compared the measurements with theory to obtain a photoionization constant ratio of $s(\lambda_0)/s(\lambda) = 16 \pm 1$. This is an expected result because it is well known that most photorefractive crystals are much more sensitive in the blue-green spectral region.

The photorefractive nonlinearity can be changed by applying an external field (see Eq. (2.44)). We applied the field $E_{\text{ext}}$ in the direction of the spontaneous polarization in order to avoid depolarization of the crystal. In this geometry $E_{\text{ext}}$ produces an effect of opposite sign with respect to the photogalvanic effect, because the photogalvanic current is flowing in the direction opposite to the spontaneous polarization. This prediction is also reproduced in our experiments presented in (Fig. 2.11), where the maximum change in transmission $\Delta T_{\text{max}}$ decreases linearly
with increasing external electric field. For a field $E_{\text{ext}} = (3.4 \pm 0.3) \text{ kV/cm}$ the nonlinear effect vanishes, whereas by applying an external field larger than 3.4 kV/cm we observe a reversal of the sign of the photorefractive nonlinearity thus producing a focusing nonlinearity. In this way it is possible to produce a focusing refractive index change of $\Delta n_0 = 1.1 \times 10^{-5}$ for every kV/cm of applied field.

The zero-crossing point in Fig. 2.11 can be predicted from Eq. (2.44) for $E_0 = E_{\text{ph}}$. From this relation and our definition of screening Eq. (2.11) we determine the screening factor $\xi$ as the ratio $E_{\text{ph}}/E_0$ which has the value of $\xi = 0.4 \pm 0.07$ in the case of no external illumination. As known, the screening of the external field at the electrode is particularly strong in doped crystals.

To summarize, the one dimensional Z-scan experiments can be used to determine the dark intensity, the photovoltaic field parameter, the screening factor of the external field, and the photoionization constant ratio between the Z-scan probing beam and the homogeneous illumination. These parameters strongly influence the photorefractive soliton formation, therefore the simple experiments presented here are a valuable tool to optimize materials suitable for applications based on photorefractive self-focusing. The method allows also a precise determination of the photovoltaic field parameter which is a key parameter for holographic storage in materials which exhibit a strong photogalvanic effect like LiNbO$_3$ and LiTaO$_3$. An additional advantage of Z-scan is that the technique can be applied on very small ($200 \times 200 \times 200 \mu\text{m}^3$) samples.

### 2.3 Interband photorefraction

Interband photorefraction is a very effective method to produce fast holographic gratings. If the energy $h\nu$ of the incident photons is larger than the band gap energy $E_B$ of the material, electrons in the valence band can be directly photoexcited in the conduction band (see Fig. 2.12). As the number density of the electron which can be excited can be very large this process presents a very strong absorption. The resulting photorefraction effect will be very fast (several orders of magnitude faster than in conventional photorefraction), and present thin gratings which are robust to illumination by light with longer wavelength. These properties are particularly well suited for optical parallel processing demonstrations like optical controlled coherent to incoherent conversion, light induced wave-guiding, or optical joint Fourier transformed correlation. The first two have been recently demonstrated in KNB0$_3$ using ultra violet light with very promising results, whereas the third one is the topic of chapter 5.2.

#### 2.3.1 Model for the interband photorefractive effect

In the interband model the photorefractive space charge field $E_{sc}$ is driven by the highly efficient direct band-to-band photoexcitation. In this first model we will neglect the eventual presence of traps in the band gap. The model is valid in the case that excited charge carriers concentration generated by interband photoexcitation is much higher than the density of the neglected traps. The interband process is
2.3. Interband photorefraction

The conduction band and the valence band are shown in Figure 2.12. The Fermi level lies between them. Photoexcitation of an electron from the valence band to the conduction band generates a mobile electron in the conduction band and simultaneously a mobile hole in the valence band (a). Charge transport of the mobile electrons and holes occurs in the conduction band (b). Interband recombination of an electron in the conduction band with a hole in the valence band (c).

The band scheme of the interband photorefractive process is described by the following equations:

\[
\begin{align*}
\frac{\partial n}{\partial t} &= gI - \gamma_{\text{dir}}np + \frac{1}{e} \nabla J_e, \\
\frac{\partial p}{\partial t} &= gI - \gamma_{\text{dir}}np - \frac{1}{e} \nabla J_e, \\
J_e &= en\mu_e E_{\text{sc}} + k_B T \mu_e \nabla n, \\
J_h &= en\mu_h E_{\text{sc}} - k_B T \mu_e \nabla n, \\
\nabla (\varepsilon_0 eE_{\text{sc}}) &= e(p - n),
\end{align*}
\]

where \( n \) and \( p \) are the free electron and hole density numbers, \( J_e \) and \( J_h \) are the electron and hole current density vectors, \( \gamma_{\text{dir}} \) is the band-to-band recombination constant, and

\[
g = \frac{\alpha}{h\nu} \Phi
\]

is the band-to-band photoexcitation rate introduced assuming that the valence band is not appreciably depleted by the photoexcitation. The constant \( \alpha \) is the linear absorption in a trap free crystal, \( h \) is Planck’s constant, \( \nu \) the frequency of the light related to the wavelength by \( \nu = c/\lambda \), and \( \Phi \) is the quantum efficiency which in the following is assumed to be \( \Phi = 1 \). Compared to the single level model (see Eqs. (2.13)-(2.16)) the interband model presents one equation more. The model shows...
however a higher degree of symmetry with respect to electron and hole density. In addition the equation present only 4 free parameters (the single level model has 5-7 parameters). Of the 4 free parameters 3 can be accessed through absorption and photoconductivity measurements.

2.3.2 Solution for homogeneous illumination

Under homogeneous illumination the spatial derivative in Eqs. (2.58)-(2.62) can be neglected. For the free charge carrier density we obtain

$$n_0(t) = p_0(t) = \frac{\sqrt{gI_0}}{\gamma_{dir}} \frac{1 - e^{-2\sqrt{gI_0}\gamma_{dir} t}}{1 + e^{-2\sqrt{gI_0}\gamma_{dir} t}},$$

(2.64)

whereas the decay in the dark, after the light is turned off is given by

$$n_0(t) = p_0(t) = \frac{\sqrt{gI_0}}{\gamma_{dir}} \frac{1}{1 + \sqrt{gI_0}\gamma_{dir} t}.$$

(2.65)

The photoconductivity in the interband model is given by electron and hole conduction

$$\sigma_{tot} = e(n_0\mu_e + p_0\mu_h) = en_0(\mu_e + \mu_h)$$

(2.66)

The steady state of the photoconductivity in the interband regime is proportional to $\sqrt{I_0}$. This in contrast to the single level model where the photoconductivity was proportional to $I_0$, and allows a distinction of the dominating photoconduction mechanism by photoconductivity measurements.

2.3.3 Photoconductivity measurement

The photoconductivity can experimentally be determined by applying an electric field $E$ (for example in the $x$ direction), measuring the electric current $I_{el}$ through the crystal and changing the light intensity in the transversal direction ($z$ direction). This is however directly possible only in very thin plate because of the large absorption. The absorption is strongly reducing the incident light intensity $I_0$ along the perpendicular of the light incidence surface direction $z$ following

$$I_a(z) = I_0 e^{-\alpha z}. $$

(2.67)

The electric current $I_{el}$ under illumination in an absorbing material is given by Ohm’s law by

$$I_{el}(I_0) = \int_0^d E\sigma_{tot}(I_0 e^{-\alpha z}) b \, dz$$

(2.68)

where $b$ is the crystal dimension in the $y$-direction, $d$ is the thickness in $z$-direction, and $E$ is the applied electric static field. If the incident light intensity $I_0$ is increased by a small amount $\Delta I_0$, the expression for the electric current can be rewritten as

$$I_{el}(I_0 + \Delta I_0) = \int_0^d E\sigma_{tot}(I_0 e^{-\alpha(z+\Delta z)}) b \, dz$$

(2.69)
where the property
\[ \Delta I_0 \approx -\alpha I \Delta z \] (2.70)
obtained by linearization of Eq. (2.67) was used. Calculating the difference \( \Delta I_e(I_0) = I_e(I_0 + \Delta I_0) - I_e(I_0) \) between the electric currents, and after some rearrangement of the integration boundaries, we obtain
\[ \Delta I_e(I_0) = Eb \left( \int_{\Delta z}^{0} \sigma_{\text{tot}}(I_0 e^{-\alpha z}) \, dz - \int_{d+\Delta z}^{d} \sigma_{\text{tot}}(I_0 e^{-\alpha z}) \, dz \right). \] (2.71)

In the case of strong absorption \( (d \gg 1/\alpha) \) the total conductivity \( \sigma_{\text{tot}} \) in the second integral of Eq. (2.71) can be replaced by the intensity independent dark conductivity, which is defined as \( \sigma_{\text{dark}} = \sigma_{\text{tot}}(I_0 = 0) \). Further for small light intensity changes \( (\Delta I_0 \ll I_0) \) the conductivity in the first integral of Eq. (2.71) can be assumed as constant over the integration range. For the photoconductivity \( \sigma_{ph} = \sigma_{\text{tot}} - \sigma_{\text{dark}} \) we finally obtain
\[ \sigma_{ph}(I_0) = -\frac{\alpha}{bE} \frac{\Delta I_e}{\Delta I_0/I_0} \] (2.72)
where we made use again of Eq. (2.70).

The photoconductivity in thick samples can therefore be calculated by the measurement of the \( I_e(I_0) \) characteristic after numerical differentiation. An experiment is presented in section 5.3 where the method is used to determine the photoconductivity of a Sn_2P_2S_6 crystal.

### 2.3.4 Solution for sinusoidal illumination

In analogy to the single level model, also for the interband model an analytical solution can be found for a continuous sinusoidal illumination. The resulting amplitude of the space charge field \( E_{sc} \) is given by
\[ E_{sc} = -im \frac{E_g}{A} \left[ E_D(E_{Rh} - E_{Re}) - iE_0(E_{Rh} + E_{Re}) \right] \] (2.73)
\[ A = (E_D + E_{Re} + E_{Rh})(E_D + E_g) + iE_0(E_{Re} - E_{Rh}) + E_0^2, \] (2.74)

where we made use again of Eq. (2.70).

The photoconductivity in thick samples can therefore be calculated by the measurement of the \( I_e(I_0) \) characteristic after numerical differentiation. An experiment is presented in section 5.3 where the method is used to determine the photoconductivity of a Sn_2P_2S_6 crystal.
recombination takes place. In the case that no external field is applied the space charge field simplifies to

\[ E_{sc} = -im \frac{E_{gf} E_D (E_{Rh} - E_{Re})}{(E_D + E_{Re} + E_{Rh})(E_D + E_{gf})} \quad (2.77) \]

Eq. (2.77) is a little bit more complex than Eq. (2.23), valid for the conventional single level model. Different regimes dominated by one of the fields \( E_{gf}, E_D, E_{Rh}, \) and \( E_{Re} \) are possible. In interband photorefraction the steady state of the space charge field depends on the light intensity \( I_0 \). For high intensities \( (E_{gf} \gg E_D) \) the \( E_{sc} \) will grow proportional to \( \sqrt{I_0} \).

The dynamic of the build-up is given by a double exponential function. Where the build-up time constant is proportional to

\[ \tau_{lb} \propto \frac{1}{\sqrt{\gamma_{dir} g I_0}} \quad (2.78) \]

The exact solution is reported in detail in Ref.[59].

The build-up time of interband photorefractive effect which is typically around 100 \( \mu s \) in \( \text{KNbO}_3 \) at 100 mW/cm\(^2\) can be improved further by the use of pulsed illumination. This will be the topic of next section.

### 2.4 Interband photorefraction with pulsed light

Illuminating a photorefractive material with pulses instead of continuous illumination allows a deeper insight into the charge transport process because photoexcitation and charge transport can then be considered as separate processes. In addition, the high density of excited charge carriers produced by the light pulse can dramatically reduce the build-up time and the maximum amplitude of the space charge field modulation.

Under the assumption that charge transport can be neglected during the pulse duration and considering the symmetry with respect to \( n \) and \( p \) of the equation and boundary conditions \( n(x,t=0)=p(x,t=0)=0 \), Eqs. (2.58)-(2.62) can be simplified to

\[ \frac{\partial n}{\partial t} = g I(t) - \gamma_{dir} n^2. \quad (2.79) \]

For a general pulse shape \( I(t) \) Eq. (2.79) can be solved by numerical integration. Approximating the pulse with the rectangle

\[ I(x,t) = \begin{cases} 0 & : t < 0 \\ \bar{I}(x) & : 0 \leq t \leq \tau_{pulse} \\ 0 & : t > \tau_{pulse} \end{cases} \quad (2.80) \]

where \( \bar{I} \) is the instantaneous maximum intensity and \( \tau_{pulse} \) is the pulse duration, Eq. (2.79) presents following solution

\[ n(x) = p(x) = \sqrt{\frac{g \bar{I}(x)}{\gamma_{dir}}} \frac{1 - e^{-2\sqrt{g \bar{I}(x) \gamma_{dir} \tau_{pulse}}}}{1 + e^{-2\sqrt{g \bar{I}(x) \gamma_{dir} \tau_{pulse}}}} \quad (2.81) \]
which gives the electron and hole densities at the end of the pulse. For short pulses with moderated power \( (g \gamma_{\text{dir}} \bar{I} \tau_{\text{pulse}}^2 \ll 1) \), which is the case in the experiments presented in this work, Eq. (2.81) can be simplified to

\[
n(x) = g \bar{I}(x) \tau_{\text{pulse}}.
\]  

The density of excited electrons or holes is then directly proportional to the pulse fluence \( F \approx \bar{I} \tau_{\text{pulse}} \).

After the hole and electron density modulation are built up by the laser pulse, the charge carriers will move and recombine. Let’s first consider the case of recombination only, corresponding to the case for a pulse with homogenous illumination.

### 2.4.1 Solution for homogeneous illumination

For a spatial constant light intensity the time dependence of the free electron density will have a similar form as the interband dark decay (see Eq. (2.65)). The initial charge carrier density \( n_0(t = 0) \) will however be given by Eq. (2.82). We obtain

\[
n_0(t) = p_0(t) = \frac{gF}{1 + g \gamma_{\text{dir}} F t}.
\]  

Due to the strong absorption of the illuminating light the fluence \( F \) will change with

\[
F(z) = F_0 e^{-\alpha z}
\]  

for the light penetrating in the crystal. A way to take into account this effect was already described in section 2.3.3. A second method which allows a direct interpretation of conductivity measurements can be obtained integrating Eq. (2.83) in the \( z \) direction. For the electric current \( I_{el} \) we obtain

\[
I_{el} = e(\mu_e + \mu_h) b E \ln \left(1 + \frac{g \gamma_{\text{dir}} g F_0 t}{\alpha \gamma_{\text{dir}} t}\right).
\]  

For \( t \rightarrow 0 \) Eq. (2.85) becomes

\[
I_{el} = \frac{e}{h\nu}(\mu_e + \mu_h) b E F_0
\]  

which presents only the sum of the mobilities \( \mu_e + \mu_h \) as experimental unknown.

### 2.4.2 Solution for sinusoidal illumination

The spatially modulated laser pulse produces a sinusoidal modulation of the free electron \( n(x) \) and holes \( p(x) \) density. As both densities are equal, the initial space charge field modulation will be \( E_{sc}(t = 0) = 0 \). Driven by different diffusion rates of holes and electrons and the different drift directions for an external applied field, \( E_{sc} \) will start to raise. The growth of the space charge field will then be contrasted by the recombination of electron and holes and by the electric force of the modulated space charge field on the mobile charge carriers.
Considering the first Fourier-component of the free electron density \( n_1 = n_{10}(t)e^{K_x x} \) and of the hole density \( p_1 = p_{10}(t)e^{K_x x} \), Eqs. (2.58)-(2.62) can be simplified to

\[
\frac{\partial n_1}{\partial t} = \frac{n_0(t)}{n_0(0)} \left[ -\tau_{\text{dir}}^{-1}(n_1 + p_1) + \tau_{\text{Di},e}^{-1}(p_1 - n_1) \right] + \left[ i\tau_{\text{drift},e}^{-1} - \tau_{\text{D},e}^{-1} \right] n_1 \tag{2.87}
\]

\[
\frac{\partial p_1}{\partial t} = \frac{n_0(t)}{n_0(0)} \left[ -\tau_{\text{dir}}^{-1}(n_1 + p_1) - \tau_{\text{Di},h}^{-1}(p_1 - n_1) \right] - \left[ i\tau_{\text{drift},h}^{-1} + \tau_{\text{D},h}^{-1} \right] p_1 \tag{2.88}
\]

\[
E_{sc} = \frac{-ie}{K_o c_{0\text{eff}}} (p_1 - n_1) \tag{2.89}
\]

where for a better physical understanding we have introduced the charge carrier life time

\[
\tau_{\text{dir}} = \frac{1}{\gamma_{\text{dir}} n_0(t = 0)} \tag{2.90}
\]

the drift time

\[
\tau_{\text{drift},h/e} = \frac{e}{K_o \mu_{h/e} E_0} \tag{2.91}
\]

which describes the time needed by a charge carrier to move a distance \( \frac{\lambda}{2\pi} \) under the effect of an external applied field \( E_0 \), and the diffusion time

\[
\tau_{\text{D},h/e} = \frac{e}{k_B T \mu_{h/e} K_g^2} \tag{2.92}
\]

which is the time needed for the charge modulation to relax by diffusion. The Maxwell dielectric relaxation time

\[
\tau_{\text{Di},h/e} = \frac{c_{0\text{eff}}}{e \mu_{h/e} n_0} \tag{2.93}
\]

An analytic solution for the first Fourier-component of the space charge field \( E_{sc} \), as presented in section 2.3.1, is no longer possible under pulsed illumination, because of the explicit time dependence of the mean charge density \( n_0(t) \) (described by Eq. (2.83)). Some insight in the process can be obtained by considering the time constants introduced above separately.

The life time \( \tau_{\text{dir}} \) is responsible for the recombination of the homogeneous free charge carrier density and therefore defining the maximum time scale of the interband process. The recombination can not directly reduce the space charge field \( E_{sc} \) which depends only on the difference \( p_1 - n_1 \).

The dielectric relaxation described by the time \( \tau_{\text{Di},h/e} \) is counteracting the build-up of the space charge field produced by the other effect. Its effect is dependent on the space charge field only.

Under the influence of an external electric field \( E_0 \) the modulated charge carrier moves with a constant velocity given by \( \mu_{h/e} E_0 \). The direction and the velocity of
this movement depends on the charge carrier type. This will result in an oscillation of space charge field in the time domain with a period of

\[ T_{osc} = \frac{4\pi \tau_{drift,h} \tau_{drift,e}}{\tau_{drift,h} - \tau_{drift,e}}, \] 

which can be observed provided that \( T_{osc} \ll \tau_{dir} \). In the case that no external fields are applied (\( E_0 = 0 \)) a space charge field can only be produced if the diffusion time of electrons and holes is different. This is described by the diffusion times \( \tau_{D,h/e} \). The difference of the reciprocal diffusion times is responsible for the build-up of the space charge field, whereas the sum of the reciprocals will limit the time frame of the whole process. In the case where the diffusion times are \( \tau_{D,h/e} \ll \tau_{dir} \), Eqs. (2.87)-(2.89) can be solved and the solution is

\[ E_{sc} = \text{im} \frac{e g F_0}{K g_{0eff}} \frac{\tau_{D,h}^{-1} - \tau_{D,e}^{-1} - \Gamma_B}{\Gamma_C} \left( e^{-\left(\Gamma_A + \Gamma_C\right)t/2} - e^{-\left(\Gamma_A - \Gamma_C\right)t/2} \right), \] 

\[ \Gamma_A = \tau_{D,h}^{-1} + \tau_{D,e}^{-1} + \tau_{D,h}^{-1} + \tau_{D,e}^{-1}, \] 

\[ \Gamma_B = + i \left( \tau_{drift,h}^{-1} + \tau_{drift,e}^{-1} \right) + \tau_{D,h}^{-1} - \tau_{D,e}^{-1} + \tau_{D,h}^{-1} - \tau_{D,e}^{-1}, \] 

\[ \Gamma_C = \sqrt{\Gamma_B^2 + 4 \tau_{D,h}^{-1} \tau_{D,e}^{-1}}. \]

The solution presents an oscillating behavior as expected for short drift times. In the case that no external field are applied (\( E_0 = 0 \)), the diffusion rates are the dominating terms and the solution reduces to the sum of exponential decays (see Fig. 2.13 b)).

The space charge field under of pulsed interband photoexcitation can be found for the general case by numerical integration of Eqs. (2.87)-(2.89) or directly of Eqs. (2.58)-(2.62) if the exact spatial dependence of \( E_{sc} \) is of interest. An example of such a simulation is reported in Fig. 2.13 a), where the interband recombination constant was varied over 10 orders of magnitude. The main effect of the recombination is to reduce the maximum amplitude of the space charge field, and shorten the duration of the space charge field.

In the next section the theory will be expanded to include effects produced by the presence of traps within the band gap.

### 2.4.3 Influence of traps

Traps in the band gap are generated by crystal defects and impurities. In the case of interband excitation the photon energy \( h\nu \) is high enough to excite all accessible defects. During the illumination, however, charge carriers will be excited mainly from band to band, because of the much higher cross-section of this process. For the recombination process the situation is much more complicated and different scenarios given by different relative recombination constants are possible. Let us first expand Eqs. (2.58)-(2.62) to the case of interband effect with a single additional trap level
Figure 2.13: Time dependence of space charge field amplitude $E_{sc}$ for the case of pulsed interband photoexcitation. a) Numerical solution of Eqs. (2.58)-(2.62) where the recombination constant $\gamma_{dir}$ is varied from $2 \cdot 10^{-15} \cdots 2 \cdot 10^{-25}$ m$^3$/s. The other parameters are $\lambda = 532$ nm, $\alpha = 30$ cm$^{-1}$, $\mu_e = 8 \cdot 10^{-8}$ m$^2$/Vs, $\mu_h = 12 \cdot 10^{-8}$ m$^2$/Vs, $\Lambda_g = 6.6 \mu$m, $\epsilon_{eff} = 300$, $F = 100 \mu$J/cm$^2$, and $E = 0$. b) Algebraic solution (2.95) which is valid for small values of $\gamma_{dir}$.

which can act simultaneously as electron or hole donor$^\dagger$. The equations are

\[
\frac{\partial n}{\partial t} = g I - \gamma_{dir}np + (s_e I + \beta_e)(N_D - N_D^+) - \gamma_e n N_D^+ + \frac{1}{e} \nabla J_e, \tag{2.99}
\]

\[
\frac{\partial p}{\partial t} = g I - \gamma_{dir}np + (s_h I + \beta_h)N_D^+ - \gamma_h p(N_D - N_D^+) - \frac{1}{e} \nabla J_h, \tag{2.100}
\]

\[
\frac{\partial N_D^+}{\partial t} = (s_e I + \beta_e)(N_D - N_D^+) - \gamma_e n N_D^+ - (s_h I + \beta_h)N_D^+ + \gamma_h p(N_D - N_D^+) - \gamma_{dir}np, \tag{2.101}
\]

\[
\begin{align*}
J_e & = en \mu_e E_{sc} + k_BT \mu_e \nabla n, \tag{2.102} \\
J_h & = en \mu_h E_{sc} - k_BT \mu_h \nabla n, \tag{2.103} \\
\nabla (e_0 e E_{sc}) & = e(N_D^+ + p - n - N_A). \tag{2.104}
\end{align*}
\]

In this equation set 8 additional parameters were introduced: the photoionization constants $s_{h/e}$, the electronic dark generation rates $\beta_{h/e}$ for holes and electrons, the recombination constant of the traps $\gamma_{h/e}$, the total donor concentration $N_D$, and the concentration $N_A$ of ionized donors in the dark. Following the argument that the photoexcitation cross-section for the interband process is much larger than the

$^\dagger$This is the most general case and can be investigated by photorefractive two beam coupling experiments (see section 2.2.3).
photoionization rates of the traps, we can choose $s_{h/e} = 0$ and $\beta_{h/e} = 0$. The total

$$\begin{align*}
\text{Figure 2.14:} & \quad \text{Numerical calculation of the space charge field } |E_{sc}|^2 \text{ for the case of pulsed interband photoexcitation with an additional trap level.} \\
& \text{The occupation } N_A/N_D \text{ of the trap level in the dark was changed from 0 to 1.} \\
& \text{The other parameters are } \lambda = 532 \text{ nm, } \alpha = 30 \text{ cm}^{-1}, \mu_e = 4.8 \cdot 10^{-7} \text{ m}^2/\text{Vs,} \\
& \mu_h = 8 \cdot 10^{-7} \text{ m}^2/\text{Vs}, \gamma_{\text{dir}} = 5 \cdot 10^{-20} \text{ m}^3/\text{s}, N_D = 2 \cdot 10^{-22} \text{ m}^{-3}, \\
& \gamma_e = 2 \cdot 10^{-21} \text{ m}^3/\text{s}, \gamma_h = 2 \cdot 10^{-18} \text{ m}^3/\text{s}, \Lambda_g = 6.6 \mu\text{m, } \epsilon_{\text{eff}} = 300, \ F = 100 \mu\text{J/cm}^2.
\end{align*}$$

donor concentration can be determined by chemical analysis or from the segregation coefficient \(s[A_\text{]}\) of the dopant if the crystal is intentionally doped, whereas \(N_A\) can be estimated from two beam coupling experiment (see section 2.2.3). We are left with the unknown recombination constant \(\gamma_{h/e}\). To investigate the importance of \(\gamma_{h/e}\) on the space charge field it is useful to concentrate on the density of the ionized traps \(N^+_D\) under homogeneous illumination. Under the additional assumption that the concentration of charge carriers \(n,p \gg N^+_D - N_A\), we can solve the rate equation for the \(n\) and \(p\) independently from \(N^+_D\). Note that the assumption is valid only for a time scale in the order of \(\tau_{\text{dir}}\). For the free charge carrier concentration Eq. (2.83) will still hold, and by inserting into Eq. (2.101) we obtain

$$N^+_D(t) = \frac{\gamma_h}{\gamma_e + \gamma_h} N_D + \left( N_A - \frac{\gamma_h}{\gamma_e + \gamma_h} N_D \right) \left( 1 + \gamma_{\text{dir}} g F_0 t \right)^{\frac{\gamma_h + \gamma_e}{\gamma_{\text{dir}}} \left( \gamma_e + \gamma_h \right)}.$$  \(2.105\)

The time dependence of the ionized trap density \(N^+_D\) is strongly modified by the factor \(\frac{\gamma_h}{\gamma_e + \gamma_h}\) of the exponent in Eq. (2.105). For \(\gamma_e + \gamma_h > \gamma_{\text{dir}}\) the relative change in ionized traps density will change faster than the free carrier density in the bands (see Eq. (2.83)). In the opposite case \(\gamma_e + \gamma_h < \gamma_{\text{dir}}\) the density of ionized traps will change only slowly compared to the free carrier density in the bands, and the
Chapter 2. The photorefractive effect

Figure 2.15: Numerical calculation of the space charge field $E_{sc}$ for the case of pulsed interband photoexcitation with an additional trap level. The spatial dependence of $E_{sc}$ is plotted for different times. $N_A = 1.2 \times 10^{-22} \text{ m}^{-3}$ whereas the other parameters are the same as in Fig. 2.11.

The influence of the traps can then be mostly neglected. The ratio $\frac{\gamma_h}{\gamma_e}$ determines the final value of $N_D^+$ for long times $t$.

The influence of traps under sinusoidal illumination is more challenging. We can apply the same method as described in section 2.1.2 and consider only the first Fourier components $n_1$, $p_1$, $N_{D,1}^+$ of $n$, $p$, and $N_D^+$ respectively. For $\frac{\partial N_{D,1}^+ \phi - n_1}{\partial t}$ which is proportional to the change in space charge field $\frac{\partial E_{sc}}{\partial t}$, we will obtain an expression which will not contain $N_{pv}$. This means that the traps do not have any effect as long as the assumption $n_p \gg N_D^+ - N_A$ holds, that is for times shorter than $\tau_{dir}$. For longer times, however, the band-to-band recombination is no longer independent on the traps, and charge modulations in the traps will start to appear. The magnitude and the dynamics of the space charge field generated by the traps depend in a complicated way on $\frac{\gamma_h}{\gamma_e}$, $\frac{2 \tau_{dir}}{\gamma_{sec}}$, $N_A/N_D$, $\tau_{D,h/e}$, $\tau_{D,i,h/e}$, and $\tau_{dir}$. An example of a numerical calculation presenting a double peak behavior (which was experimentally observed in Sn$_2$P$_2$S$_6$ (see section 5.3)) is presented in Fig. 2.14.

The influence of the reduction state is considered by changing the ratio between the concentration of ionized donors in the dark and the total donor concentration $N_A/N_D$. The space charge field arising from charge modulation in the trap level exhibits a very long life time because the only way the charges can redistribute is through thermal excitation. The spatial dependence of $E_{sc}$ is shown in Fig. 2.15 for different times and for the same parameter as in Fig. 2.14. For these parameters the charge modulation produced by the interband effect and the charge modulation caused by the traps are out of phase by $180^\circ$, producing the double peak behavior.
observed in Fig. 2.14.

In this section we have presented a theoretical analysis of interband photorefractive with pulsed light observed in Fig. 2.11. For the case where the carrier life time is much longer than the carrier diffusion time an analytical solution was found. Numerical simulations were also presented for the case of short carrier life time. The influence of traps is complex, due to the numerous parameters introduced for describing the trap level. The most important quantities influencing the interband effect were found to be the reduction state of the crystal $N_A/N_D$, the ratio between the recombination constants of holes and electrons $\gamma_h/\gamma_e$, the ratio between the sum of the recombination constants for holes and electrons, and the interband recombination constant $\frac{\gamma_{h}+\gamma_{e}}{\gamma_{ir}}$. In the dynamic of the space charge field, the influence of traps will be noticed by the appearance of long living space charge field modulations.

Even if the complete theory which describes the effect is complex, many important characteristics can be modeled in a simplified way when the order of magnitude of the material parameter are known from measurements. Powerful characterization methods for this purpose are absorption, photoconductivity, and photorefractive beam coupling experiments. The last two are preferably performed with pulsed illumination, because of the simpler theoretical analysis.
Chapter 3

Photorefractive holographic storage

In holographic three-dimensional storage the information is stored in the material volume in the form of holograms. The latter can be produced by fixing the interference patterns produced by the coherent superposition of the image beam (on which the information page to be stored was encoded) and the reference beam. The stored image can be reconstructed by illuminating the hologram with the reference beam. Multiple hologram can be written and read out by choosing different appropriate reference beams.

As compared to a conventional storage systems, were the information is stored in a two-dimensional area (RAM chip, magnetic disk, magneto-optic disk, compact disk, magnetic band, etc.) holographic storage has the advantage that the information can be recorded in the whole volume of the storage medium. The idea of holographic information storage in solids was first presented by van Heerden[1, 2]. Even though the basic principle of holographic storage is simple, there are multiple ways to encode the information on the image beam and to choose the reference beams. In the next section a concise description of the most important multiplexing techniques and their impact on the storage density is presented. In the following section (3.2) the influence of the spatial dynamics of the photorefractive nonlinearity on the storage density is discussed. Special emphasis will be given to the self-focusing effect which is acting on the image beam itself. The following sections describe the set-up of the holographic storage system which was built, and the experimental results achieved with different photorefractive materials.

3.1 Principles of holographic storage

The simplest way to look at holographic storage is to consider the grating written by the interference of two plane waves (see section 2.1.4) as one bit of information*. The maximum storage density is then given by the maximum number of grating

*The amplitude of the written grating can also be interpreted as an analogic value. Theoretical considerations about storage density[88] based on the signal to noise ratio of the read-out beam have shown that the storage density is maximized if the amplitude is interpreted as maximum three discrete levels, for the sake of simplicity in the following only two level 0 and 1 will be considered.
vectors $\mathbf{K}_{g,mn}$ which can be stored in the material. Following Wullert et al.\cite{9} the
equations for the electric field of the plane wave writing the hologram are

$$
E_m = E_0 e^{i\mathbf{k}_m \cdot \mathbf{x}} = E_0 e^{ik(m_1 x + m_2 y + m_3 z)}
$$

where $k = 2\pi/\lambda$. Because the wavelength $\lambda$ is fixed, these two equations must meet
the condition

$$
(m_1^2 + m_2^2 + m_3^2) = (n_1^2 + n_2^2 + n_3^2) = 1. \quad (3.2)
$$

In order to be distinguished, the grating vectors $\mathbf{K}_{g,mn} = \mathbf{k}_n - \mathbf{k}_m$ must be orthogonal
within the $k$-space of the storage medium, that is

$$
\begin{align*}
    k(n_1 - m_1) &= q \frac{\pi}{l_x} \\
    k(n_2 - m_2) &= r \frac{\pi}{l_y} \\
    k(n_3 - m_3) &= s \frac{\pi}{l_z}
\end{align*} \quad (3.3)
$$

where $q, r, s$ are integers and $l_x, l_y, l_z$ are the dimensions of the storage medium in
$x, y, z$ direction respectively. This second condition (Eq. (3.3)) allots a minimum
portion of the $k$ space for each hologram

$$
V_k = \frac{\pi^3}{l_x l_y l_z}. \quad (3.4)
$$

From Eq. (3.2) we know that the total available volume of $k$-space is spherical and
given by

$$
V_{tot} = \frac{1}{8} \left(\frac{4\pi}{3} k^3\right), \quad (3.5)
$$

where the factor $1/8$ takes into account degenerate states. The maximum number
of gratings $N$ which can be stored in the medium is found by division of the total
volume in $k$-space by the volume per grating

$$
N = \frac{4\pi}{3} \frac{l_x l_y l_z}{\lambda^3}. \quad (3.6)
$$

The storage density $\rho$ is given by the total number $N$ of stored grating divided by
the volume of the storage material

$$
\rho = \frac{4\pi}{3} \frac{1}{\lambda^3}. \quad (3.7)
$$

Evaluating Eq. (3.7) for a wavelength $\lambda = 532$ nm we obtain a storage density of
= 3 Tbyte/cm³. As we will see in following sections this simple estimation of the
storage density is far too optimistic. The real storage density is reduced by cross-
talk noise, which is generated by the light scattered from the non Bragg-matched
gratings. This is equivalent to the side lobes of the diffraction efficiency curve as
function of the read-out angle presented in Fig. 2.2.
3.1.1 Encoding the data on the image beam

Considering each bit of information encoded in the direction of a plane wave as described above, may appear a very impractical method to store data pages with several thousands or even millions of bits. This can however be achieved very simply by a lens in the Fourier transform arrangement put in front of a spatial light modulator. The written holograms are then called Fourier plane holograms (see Fig. 3.1 b)). A spatial light modulator is a two-dimensional array of optical modulators which can be independently controlled. The data can be encoded on the image beam either by amplitude modulation or phase modulation or both together\cite{89}. Amplitude modulation is the preferred method because the read-out image can be directly detected on a CCD-Camera.

A second possibility to encode the data on the image beam is the so called image plane holography, where the two-dimensional array of the spatial light modulator is imaged directly on the storage material (see Fig. 3.1 a)). Compared to Fourier plane holography this method presents a more favorable distribution of the light intensity in the storage material if amplitude modulated images are used. As drawback small localized defects in the storage material are directly mapped on the read-out image. This is not the case in Fourier plane holograms where the same defect would be

![Schematic setup for writing and reading holograms in photosensitive materials: a) Image plane hologram: the input of the spatial light modulator is imaged on the storage material. b) Fourier plane holograms: the spatial Fourier transformed of the input image is stored in the storage material.](image-url)
smeared out on the whole output image and therefore the output image can always
be recognized and only the spatial resolution is affected.

From a theoretical point of view image plane and Fourier plane holograms are
equivalent because they are related to each others by the spatial Fourier transform.
Consideration of cross-talk in particular arrangements and the dynamics of the holo-
graphic material make one preferable over the other.

3.1.2 Multiplexing methods

Holographic storage is interesting because many pages can be stored in the same
volume. As was pointed out by the simple consideration with two plane wave at
the beginning of section 3.1 multiple holograms can be stored if their grating vector
distributions are disjunct. This can be achieved by an appropriate set of reference
waves. The methods to produce this set of waves are called multiplexing techniques.
The mostly used is angular multiplexing, where the direction of the reference beam
is changed, for example by a rotating mirror. Other possibilities are wavelength
multiplexing where the wavelength of the image and reference beam is changed, or
phase code multiplexing where the reference beam consists of a set of orthogonal
phase coded images. Sometimes different methods are combined to achieve higher
storage capacities or because of the geometry of the system.

Angular multiplexing

In angular multiplexing the reference beam consist of a plane wave incident on the
storage material at different angles, and therefore different wavevectors $k_{ref,n}$ (see
Fig. 3.2 a). The angle of the reference beam can be changed by a rotating mirror
or an acusto-optic deflector. These components are often used in combination with
a telescope arrangement, allowing to change the direction of the beam maintaining
the incidence position on the storage material. Angular multiplexing can also be
achieved by rotating the storage medium for each object to be stored and fixed
angle between object and reference beam. The highest number of holograms stored
with angular multiplexing is 5000 and was reported by Mok\[90\]. The highest number
of stored holograms ever reported is 20000 and was achieved\[91\] by the combination
of 1000 angle multiplexed images spatially multiplexed at 20 different positions.

Wavelength multiplexing

In wavelength multiplexing the wavelength of the reference beam (a plane wave) and
the object beam are changed for each stored image. The technique depends critically
on the availability of fast tunable laser. Recently the technique was used with an
external cavity laser diode to store up to 27 images for a total wavelength tuning of
2 nm (see Ref. \[92\]). Reflection gratings are often used in wavelength multiplexing
to maximize the number of images that can be stored by the given wavelength
tuning range. The grating vectors $K_{g,n}$ are then oriented nearly parallel to the
wavevector $k_{ref,n}$ and $k_{img,n}$ of the reference beam and image beam respectively and
are of magnitude approximately twice that of $|k_{ref}|$ (see Fig. 3.2 b). This is the
3.1. Principles of holographic storage

Figure 3.2: Representation of multiplexing schemes for multiple hologram storage. The grating vectors of the stored holograms are denoted by $K_{g,n}$. Their unsharpness due to the transversal modulated image beams with wavevector $k_{img,n}$ are represented by the shaded spots. a) Angular multiplexing: The angle of the reference beam wavevector $k_{ref,n}$ is changed whereas the wavevector $k_{img,n}$ of the image beam is held constant. b) Wavelength multiplexing: The direction of $k_{ref,n}$ and $k_{img,n}$ is always the same but the length is changed by changing the wavelength of the light source.

maximum magnitude of $K_g$ which can be achieved by interference of two plane wave, and therefore a very efficient use of the $k$-space.

Phase-code multiplexing

In phase-code multiplexing the reference beam is expanded and modulated by a one or two-dimensional phase modulator. The method can be understood considering an angle multiplexed scheme were all the reference beams are present simultaneously. The image selectivity is achieved by changing the relative phases of the single plane waves. Different phase code (or phase images) are possible. The most ones used are the Walsh-Hadamard codes[93]. For a linear phase modulator with $N$ pixels, $N$ different images can be stored. The method is particularly interesting because there are no moving parts involved, and fast configuration could be achieved by the use of ferro-electric liquid crystal phase modulators. Based on phase-code modulation it is also possible to realize parallel optical addition, subtraction, and inversion operations of stored images. The method was demonstrated by Denz et. al.[94] and is based on the sub-sampling of the set of reference beams during the recall operation, which allows one to combine images in such a way that different linear combinations of the images can be realized at the output of the memory.
Shift multiplexing

Shift multiplexing\cite{95} is a method for holographic storage that uses non-planar reference beams. A hologram recorded with a spherical-wave reference becomes Bragg mismatched when it is translated with respect to the read-out beam. This effect can be used to multiplex holograms in the same recording medium. The required amount of translation $\Delta l$ between holograms is called shift selectivity and is typically in the order of a few to a few hundreds $\mu$m, depending on the geometry, the material thickness, and the distance of the focus of the spherical wave to the hologram. Shift multiplexing is particularly convenient for holographic three-dimensional disks\cite{96, 97}.

Random phase multiplexing

In random phase multiplexing the reference waves are generated by random inhomogeneity on the surface of an optical component. In Ref. \cite{98} for example, 100 holograms could be multiplexed by the use of a simple ground glass plate. The method is particularly interesting because of its simplicity: Only one translation stage is needed to move the ground glass. Another property of the method is the securing of stored data, because an exact random phase can only be generated using the original random mask. A random multiplexing method based on speckle patterns was demonstrated by Tebaldi et. al. in Ref. \cite{99}.

Spatial multiplexing

All the multiplexing methods described till here were based on the separation of the hologram grating vectors in the $k$-space. In spatial multiplexing the holograms are separated in real space. It is often used in combination with another multiplexing technique, and is specially interesting for holographic three-dimensional disks\cite{97}. The advantage is that as the holograms do not overlap in space inside the material, no cross-talk is produced, this however on the expenses of the storage density.

3.1.3 Cross-talk limited storage density

The storage density in holographic memories described by Eq. (3.7) has to be modified if light diffracted by non Bragg matched holograms during read-out is taken into account. The effect is called cross-talk noise and can be quantitatively characterized by the signal-to-noise ratio

$$\text{SNR} = \frac{I_{\text{image}}}{I_{\text{ct}}}$$

where $I_{\text{image}}$ is the intensity of the read-out image and $I_{\text{ct}}$ is the intensity of the light scattered by the non Bragg matched gratings. Note that beside cross-talk also other effect such as light scattering from material defects or the material dynamics during the recording process can affect the storage density. In this section we concentrate on cross-talk noise only, whereas the other effects limiting the storage capacity will be addressed in section 3.2.
3.1. Principles of holographic storage

The impact of cross-talk noise in holographic storage was first considered by Ramberg\cite{100} in 1972. In his work Fourier plane holograms produced by angle multiplexing in two orthogonal directions were considered. The method presented strong cross-talk noise due to the presence of degenerate gratings introduced by the unfavorable multiplexing scheme. The cross-talk in Fourier plane holograms with the standard angular multiplexing method was analyzed by Gu et. al.\cite{101} based on a scalar diffraction theory and simple statistical methods. The signal-to-noise ratio was found to be maximum for an angular separation between subsequent reference beams of

$$\Delta \theta = \frac{\lambda}{l_z \sin \theta} \quad (3.9)$$

where $l_z$ is the crystal thickness and $\theta$ is the angle between the $z$-axis and the wave-vector $\mathbf{k}_{ref}$ of the reference beam inside the medium. This can be qualitatively understood considering the $(\sin (x)/x)^2$ like diffraction function depicted in Fig. 2.2. Cross-talk noise is mainly produced by the holograms written by the reference beams adjacent to the read out beam. For the angle difference reported in Eq. (3.9) the light diffracted by an hologram read out with the adjacent reference beam has its minimum exactly where the Bragg matched hologram presents the maximum. The cross-talk was also found to be dependent on the position in the output plane under investigation. The $\text{SNR}$ was highest at the center of the image and lowest away from the center in the $y$ direction (see Fig. 3.1) near the border. For the worst case $\text{SNR}$ under the separation angle of Eq. (3.9) the following expression was found

$$\text{SNR} \approx \frac{l_z f}{N a_y} \quad (3.10)$$

where $N$ is the number of stored holograms, $f$ is the focal length of the Fourier lens (see Fig. 3.1 b)) and $a_y$ is the image width in the $y$ direction in the image or output plane. Let’s consider the impact of the $\text{SNR}$ on the storage density, which per definition is given by

$$\rho = \frac{a_x a_y N}{\delta a_x \delta a_y \sqrt{V}} \quad (3.11)$$

where $\delta a_i$ are the pixel spacing in the $i$ direction of the image plane. Introducing Eq. (3.10) in Eq. (3.11), and considering an optimal lens system for maximum storage, where the image extension $l_{x,y}$ in the Fourier plane is equal to the image width $a_{x,y}$, we obtain

$$\rho = \frac{f \lambda}{a_y \delta a_x \delta a_y \text{SNR} \lambda^3} \quad (3.12)$$

Analyzing the first three terms of Eq. (3.12) one can see that for the case of maximum storage density the terms are simply related to the numerical aperture $N_a$ of the lens by $N_a \approx \lambda / \delta a_i \approx a_y / f$. Finally we can write the storage density as

$$\rho = \frac{N_a}{\text{SNR} \lambda^3} \quad (3.13)$$
For a wavelength of $\lambda = 532$ nm, a bit-error rate of $10^{-9}$, which requires a $\text{SNR} \approx 75$, and assuming $N_a = 0.2$ we obtain a storage density of $> 2 \text{ Gbyte/cm}^3$, which is approximately 3 orders of magnitude less than the storage density expected by neglecting cross-talk. It is noteworthy that the maximum storage density is independent of the number of stored holograms and the number of pixels in the image. The maximum number of stored images is however limited by the required $\text{SNR} (3.10)$. The number of stored holograms can be increased by increasing the crystal thickness or reducing the number of pixels in the $y$ direction. Also the number of pixels in the $y$ direction is related to the $\text{SNR}$, whereas the number of pixels in the $x$ direction is limited by the aperture of the lens system. For both directions a large number of input pixels (for example $1000 \times 1000$) will require high quality multiple lens systems (see for example Ref. [102]).

The case of angle multiplexed image plane holograms was analyzed in Refs. [12] and [16]. They show that, in contrast to Fourier plane hologram, the $\text{SNR}$ is no longer dependent on the the position in the output plane. Maximum $\text{SNR}$ is still obtained with angular separation given by Eq. (3.9) and is given by

$$\text{SNR} \approx \frac{\delta a_y l_z}{\gamma \lambda^2 N} \quad (3.14)$$

where $\gamma = \int^ {+\delta a_y/\lambda} _{-\delta a_y/\lambda} \text{sinc}^2(p)|p|dp$ is a slowly varying logarithm-like function of $\delta a_y/\lambda$. As an estimation, taking $\delta a_y/\lambda = 10$, we have $\gamma \approx 0.5$. For the storage density one obtains

$$\rho = \frac{N_a}{\gamma \text{SNR} \lambda^3} \quad (3.15)$$

and is therefore approximatively twice as large as the storage density in the case of Fourier plane holograms. Whereas the scaling properties for the maximum number of holograms and the number of pixels per image are similar as for Fourier plane holograms. Strangely enough the $\text{SNR}$ reported in Ref. [12] qualitatively agree with the ones reported in Ref. [16] whereas quantitatively a $\text{SNR}$ for image plane holograms which is 5 orders of magnitude larger than for Fourier plane holograms is reported in the Ref. [16]. The results of Ref. [12] are also in agreement with Ref. [103].

Cross-talk was also analyzed in the case of wavelength multiplexing[15, 16, 92, 104]. For nearly antiparallel propagating image and reference beams (see Fig. 3.2 b)) the $\text{SNR}$ is maximum in the case the frequency shifts $\Delta \nu$ between subsequent reference and image beams frequencies is chosen as

$$\Delta \nu = c/l_z \quad (3.16)$$

where $c$ is the speed of light in vacuum. The $\text{SNR}$ in the case a large number of holograms ($N > 1000$) can be approximated by

$$\text{SNR} \approx 4f^2 \frac{a_y^2}{a_x^2 + a_y^2}. \quad (3.17)$$
As can be noted the SNR is no longer dependent on the image number N. For the storage density one obtains
\[
\rho \approx \frac{N\lambda}{l_z} \frac{1}{SNR} \frac{1}{\lambda^3},
\]  
(3.18)
where we have considered only small relative changes of the wavelength \( \lambda \). Transforming the frequency shift of (3.16) in a wavelength shift \( \Delta \lambda = -\frac{1}{\lambda^2} \), and introducing the tuning range \( \delta \lambda \) of the light source, the first term of Eq. (3.18) can be rewritten as
\[
\rho \approx \frac{\delta \lambda}{\lambda} \frac{1}{SNR} \frac{1}{\lambda^3},
\]  
(3.19)
Wavelength multiplexing is therefore critically dependent on the tuning range of the laser. In order to compete with the storage density of angular multiplexed systems a tuning range of \( \delta \lambda/\lambda > 0.1 \) is required. The number of stored images can be controlled by the crystal thickness as in angle multiplexing, whereas the maximum image size and the number of pixels are limited by the required SNR. In analogy to angle multiplexing, the SNR depends on the position in the output plane for Fourier plane holograms and is independent on the position and larger in the case of image plane holograms.

Cross-talk was also investigated for phase-encoded multiplexing in Refs. [105, 103, 14, 106, 93]. Phase-code multiplexing is very similar to angle multiplexing from a theoretical point of view[105, 103]. The angle separation between the reference beams is the same as Eq. (3.9) and the SNR and the storage density are twice the respective values in angle multiplexing. The storage density can however also be reduced by imperfections in the phase modulator, which has to satisfy very high requirements for the reciprocal phases (Phase error of less than \( 2\pi/100 \) are required).

To summarize we have seen that cross-talk noise can dramatically decrease (by three orders of magnitude, i.e. \( \approx 8 \, \text{Gbyte/cm}^3 \)) the storage density predicted by simple optical arguments. The highest storage densities can be achieved with image plane holograms and phase-encoded multiplexing. The last method suffers from the high requirements of the phase modulator needed for multiplexing. The storage density in angle multiplexed holograms is smaller by only a factor of two and represents a simpler solution from a technological point of view. Besides cross-talk there are other sources of noise and limitations of the storage density. They will be addressed in the following section.

### 3.2 Holographic storage in photorefractive materials

Volume holograms are produced by the interference of the object and reference beam in a photo-sensitive material. Photorefractive materials are particularly well suited for this purpose because they exhibit a high photosensitivity and because of the reversible nature of the effect. These facts make them especially attractive for real time holographic applications and also for storage applications because of the long
storage time in the dark. In holographic storage numerous holograms have to be stored in the same material. This can be achieved writing each hologram in a sequential way. However during the recording of a hologram, previously written holograms are partially erased. Homogeneously diffracting holograms can be obtained in two ways, by adjusting the writing time of the holograms carefully (sequential recording) or by recording the holograms for very short time but in multiple cycles (incremental recording). Recording scheme will be the topic of section 3.2.1. The fact that during recording of a hologram the already written ones are partially erased will also limit the diffraction properties of the holograms. It will be shown that the diffraction efficiency of the holograms will scale with $1/N^2$. For large numbers $N$ of stored holograms this means that highly sensitive image detection devices are required. The intrinsic scattering properties of the material will give the final limitation on the number of stored holograms. The impact of the photorefractive self-focusing effect (see Appendix A) on Fourier and image plane holograms which is limiting the maximum image resolution will be addressed in section 3.2.3.

### 3.2.1 Hologram recording schemes

The dynamics of a single photorefractive grating produced by the conventional photorefractive effect is given by the exponential build-up of Eq. (2.24). In materials which present a strong photogalvanic effect as for example in LiNbO$_3$ or LiTaO$_3$ the dynamics of the build-up presents a much more complicated structure (see Ref. [82]). For the sake of simplicity we hold on the simple exponential build-up of Eq. (2.24).

If more than one hologram have to be written, this can be done by writing the holograms sequentially one after the other. The build-up of the refractive index grating $\Delta n$ can be described by

$$\Delta n_w(t) = \Delta n_{\text{max}} \left(1 - e^{-t/\tau_w}\right)$$

where $\tau_w$ is time constant of the build-up and $\Delta n_{\text{max}}$ can be calculated from Eqs. (2.3) and (2.19). The decay during the writing of the next hologram is given by

$$\Delta n_e(t) = \Delta n_e(t_0) e^{-(t-t_0)/\tau_e}$$

where the image and reference beam are switched to the next image/reference pair at the time $t_0$, and $\tau_e$ denotes the time constant of the erasure process. In general, and especially in the case of strong photogalvanic material, the time constant of the erasure process $\tau_e$ and hologram build-up $\tau_w$ are not equal.

Writing all hologram sequentially for an equal time will result in holograms with non homogeneous $\Delta n$. The last written holograms will have the strongest $\Delta n$ whereas the first ones will have undergone the strongest erasure. A simple way

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1In the case of angular or phase-encoded multiplexing the holograms can not be stored simultaneously because of the parasitic grating produced by the interference of the multiple images and reference waves. This is however possible in wavelength multiplexing. Because each image/reference wave pair has a different wavelength. The total writing time of the hologram is then reduced by a factor $N$. 

---
to overcome this problem is to use variable writing times. For the hologram written earlier a longer writing time is chosen. The method is known as sequential storage procedure\cite{107, 8, 108}. The index change $\Delta n$ is equalized if the condition

$$
(1 - e^{-t_{k+1}/\tau_w}) = (1 - e^{-t_k/\tau_w}) e^{-t_{k+1}/\tau_e}
$$

(3.22)
is fulfilled. There is no analytical closed solution for Eq. (3.22) which has to be evaluated numerically, except for the special case that $\tau_e = \tau_w$. For this case one obtains

$$
t_k = \tau_w \ln \left( \frac{k}{k-1} \right),
$$

(3.23)

where $k > 1$ is the hologram sequence number. The maximum refractive index change per hologram $\Delta n_{\text{max},N}$ which can be achieved with this method is

$$
\Delta n_{\text{max},N} = \Delta n_{\text{max}} \frac{\tau_e}{\tau_w \frac{1}{N}}.
$$

(3.24)

The critical point of the sequential recording scheme is that the dynamics of the photorefractive material has to be known with a high accuracy to calculate correct exposure times $t_i$. Especially for the hologram at the beginning which are written near to the saturation, the $\Delta n$ can change dramatically because of the oscillatory behavior introduced by the photogalvanic effect (see Ref. [82]).

A second way to equalize the refractive index change of the holograms, is the so called incremental recording scheme \cite{109, 110, 111}. In this method all the holograms are written for the same time $\delta t < \tau_w$, but the writing procedure is repeated for $M$ cycles. The index modulation of the first hologram after the first recording cycle is

$$
\Delta n_{1,1} = \Delta n_{\text{max}} (1 - e^{-\delta t/\tau_w}) e^{-(N-1)\delta t/\tau_e}.
$$

(3.25)

After the second writing cycle the refractive index change of the same image is

$$
\Delta n_{2,1} = \Delta n_{\text{max}} (1 - e^{-\delta t/\tau_w}) (1 + e^{-\delta t/\tau_w} - (N-1)\delta t/\tau_e) e^{-(N-1)\delta t/\tau_e}.
$$

(3.26)

Proceeding in this manner, it can be shown that the index modulation for the $k$th image after $l$ cycles is given by

$$
\Delta n_{l,k} = \Delta n_{\text{max}} (1 - e^{-\delta t/\tau_w}) \left( \sum_{m=0}^{m=k-1} e^{-m\delta t/(\tau_w - (N-1)\delta t/\tau_e)} \right) e^{-(N-1)\delta t/\tau_e}.
$$

(3.27)

After some algebraic transformation and assuming that $N\delta t \ll \tau_e$, Eq. (3.27) can be simplified to

$$
\Delta n_{l,k} = \Delta n_{\text{max}} \frac{\tau_e}{\tau_w \frac{1}{N}} (1 - e^{-t_{\text{tot}}/\tau_e}) e^{-(N-1)\delta t/\tau_e}
$$

(3.28)

where $t_{\text{tot}} = MN\delta t$ is the total writing time. The maximum diffraction efficiency that can be reached is the same as in the sequential recording scheme (Eq. (3.24)). The difference in refractive index modulation between the images is given by the
second exponential function of Eq. (3.28). If we require that all the holograms have a reciprocal relative difference smaller than $\epsilon_n$, the maximum recording time $\delta t$ will be given by

$$\delta t < \frac{\tau_+}{N} \ln (1 + \epsilon_n).$$  \hspace{1cm} (3.29)

It was demonstrated in Ref. [110] that for equivalent refractive index modulation the total writing time of sequential recording and incremental recording are equal. The total writing time will be however longer for the incremental recording method if we also consider the death time required to set the next image and reference beam in the setup. The method is however superior if we consider the homogeneity of the refractive index change, because the method is completely independent on the temporal dynamics of the recording material.

### 3.2.2 Influence of intrinsic light scattering in holographic medium

In the previous section it was shown that the refractive index modulation of the multiple stored holograms scales with $1/N$. For a large number of hologram we can therefore expect very low diffraction efficiency. If we consider Eq. (2.6-2.10) and (3.24) for the case of Bragg matched holograms we obtain a diffraction efficiency

$$\eta_N = \left( \frac{M/\#}{N} \right)^2 \approx \left( \frac{\pi l_2}{\lambda \cos \theta} \frac{\tau_c}{\tau_w} \frac{1}{N} \frac{1}{2} \eta^3 \tau_{eff}^2 E_{sc} \right)^2$$ \hspace{1cm} (3.30)

where the space charge field $E_{sc}$ is given by Eq. (2.23) and $M/\#$ is a figure of merit for the number of hologram which can be stored in a crystal with a certain thickness[112]. As an example, in a 1 cm thick LiNbO$_3$ crystal where 5000 angular multiplexed images are stored, a diffraction efficiency of $\eta_{5000} = 10^{-6}$ can be expected.

For such small diffraction efficiency the noise due to light scattered by intrinsic defects starts to be important. High requirements are then posed to crystal quality. The scattering noise can affect the image quality in two ways. First the reconstructed image beam with intensity $I_{rec}$ can be distorted by superposition of the light intensity $I_{scat,rec}$ scattered from the $I_{rec}$ itself. This kind of noise can be quantified by direct image transmission measurements through the crystal and is described by the signal to noise ratio $SNR_{trans} = I_{rec}/I_{scat,rec}$. We will see in next section that also the photorefractive self-focusing effect is responsible for distortion of the image beam. The second type of noise is generated by the light intensity $I_{scat,ref}$ scattered from the reference beam $I_{ref}$ in the direction of the reconstructed image which is described with the signal to noise ratio $SNR_{ref} = I_{ref}/I_{scat,ref}$. The total signal to noise $SNR_{tot}$, defined as $I_{rec}$ divided by the total scattered light in the direction of the reconstructed beam, can be calculated considering that $I_{rec} = \eta_N I_{ref}$. We obtain

$$SNR_{tot} = \left( \frac{1}{SNR} + \frac{1}{SNR_{trans}} + \left( \frac{N}{M/\#} \right)^2 \frac{1}{SNR_{ref}} \right)^{-1}.$$ \hspace{1cm} (3.31)
where in addition to scattering noise also cross-talk noise is considered. The noise generated from the reference beam is the most critical in the case that high storage capacity are targeted, because it scales with $N^2$. For $N = 1000$ for example a $SNR_{ref} > 10^8$ is required. As scattering by intrinsic defects is strongly dependent on the scattering angle, the angle between image beam and reference beam in the crystal is often chosen to be 90°. This can be achieved by entering the crystal from different faces. The method presents also other advantages which are described in section 3.2.4.

### 3.2.3 Impact of self-focusing on image quality

The photorefractive effect is responsible for fixing the light modulation produced by the interference pattern of the image and reference beam. Unfortunately the photorefractive effect will also interact with the light intensity modulation of the image beam itself. The inhomogeneous refractive index change produced by this interaction can cause a strong distortion of the image and the reference beam. The distortion crucially depends on the type of input image. The strongest distortion can be observed in the case of Fourier plane images (see Fig. 3.3). The reason for the strong distortion is connected with the spatial intensity distribution in the Fourier plane of an amplitude modulated image. An example of the Fourier transformed of a random bitmap is shown in Fig. 3.4. The small central peak for the spatial frequency 0 corresponds to the mean amplitude in the image plane. For the case of a binary amplitude modulation (the transmission of the “on” pixels is 1 and the transmission
Figure 3.4: Random bitmap image and its Fourier spectrum. The Fourier spectrum (which is obtained from the squared amplitude of the Fourier transformed) is shown for a cut along one axis in the spatial frequency plane.

of the “off” pixels is 0), the mean amplitude is non vanishing and generates the peak. Due to the self-defocusing effect (see also Appendix A.2) driven by the photogalvanic effect or an applied field, the peak will be broadened till it will start to overcome the amplitude of the lowest spatial frequencies which are located adjacent to the 0 spatial frequency point. This can be clearly observed in Fig. 3.3, where the contrast of the check pattern is slowly disappearing in time. The obvious way to eliminate the peak in the Fourier plane would be to use binary phase modulation (“on” pixel have a transmission of 1 and the “off” pixels a transmission -1). In this case, if the number of “on” and “off” pixels is equal, the mean amplitude in the image plane can be forced to zero. Phase modulation has however the drawback that the reconstructed image can not be detected directly on a CCD camera. In most Refs. reporting experiment with Fourier plane holograms another method is tacitly used: The Fourier plane is not imaged directly in the storage medium but slightly (some cm) before or after the crystal. The method is known as quasi Fourier plane holography.

For image plane holograms the situation is different. The intensity distribution inside the crystal corresponds to the image itself. There are therefore also small peaks present which represent the “on” pixels entoured by “off” pixels. The “on” pixels are self-focused. The global effect on image quality is however much smaller compared to the case of Fourier plane holograms.
In both cases the peaks appearing in the storage material are in the order of some 10 μm. This means that in order to build-up the self-focusing effect, charge carrier will have to move this distance. The build-up time required for this process is comparable with the build-up time of a holographic grating with grating spacing $\Lambda_g > 10 \, \mu m$ which was observed to be longer than the build-up time of the stored holograms in our investigated crystals.

When multiple holograms are written, the self-focusing effect is acting on all of the image waves. In the case of Fourier holograms the central peak of the 0 order will appear for every image at the same place, thus for every writing step the self focusing is enhanced. The situation is more complicated for image plane holograms where the peaks, which contains the actual information, are changing from page to page.

As previously mentioned the self-focusing effect is slower than the build-up time of the holograms. With this effect it is possible to write high quality holograms as reported in many publications also in material with strong photogalvanic effect. The problems however will show when the system is used as “write many” memory or with a periodical refreshing scheme. The image quality will then start to lower in a non reversible way.

A simple solution would be to use photorefractive materials with negligible photogalvanic effect. In this case however also the strong asymmetry between the erasure time $\tau_e$ and the writing time $\tau_w$ will be lost and 5-10 times smaller $M/#$ numbers (see Eq. (3.30)) must be taken into account.

### 3.2.4 Geometries for holographic storage

The hologram writing and read-out, the signal-to-noise ratio, and the storage density depend strongly on the possible range of the angle $2\theta$ between the image wave and the reference wave inside the material. In materials with high refractive index ($n > 2$) with normal cut not all angles $2\theta$ can be reached. There are basically three possible geometries which are represented by the small sketches in Fig. 2.5, where the photorefractive space charge field $E_{sc}$ is plotted as a function of the grating spacing $\Lambda_g$. The geometries are:

**Copropagating image and reference beam**

In this geometry the image and reference beam are entering the crystal form the same face. The grating spacing $\Lambda_g$ which can be produced in this way ranges from $\lambda/2$ till $\infty$. The method presents the advantage that it works also for thin storage media and only a short coherence length of the laser source is required. From the point of view of scattering, the method is not favorable because of the small angle between image and reference beam in the medium.

**90° geometry**

In the case of $2\theta = 90°$ the image and reference beam are entering the storage material at different faces. The obtained grating spacing is centered around
\[ \Lambda_g = \lambda/(\sqrt{2}n) \]. The method which was first proposed by Leith et. al\cite{113} is advantageous for several reasons: the effect of intrinsic scattering is minimized, the Bragg selectivity for angular multiplexing is greatest, and compact storage system can be implemented because the two beams are clearly separated. As a drawback the effective electro-optic geometry is in general smaller than in the copropagating geometry. This can be partially avoided using specially cut crystals which exhibit the polar axes at 45° with respect to the surface. In addition the effective electro-optic coefficient for light with polarization in the plane of incidence (p-polarized), which is generally larger than for s-polarization, can not be used because of the vanishing interference between the beams during hologram writing. For LiNbO₃ this means for example a \( M/\# \) number lowered by more than 3.5 times for not specially cut crystals and 3.3 for a 45° cut. Note that it is possible to write the holograms with s-polarization and read them out with p-polarization, in this case the read-out angles will have to be slightly corrected because of the anisotropy of the refractive index. For this geometry the \( M/\# \) number will result smaller by 1.7 times for not specially cut crystals under anisotropic Bragg-diffraction and lowered by 1.3 for a 45° cut.

**Counterpropagating image and reference beam**

The shortest grating spacing can be achieved using counterpropagating beams. The minimal grating spacing is given by \( \Lambda_g = \lambda/(2n) \). The method is mostly used combined with wavelength multiplexing, because of the optimal use of the tuning range of the laser.

### 3.2.5 Fixing and non destructive read-out techniques

Many applications based on photorefractive storage require that the stored holograms are not erased during read-out. The storage time in the dark which is mainly given by the conductivity in the dark is also finite. The most promising method to overcome these problems are thermal fixing and two-color holography. The thermal fixing procedure takes advantage of the mobile ions resident in the crystal as a consequence of the growth process, which produce a replica of a hologram or a series of holograms formed by trapped holes or electrons. This process can be described as follows \cite{114, 115, 116}: During or after hologram recording, the sample is heated to temperature in the 100°C - 200°C range. At that temperature the mobiles ions move to form another grating which is compensating the grating stored in the traps (fixing stage). On cooling at room temperature, a homogeneous illumination is used to erase the grating in the traps and brings out the replica (developing stage). The complementary grating remains and can not be optically erased at low temperatures. This technique was first demonstrated in LiNbO₃ \cite{117, 107, 118} and later in Bi₁₂SIO₂₀ \cite{119} and KNbO₃ \cite{120, 121}. With this process an erased time constant at room temperature of several years in LiNbO₃ and several months in KNbO₃ can be achieved even under illumination of the crystal.

Another method to which recently much attention has been devoted is the so called two-color or photon-gated holography\cite{26}. In this method the holograms are
written in the near infrared, where the photorefractive material does initially not show any sensitivity. A short illumination with the gating beam however will sensitized the material at the wavelength of the writing beams. The light induced sensitivity is produced by the population of a metastable level by the illumination with the gating beam at shorter wavelength (usually from the violet-blue region). The method requires a deep trapping level which is not accessible for the writing beams, where finally the holograms will be stored, and a higher empty trap level which can be filled by illumination with the gating beam. The method was demonstrated in LiNbO$_3$[28, 122, 123, 124], LiTaO$_3$[125], and La$_3$Ga$_5$SiO$_{14}$ [126].

3.3 Materials for holographic storage

Basically all photo-sensitive materials can be used as holographic storage material. The requirements coming from storage density and the storage time however reduce drastically the choice. We will focus here on two main groups namely ferroelectric oxides, and organic polymers.

The oxides were investigated intensively in the last 30 years, and the optimum growth condition for those materials are known, allowing to produce large high quality samples necessary for storage application. The following three materials were investigated: LiNbO$_3$, LiTaO$_3$, and KNbO$_3$. Each of these materials present different photorefractive properties which are mainly given by the dopant, dopant concentration, the reduction state of the crystal, and its chemical composition. This allows to tailor those materials for specific applications.

The polymers were investigated because they present very large change in refractive index, and can be produced in large quantities at a favorable price. The investigated polymers from DuPont[127] (HRF 600) are commercially available and produce the refractive index change by photopolymerization. The process is irreversible and therefore suited for read-only storage applications.

3.3.1 LiNbO$_3$

The most often used material for holographic storage is certainly lithium niobate (LiNbO$_3$), this is due to the long dark storage times ranging from some weeks till years, and the strong photogalvanic effect. The first property is central for long term storage systems, whereas the latter is important for an efficient recording of the hologram. In fact the photogalvanic effect is responsible for a strong asymmetry between the writing time $\tau_w$ and the erasure time $\tau_e$ where typical ratios for LiNbO$_3$ can be as high as $\tau_e/\tau_w = 5$. This means an enhancement of the diffraction efficiency $\eta_N$ by a factor of 25. However the photogalvanic effect is also responsible for the self-defocusing effect (see section 3.2.3). It takes usually more than 10 minutes to erase the self-defocusing lens by a homogeneous illumination of a Hg lamp in a congruent Fe (0.1% in the melt) doped crystal whereas other samples had to be heated in an oven at 180°C for one hour.

The dependence of the photorefractive sensitivity on the melt composition and the growth conditions were investigated in several 2 mm thick samples. The results are summarized in Tab. 3.1. The crystal were grown either with the double crucible
Table 3.1: Absorption $\alpha$ and sensitivities $S_1$ (2.11) and $S_2$ (2.12) for different LiNbO$_3$ crystals. The crystals were drawn from stoichiometric (stoic), congruent (cong), and Lithium rich melt, with different doping levels. Some of the crystal were oxidized (oxid.) after growth. The photorefractive experiments were performed with $\lambda = 488$ nm at a grating spacing $A_g = 1 \mu m$.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Composition</th>
<th>$\alpha$ (cm$^{-1}$)</th>
<th>$S_1$ (10$^{-12}$ m$^3$/J)</th>
<th>$S_2$ (10$^{-12}$ m$^2$/J)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CLN</td>
<td>cong, undoped</td>
<td>0.48</td>
<td>1.7</td>
<td>81</td>
</tr>
<tr>
<td>H100</td>
<td>cong, 50 ppm Fe</td>
<td>0.66</td>
<td>1.13</td>
<td>74</td>
</tr>
<tr>
<td>H200</td>
<td>cong, 440 ppm Fe</td>
<td>7.8</td>
<td>15.8</td>
<td>12300</td>
</tr>
<tr>
<td>H200A</td>
<td>cong, 440 ppm Fe, oxid.</td>
<td>1.6</td>
<td>7.0</td>
<td>1110</td>
</tr>
<tr>
<td>K150L</td>
<td>Li rich, 140 ppm Fe</td>
<td>1.44</td>
<td>15</td>
<td>2170</td>
</tr>
<tr>
<td>K150LA</td>
<td>Li rich, 140 ppm Fe, oxid.</td>
<td>0.20</td>
<td>3.2</td>
<td>64</td>
</tr>
<tr>
<td>K250</td>
<td>Li rich, 330 ppm Fe</td>
<td>18.2</td>
<td>11.8</td>
<td>21400</td>
</tr>
<tr>
<td>K250A</td>
<td>Li rich, 330 ppm Fe, oxid.</td>
<td>0.56</td>
<td>82</td>
<td>4560</td>
</tr>
<tr>
<td>KLN</td>
<td>stoic, undoped</td>
<td>0.20</td>
<td>3.9</td>
<td>80</td>
</tr>
<tr>
<td>KLNA</td>
<td>stoic, undoped, oxid.</td>
<td>0.21</td>
<td>2.5</td>
<td>54</td>
</tr>
<tr>
<td>SLN</td>
<td>stoic, undoped</td>
<td>0.34</td>
<td>4.6</td>
<td>156</td>
</tr>
<tr>
<td>SLNA</td>
<td>stoic, undoped, oxid.</td>
<td>0.13</td>
<td>3.8</td>
<td>50</td>
</tr>
<tr>
<td>RHKL1</td>
<td>stoic, Rh doped</td>
<td>0.67</td>
<td>2.6</td>
<td>182</td>
</tr>
<tr>
<td>FEK1</td>
<td>stoic, 270 ppm Fe$^a$</td>
<td>13.2</td>
<td>4.0</td>
<td>5230</td>
</tr>
<tr>
<td>FEK2A</td>
<td>stoic, 270 ppm Fe$^a$, oxid.</td>
<td>2.75</td>
<td>32</td>
<td>8870</td>
</tr>
</tbody>
</table>

$^a$ special composition of the melt with 6% potassium

method or from potassium rich melts at NIRIM in Japan, starting from three different ratio of Li and Nb giving congruent crystals, stoichiometric, and Li rich crystal. In stoichiometric crystal the ratio between Nb and Li in the crystal is exactly equal to the theoretical crystal structure, whereas in the congruent case Nb atoms are in abundance. Stoichiometric crystals are expected to have less intrinsic lattice defects and present an absorption spectrum were the band gap is shifted to higher photon energies $h\nu$. All the samples were intentionally doped with Fe and some with Rh.

The photorefractive properties of the samples were characterized by absorption and diffraction efficiency measurements. The absorption is mainly given by the amount of Fe in the melt and the oxidation state of the crystal. The oxidation state can be changed by heating the crystal till 600°C in a reducing or oxidizing atmosphere. The highest sensitivities were measured in Li rich 330 ppm Fe doped samples. The maximum sensitivity $S_1$ (see Eq. (2.11)) was found in the oxidized sample whereas $S_2$ was maximum in the reduced sample. This suggests that the optimal amount of iron in the melt is around 300 ppm. Comparing the nominally undoped samples we note that the stoichiometric samples show a higher sensitivity.

From measurements with a scattering tomograph it was found that the scattering is lower in stoichiometric material and can therefore be considered superior to congruent compositions for holographic storage.
3.3. Materials for holographic storage

3.3.2 LiTaO$_3$

The second investigated material which is isomorphous to LiNbO$_3$ is lithium tantalate (LiTaO$_3$). The photorefractive properties of LiTaO$_3$ are very similar to LiNbO$_3$. Also in this crystal the build-up of the holographic grating is driven by the photogalvanic effect. We observed a strong lensing effect after longer illumination, ending with a self-induced lens that could only be erased by warming up the sample (a congruent crystal doped with 300 ppm Fe in the melt) for 1 hour at 180°C. In this material, due to the high refractive index change, we also observed a self enhancing effect of the object beam during the read out with a strong reference beam. Compared to LiNbO$_3$, stoichiometric LiTaO$_3$ show a slightly lower photogalvanic effect, and higher sensitivities at UV wavelength.

3.3.3 KNbO$_3$

The last investigated material is potassium niobate (KNbO$_3$). All the samples were grown in the nonlinear optics laboratory at ETH using top seed solution growth. The crystals were grown in air and are therefore not reduced. KNbO$_3$ presents a much shorter storage time in the dark compared to LiNbO$_3$ or LiTaO$_3$. The decay times in the dark which can vary from few seconds to several days are reported for different dopants in Tab. 3.2. Longer storage times can be achieved using thermal fixing methods[120]. The main advantage of KNbO$_3$ compared to LiNbO$_3$ and LiTaO$_3$ are the photorefractive sensitivities, which are from 2 to 3 orders of magnitude higher (see Tab. 3.2). Therefore much shorter writing times can be achieved. Especially Ce doped KNbO$_3$ presents a very high ratio ($\tau_{\text{dark}}/\tau_{\text{die}} = 3 \cdot 10^6$) between dark storage time $\tau_{\text{dark}}$ and the hologram build-up time $\tau_{\text{die}}$ at an intensity of 1 W/cm$^2$.

High sensitivities however are not always an advantage in photorefractive storage. In fact the crystal is also erased by the illumination used during the read-out process.

<table>
<thead>
<tr>
<th>Crystal Nr.</th>
<th>Composition</th>
<th>$\tau_{\text{dark}}$ (h)</th>
<th>$S_2$ ($10^{-12}$ m$^2$/J)</th>
</tr>
</thead>
<tbody>
<tr>
<td>186</td>
<td>2000 ppm Ni doped</td>
<td>71</td>
<td>240</td>
</tr>
<tr>
<td>176</td>
<td>3000 ppm Ni doped</td>
<td>26</td>
<td>2000</td>
</tr>
<tr>
<td>254</td>
<td>500 ppm Rh doped</td>
<td>5.3</td>
<td>2800</td>
</tr>
<tr>
<td>250</td>
<td>1500 ppm Rh doped</td>
<td>$6.7 \cdot 10^{-2}$</td>
<td>1500</td>
</tr>
<tr>
<td>249</td>
<td>3000 ppm Rh doped</td>
<td>1.4</td>
<td>6400</td>
</tr>
<tr>
<td>279</td>
<td>1500 ppm Rh/Mn doped</td>
<td>2.9</td>
<td>1600</td>
</tr>
<tr>
<td>208</td>
<td>1000 ppm Mn doped</td>
<td>$5 \cdot 10^{-2}$</td>
<td>3100</td>
</tr>
<tr>
<td>217</td>
<td>1000 ppm Mn doped</td>
<td>103</td>
<td>18</td>
</tr>
<tr>
<td>222</td>
<td>1 ppm Ce doped</td>
<td>31</td>
<td>78000</td>
</tr>
</tbody>
</table>
The intensity of the readout beam has to be kept as high as required by the detector, and considering that the diffraction efficiency will decrease with the number of stored holograms squared, this represents a serious limitation for the maximal number of holograms that can be stored. The limitation can be somehow alleviated using thicker samples, this however to the expense of the storage density.

The main difference between KNbO₃ and LiNbO₃ is that the photogalvanic effect is much smaller (and usually negligible) in KNbO₃. During our experiments we did not observe any distortion produced by the self-induced lensing, and the crystal could be erased in a short time by an homogeneous illumination. As drawback KNbO₃ lacks the asymmetry between writing time of the hologram and the erasing time by homogeneous illumination. Tomographic scattering measurements (presented in Fig. 3.5) were performed at NIRIM in Japan. The scattering properties strongly depend on the growth condition and dopant. The measurements have demonstrated that Mn doped KNbO₃ shows very low scattering compared to other KNbO₃, LiNbO₃, and LiTaO₃ crystals.

In copper doped KNbO₃ crystals we observed a self fixing effect leaving the image stored in the crystal for several days. This is probably related to ionic compensations[121] which occurs on a longer time scale already at room temperature. The hologram is then stored by the inhomogeneous ionic concentration and can not be erased anymore by light illumination. The effect could be used for the implementation of a photorefractive read only memories.

![Figure 3.5: Tomographic scattering measurement of a Mn doped KNbO₃ crystal. The image is obtained by detecting the light scattered perpendicular to a strong focused laser beam. A two-dimensional image is obtained by scanning the crystal perpendicularly to the observation direction.](image-url)
3.3.4 Photo-polymers

Photo-polymer materials are attractive candidates for write-once-read-many-times (WORM) data storage applications because they can be designed to have large refractive index contrast and high photosensitivity, record permanent holograms, and be easily processed. Different implementations of holographic disks based on photo-polymer have been proposed. There are many possible mechanisms for fixing permanent or reversible refractive index gratings in organic materials as for example photodimerization, photoisomerization, photoinduced intramolecular structural changes, photo-oxidation, photo-bonding in PMMA, and photo-polymerization. Drawbacks of photo-polymers are the low optical quality, the limited thickness, and the shrinkage during recording.

![Figure 3.6](image.png)

**Figure 3.6:** Recording dynamic and angular dependence of the diffraction efficiency $\eta$ for a photo-polymer (DuPont HRF 600). The hologram was written by an intensity of $I_0 = 6 \text{ mW/cm}^2$ and a grating spacing of $\Lambda_g = 0.5 \mu\text{m}$. The polymer has a thickness of 20 $\mu\text{m}$.

In this work we have investigated commercially available polymers (HR 600 from DuPont). The polymer had a thickness of 20 $\mu\text{m}$ and were laminated on glass substrates. The refractive index change in these polymers is produced by photo-polymerization of free monomers. The process is irreversible. After exposition of the interference pattern the hologram can be indefinitely fixed by a short exposition with a Hg lamp. The refractive index contrast can be further increased by baking the polymer at 100°C. Despite the small thickness of 20 $\mu\text{m}$, a diffraction efficiency as high as 95% was observed. The dynamics of the recording process is shown in Fig. 3.6. The holographic grating was written by the interference of two Ar-ion laser beam at 488 nm and monitored in real time by a red beam at 633 nm. The light intensity was 6 mW/cm$^2$. Note that the recording process can not be made faster by simple increasing the intensity like in conventional photorefractive effect, because the build-up time is mainly given by the diffusion time of the free monomers. In Fig. 3.6 also the angular dependence of the grating diffraction is presented. The angular selectivity for the grating is around 4°. Much thicker polymers are therefore required.
to store multiple holograms. Important improvements were achieved recently by Steckman et. al.[132], and Dhar et. al.[134, 133] which showed high quality 1 mm thick materials with large effects and photosensitivity. In their polymers Dhar et. al. also demonstrated the highest ever reported $M/#$ of 42.

### 3.4 Set-up for holographic storage

In this section we present a set-up based on photorefractive holographic storage capable to produce image sequences with a high frame rate. The system is used as image source for a joint Fourier transform correlator described in Chapter 5. After some little modifications the system can also be used as Vander Lugt type correlator, this is presented in Chapter 4. A schematic representation of the set-up is given in Fig. 3.7 a). As light source a Q-switch diode pumped Nd:YAG at 532 nm wavelength is used. The laser delivers a maximum pulse energy of 0.5 mJ with a pulse length of 40 ns and can also operate in continuous wave producing up to 0.6 W of green light. In the experiments the Q-switch of the laser is triggered externally by a digital word generator. The beam from the laser is then expanded by a pair of lenses and the polarization adjusted by a Fresnel rhombus (see Fig. 3.7 a)). After expansion the beam passes through a Polarizing Beam Splitter (PBS) where the beam is split in two beams, the image and the reference beam. The splitting ratio of the beams can be adjusted using the previously mentioned Fresnel rhombus. The reference beam is transmitted by the PBS, has horizontal polarization and ends on a rotating mirror.

The mirror (a M3 galvanic scanner from General Scanning) is producing the different reference waves, whereas the following telescope system images the beam always at the same spot on the photorefractive crystal.

The image beam is deflected from the PBS and is vertical polarized. It is reflected by a ferroelectric liquid crystal spatial light modulator with $256 \times 256$ pixels (from Displaytech). Ferroelectric modulators have only two possible states referred as “on” and “off” state in the following. The modulator can be used as amplitude or phase modulator. In the configuration as amplitude modulator, the fast axis of the liquid crystal is rotated $45^\circ$ from the vertical. The polarization of the “on” pixels in the incoming beam are turned to horizontal whereas the polarization of the “off” pixels is left unchanged. The beam reflected from the modulator goes back to the PBS. The light of the “on” pixels which is horizontally polarized is transmitted, whereas the light of the “off” pixels is deflected back to the laser. Phase modulation can be achieved by rotating the modulator till the angle between the fast axis and the vertical is $22.5^\circ$.

The image modulated on the beam is then imaged on the storage crystal. Two different lens systems were used to produce image plane and Fourier plane holograms. The lens system was designed with an optical design program. Achromatic doublets lenses which exhibit small spherical aberrations were used. For Fourier plane holograms three lenses (lens 9 with focal length $f = 100$ mm, lens 1 with $f = 40$ mm, and lens 2 with $f = 60$ mm) where used. To obtain image plane holograms the lenses 1 and 2 where replaced by a single lens with $f = 100$ mm. As mentioned in section 3.2.3 the self-focusing effect in LiNbO$_3$ and LiTaO$_3$ crystals is very strong,
3.4. Set-up for holographic storage

Figure 3.7: Set-up for pulsed holographic memory with high read-out frame rate (Fourier plane geometry). See text for description. a) Schematic representation. b) Photograph of the set-up.

for this reason in the case of amplitude modulated image the Fourier plane of the
image was set 2 cm behind the crystal. This offers the additional advantage that the light coming from scattering inside the crystal, can be partially blocked using a diaphragm placed in the Fourier plane. In this geometry the self-phase modulation was reduced, but still observed.

As the coherence length of the used laser is rather short (5-11 mm depending on the pulse repetition rate), the optical path within object and reference beams must be carefully adjusted. This was verified optimizing the two beam coupling gain of a KNbO₃ put at the same place of the storage crystal. The holograms were written using copropagating geometry and 90° geometry. Highest image quality is obtained with the 90° geometry, but due to the small coherence length of the laser and the less favorable electro-optic configuration, the diffraction efficiency in this geometry was one order of magnitude smaller. Most experiments were therefore performed in the copropagating geometry with an external angle $\theta_{\text{ref}} = 45°$ for which a proper overlap of the image beam and reference beam could be achieved.

The angular separation between adjacent reference beams was $\Delta \theta_{\text{ref}} = 0.03°$ giving a total deflection of 3° for 100 stored images.

A picture of the set-up which has a size of about 50×50 cm is shown in Fig. 3.7 b). In the next sections the writing and read-out procedure are described in detail and finally some experimental results are shown.

### 3.4.1 Writing procedure

In the presented set-up the holograms can be written either by pulsed or continuous wave (CW) illumination. In both cases the holograms are written using the incremental recording scheme (see section 3.2.1) by multiple cycles. For each position of the rotating mirror a different image is displayed on the spatial light modulator. The mirror is moving with a periodic oscillation driven by the digital word generator or a computer, which is also responsible for displaying the next image on the SLM and trigger the laser pulse (in pulsed mode) or the mechanical shutter (in CW mode).

Writing the holograms with pulses presents several advantages: The writing pulse is very short and the cycling time in the incremental recording scheme is then mainly given by the setting time of the spatial light modulator and the rotating mirror. Another advantage is that the laser light is used more efficiently compared to a CW laser. The population inversion in the active laser material can be produced during the setting times of the mirror and modulator, and moderate energy will be ready for the next image to write. The final advantage of pulses is that there is no need for an external shutter to cut the light during mirror and modulator setting.

In our system the writing repetition rate in pulsed operation was limited by the ferroelectric SLM, which can operate at a speed of 1200 images/s\(^4\). In this way 100 images can be written, with 50 cycles (which are enough to produce homogeneous diffraction efficiency) in less than 10 s or 100 ms for each written image. The same result can be achieved in CW illumination but would require 500 s in our set-up.

\(^4\)The modulator can work at 2400 images/s, but the liquid crystal requires to be operated with the image and the reverse image exposed for the same length of time.
3.4. Set-up for holographic storage

3.4.2 Image read-out

The image are read out by illuminating the storage crystal with the reference beam only. The corresponding image beam is then reconstructed. By illuminating the crystal with CW illumination for a fixed position of the rotating mirror it is possible to detect the image on a conventional CCD camera. An example of input and read-out images for 100 images plane holograms stored in a LiNbO$_3$ crystal is shown in Fig. 3.8. The crystal was doped with 150 ppm Fe and has a size $10 \times 10 \times 10$ mm$^3$. Unfortunately it was not possible to achieve pixel matching between the pixels of the SLM and CCD camera because the pixel form of the modulator is quadratic whereas our camera has rectangular pixels. Due to the higher resolution of the camera ($748 \times 532$) most pixels in Fig. 3.8 can be correctly recognized. In this crystal a signal to noise ratio $SNR_{tot} = 46$ was observed. Measuring the total diffracted light power the number $M/\# = 0.3$ (see Eq. (3.30) for definition) was obtained. Illuminating the crystal with the reference beam for longer times, a self-enhancing of the diffraction efficiency and a subsequent image degradation probably due to self-focusing was observed. The experiments were repeated with other differently doped LiNbO$_3$ and LiTaO$_3$ crystals, giving similar results. 100 holograms were also stored in different Cu, Mn, and Ce doped KNbO$_3$ crystal. The image quality is comparable to the one obtained in LiNbO$_3$. The self-focusing effect was mostly negligible except
for the case of Fourier plane hologram. The achieved diffraction was smaller, we obtained a number $M/\# = 0.08$. This is due to the lack of the photogalvanic effect. As the sensitivity in KNbO$_3$ is much higher, the image can be stored much faster. However they are also erased faster during the read-out.

The image read-out rate under pulsed illumination is not limited anymore by the SLM as during the writing procedure, and therefore much higher frame rates can be achieved. With our set-up we were able to demonstrate a read out frame rate of 5000 images/s. This was limited by the reduced linearity of the rotating mirror at higher angular velocities. In the next section a method is presented to overcome the problem and achieve much higher repetition rates.

### 3.4.3 Fast image sequence generation

Fast image sequences are obtained synchronizing the position of the rotating mirror with the read-out pulse. At highest read-out rate the angular velocity of the rotating mirror is slightly changing (in a reproducible way) during rotation. The read-out pulses fired at constant rate are not fulfilling the Bragg condition for all holograms anymore.

The problem can be solved in two ways: changing the delays between the laser pulses or change the writing angle of the reference beams. We have implemented the second possibility. The procedure works as follows: The same image is written for all reference beams at the desired read-out frame rate. The diffraction efficiency as function of the rotating mirror angle is then measured and the Bragg matched angle is interpolated. The memory can then be rewritten with the newly found angles at low repetition rates and finally read-out at the desired high repetition rate. With this method we were able to produce a frame rate of 50'000 Frames/s. For such high frame rates our computer (PowerPC 300 MHz) was not able to produce a reliable synchronization anymore and was replaced by a digital word generator (Interface Technology RS670). The read-out frame rate is now limited by 50'000 Frames/s because of the limited repetition rate of our laser. Preliminary experiments have shown that the rotating mirror would be capable to produce frame rates reaching up to 200'000 Frames/s.

Due to the lack of a fast enough CCD detector, the image sequence was detected by a single photomultiplier (see Fig. 3.9). Using a stroboscopic technique it was possible to detect the single images. This was achieved by reading out the holograms with the high frame rate method but triggering the laser only for one particular image.

50'000 Frames/s is the fastest ever reported frame rate for a system based on rotating mirror. Other holographic memory systems with rapid access based on acusto-optical deflectors have been demonstrated in Refs. [91, 135]. In these works an access time of 15 $\mu$s, which is mainly given by the setting time of the acusto-optic deflector is given. This is slightly faster than the 20 $\mu$s demonstrated in this work and presents the additional advantage that the images can be accessed in a random way. An important point which has however also to be considered is the

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5For a slower read-out rate of 800 frames/s it is possible to detected the images directly using a high speed camera ($128 \times 128$ pixels) from Dalsa.
set-up for holographic storage

Figure 3.9: Intensity of 100 read-out images detected on a photomultiplier. Each peak corresponds to a different image. The images were written in pulsed mode using an incremental writing scheme with 200 iteration. The read out time of 2 ms corresponds to a frame rate of 5000 Frame/s.

detection time of the images. The diffraction efficiency $\eta_N$ for a large number $N$ of stored images will be very low, and a small light intensity will have to be detected on the CCD camera. Also a highly sensitive camera will require at least some ms of integration time to obtain an image with a reasonable signal to noise ratio. In our approach based on a pulsed laser the detection can be made much faster because of the large number of photons contained in a pulse. The detection is also less sensitive to the thermal noise of the CCD detector if a gated detection method is used.

The total throughput produced by our system given by the product of the total number of pixels and frame rate is 3.3 Gbit/s. The data rate can be easily increased using spatial light modulators with higher resolution and a laser with higher repetition rate. For an SLM with a resolution of $1280 \times 1024$ pixels and a laser with 200 KHz repetition rate which will be commercially available within one year, the throughput would scale up to 260 Gbit/s.

No electrically addressable spatial light modulator is capable of producing such a high data rate. This means that holographic storage is well suited to be used as fast SLM for application where the images are already known in advance, as for example in correlation experiments where an external input image has to be compared with a library of images.

$\textsuperscript{4}$The camera can be additionally cooled to reduce the effect of thermal charge excitation in the CCD cells.
Chapter 4

Vander Lugt type correlator

In the Frequency Plane Correlator or Vander Lugt type correlator (VTC), correlation between the two input beams is achieved in two steps. First a matched filter is holographically recorded by fixing the interference pattern produced by the first input beam and a plane wave. In a second step the hologram is read out by the second input beam. If the input beams are similar, the plane wave is reconstructed. The method is similar to a holographic memory which is read out with the image beam instead of the reference beam. In fact multiple matching filters can be stored by the same multiplexing method as presented in section 3.1.2.

In the next section a theory for the Vander Lugt correlator, first for thin media and later for multiple hologram in thick media is presented. Section 4.2 contains experimental results obtained adapting the set-up for holographic storage described in Chapter 3. The chapter ends with a section presenting some examples of applications.

4.1 Principle of Vander Lugt type correlator

In the correlator proposed by Vander Lugt[17] the input pattern modulated on a collimated beam by the transmittance $s(x_0, y_0)$ is compared with a fixed hologram. The optical generation of the hologram (or matched filter) that contains the frequency spectrum of the object pattern $s_1(x_0, z_0)$ is shown in Fig. 4.1. The input device is located in the front focal plane of a lens and a holographic recording media (for example a photographic film) is placed in the back focal plane. Further these two devices are located perpendicular to the optical axis so that the Fourier spectrum is in focus across the plane of the holographic medium.

The amplitude and phase $S_1(x, y)$ of the spatial Fourier transformed of $s_1(x_0, z_0)$ are recorded by interference with a plane reference wave $R(x, y) = R_0 e^{ik(-y \sin \theta_{ref} + z \cos \theta_{ref})}$, where $\theta_{ref}$ is the angle between signal and reference beam and determine the grating spacing $\Lambda_y$ of the interference fringes. If aberrations are neglected, the observed intensity pattern is given by

$$I(x, y) = |R(x, y) + S_1(x, y)|^2$$
$$= |R_0|^2 + |S_1(x, y)|^2 + R^*(x, y) S_1(x, y) + R(x, y) S_1^*(x, y) \quad (4.1)$$
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**Figure 4.1:** Realization of a matched filter for Vander Lugt correlator, where \( f \) is the focal length of the lens.

The intensity pattern \( I(x, y) \) is fixed in the holographic medium either as phase or amplitude hologram. When a transparency \( s(x_0, z_0) \) is present at the input plane and illuminated with a collimated beam, the intensity distribution at the output plane (see Fig. 4.2) can then be divided into three regions. The first region which appears on the optical axis is the image of the transparency \( s(x_0, y_0) \) transmitted through the hologram. The observed intensity distribution at the second region centered at \( (x_c = 0, y_c = c) \) with \( c = f \tan \theta_{ref} \) is proportional to the absolute square of the amplitude convolution function of the input and the stored scene:

\[
I_{conv}(x_c, y_c - c) \propto |\mathcal{F}\{s(x, y) \cdot S_1(x, y)\}|^2 \\
\propto |s(x_0, y_0) \oplus s_1(x_0, y_0)|^2 \\
\propto \left| \int \int_{aperture} s(x_0, y_0) s_1(x_c - x_0, y_c - y_0) \, dx_0 \, dy_0 \right|^2 \tag{4.2}
\]

where \( \oplus \) denotes the convolution. The third region appears with its center off the optical axis by an amount \( (x_c = 0, y_c = -c) \). The intensity distribution is
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proportional to the absolute square of the amplitude correlation function of the input object \( s(x_0, y_0) \) and the stored object \( s_1(x_0, y_0) \):

\[
I_{\text{corr}}(x_c, y_c + c) \propto |\mathcal{F}\{S_1^*(x, y) \cdot (x, y)\}|^2 \\
\propto |s_1(x_0, y_0) \otimes s(x_0, y_0)|^2 \\
\propto \left| \int_{\text{aperture}} s_1^*(x_0 - x_c, y_0 - y_c) s(x_0, y_0) \, dx_0 \, dy_0 \right|^2 \tag{4.3}
\]

The cross-correlation function \( I_{\text{corr}}(x_c, y_c + c) \) is peaked at the coordinate \((x_c, y_c - c)\) where the two images are most similar. This property can be used to identify the position of a smaller image inside a larger image, or to find out if a certain object is present in a scene. If the input object \( s(x_0, y_0) \) is similar to one of the objects contained in the reference set \( s_1(x_0, y_0) \), intensity peaks will occur at the corresponding places, as sketched in Fig. 4.3. The height of the peak is a measure of the degree of similarity of two objects. The object can thus be identified by detecting the position and the relative intensities of the correlation peaks in the output plane.

![Figure 4.3: Input images and output plane of a 4 channel correlator. The reference set \( s_1(x_0, y_0) \) contains 4 objects which have to be compared with the object \( s(x_0, y_0) \).](image)
4.1.1 Vander Lugt correlator using multiple volume Holograms

Thick holographic material like photorefractive crystal can be used to implement a Vander Lugt correlator. The advantage is that multiple matching filters (1000 or more) can be used simultaneously, thus allowing to perform \( N \) correlations (where \( N \) is the number of stored images) during one single read-out step. The price of this parallelism is the lost of the shift invariance in one direction, as we will see later.

A detailed analysis of the volume holographic Vander Lugt correlator, which considers also the cross-talk noise was presented by Gu et. al.[136, 137]. In this work we consider a similar approach in paraxial approximation. The advantage of our approach is that a simple analytic expression for the shift invariance can be derived. Referring to Fig. 4.4 consider an optical correlator system in which multiple holograms are stored in a thick recording medium near the center of the Fourier-transform plane of the object beam. The writing method is the same as in a holographic storage system based on Fourier plane image (see section 3.2). Assume that \( N \) holograms are stored. The refractive index change produced by the photorefractive effect after the hologram recording can be approximated by

\[
\Delta n \propto \sum_{m=1}^{N} R_m S_m^* + c.c.,
\]

where \( R_m \) and \( S_m \) are the amplitude of the reference beams and the object beams inside the holographic medium, respectively.

During the correlation operation an input object wave \( s(x_0, y_0) \) is Fourier transformed and incident upon the volume holograms. The inhomogeneous refractive index (Eq. (4.4)) will cause diffraction of the incident wave. According to scalar

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*The refractive index change produced by the photorefractive effect is in general more complicated and depends on the intensity ratios between the reference beam and the image beam. The relevant term producing the correlation peak is however given by Eq. (4.4)*
4.1. Principle of Vander Lugt type correlator

Diffraction theory\[138\] the amplitude of the diffracted electric field can be written as

\[ E(r) \propto \int dr'e^{-ik_d r'S(r')\Delta n(r')}, \tag{4.5} \]

where \( k_d \) is the wavevector of the diffracted plane wave and \( S \) is the incident object wave. The integration in the position vector \( r' \) is evaluated over the whole volume of the hologram. The diffracted light consists of both the desired correlation pattern and noise due to cross correlation.

In the configuration shown in Fig. 4.4 the reference wave

\[ R_m = R_0 e^{ik_m \cdot r} \tag{4.6} \]

is a plane wave corresponding to the \( m \)th point distributed along the \( y_m \)-axis in the reference plane. According to Fourier optics in paraxial approximation the components \((k_{mx}, k_{my}, k_{mz})\) of the wavevector of a plane wave inside the medium, resulting from a point \((x_m = 0, y_m)\) at front focal plane of the lens 1 (see Fig. 4.4) are

\[
\begin{align*}
    k_{mx} &= 0 \\
    k_{my} &= \frac{2\pi y_m + h}{\lambda} \\
    k_{mz} &= nk_0 \sqrt{1 - \left(\frac{k_{my}}{nk_0}\right)^2} \\
    &\approx \frac{2\pi n}{\lambda} - \frac{\pi}{n\lambda} \left(\frac{y_m + h}{f}\right)^2,
\end{align*}
\tag{4.7}\]

where \( k_0 \) is defined as \( k_0 = 2\pi/\lambda \), and \( f \) is the focal length of all lenses.

The \( m \)th object beam \( S_m \) is the Fourier transformed near the storage medium of the \( m \)th object pattern given by \( s_m \) in the object plane. Using standard Fourier-optics analysis\[139\], one obtains

\[
\begin{align*}
    S_m &= \frac{e^{2\pi i f}}{i\lambda f} e^{i k_{mz} z} \int dx_0 dy_0 s_m(x_0, y_0) e^{-i \frac{2\pi}{\lambda f}(xx_0 + yy_0 - h)} e^{-i \frac{2\pi}{\lambda f}(y_0^2 + h^2)} \Delta z' \\
    \Delta z' &= n \Delta z - \frac{f_e}{2}(n - 1),
\end{align*}
\tag{4.8}\]

where \( \Delta z \) is the distance difference between the center of the medium and the position of the Fourier plane of lens 1.

Eq. (4.8) takes into account the fact that inside the holographic medium with finite thickness, the optical field distribution varies as the object wave propagates through the recording medium. Similarly, the incident wave \( S(r) \) during correlation can be obtained by the replacement of \( s_m(x_0, y_0) \) in Eq. (4.8) by the input pattern \( s(x_0, y_0) \).

The diffracted wavevector \( k_d \) in Eq. (4.5) can be related to the coordinates of the output correlation plane \((x_c, y_c)\). A plane wave at the front focal plane of lens 2
(see Fig. 4.4) is converted to a point at the rear focal plane of lens 2. The relation between the wavevector $k_d = (k_{dx}, k_{dy}, k_{dz})$ and the coordinates of the points is given by

$$
\begin{align*}
    k_{dx} &= \frac{2\pi x_c}{\lambda f} \\
    k_{dy} &= \frac{2\pi y_c - h}{\lambda f} \\
    k_{dz} &= \frac{2\pi n}{\lambda} \left( \frac{(y_c - x)^2 + x_c^2}{f^2} \right)
\end{align*}$$

(4.9)

Using relation (4.4) and Eqs. (4.6)-(4.9), we can evaluate the Eq. (4.7). The result is

$$
I_{corr}(x_c, y_c) \propto \left| \sum_{m=0}^{N} \int dx_0 dy_0 dx_1 dy_1 s(x_0, y_0) s^*_m(x_1, y_1)e^{-i\Delta\phi_0 \frac{x_0^2 + y_0^2 - (y_m + h)^2 + (y_c - y_0 - y_m)^2}{f^2}} \right|^2
$$

(4.10)

where $l_x, l_y, l_z$ are the dimensions of the holographic medium in the $x, y, z$ directions respectively and $x_1, y_1$ are variables introduced by the integration. Expression (4.10) also give the Bragg-matching condition in volume holography. The $\text{sinc}(x) = \sin(\pi x)/\pi x$ functions indicate that when a specific grating component is Bragg matched (argument of the sine functions is equal to 0), the diffraction efficiency is maximum. Otherwise the intensity of the diffracted light decreases as a $\text{sinc}^2(x)$ function with respect to the momentum mismatch.

The first two sinc functions relate the shift in the $x_0$ and $y_0$ direction between the two input patterns in the object plane with the position of the correlation peak in the correlation plane $x_c$ and $y_c$, respectively. These sinc functions can be approximated by $\delta$ functions when the extension of the Fourier transformed $S_m$ in the crystal plane $(x, y, z)$ is smaller than the crystal widths $l_x$ and $l_y$. Evaluating Eq. (4.7) and (4.9) we obtain the following relations between the input coordinates and the coordinates in the output plane

$$
\begin{align*}
    x_c &= x_1 - x_0 \\
    y_c &= y_1 - y_0 - y_m
\end{align*}$$

(4.11)

(4.12)

Taking into account Eqs. (4.11)-(4.12) and after some simple algebraic transformations, the intensity in the output plane can be written as

$$
I_{corr}(x_c, y_c) \propto \left| \int s(x_0, y_0) s^*_m(x_0 + x_c, y_0 + y_c + y_m) \right|^2
$$

(4.13)
4.1. Principle of Vander Lugt type correlator

where $\Delta z'$ was set to 0. This form is already very close to the correlation intensity described by Eq. (4.3). The sinc function in Eq. (4.13) takes into account the amplitude change of the image along the propagation direction in the thick holographic medium. The effect limits the maximum allowed shift between the input patterns $\Delta x_{\text{max}}$ and $\Delta y_{\text{max}}$ in the $x_0$ and $y_0$ direction, respectively. Considering that the correlation peak in the output plane is centered at $(x_c, y_c) = (0, -y_m)$, and introducing the image extension $D_x$ and $D_y$ of the input patterns in the input plane in the $x_0$ and $y_0$ direction respectively, we obtain the following condition for the maximum crystal thickness

$$l_z < \frac{0.9n\lambda f^2}{D_x \Delta x_{\text{max}} + (D_y + 2h)\Delta y_{\text{max}}},$$

(4.14)

where the sinc function of Eq. (4.13) was evaluated at half intensity ($\text{sinc}^2(x/2) = 1/2$).

The maximal crystal thickness $l_z$ is therefore determined by the required shift invariance $\Delta x_{\text{max}}, \Delta y_{\text{max}}$ in both directions. The maximal allowed shift between the input patterns, for a given crystal thickness $l_z$, can be evaluated in both directions independently. For the $x_0$ direction we obtain

$$\Delta x_{\text{max}} < \frac{0.9n\lambda f^2}{D_x l_z},$$

(4.15)

For a system with a focal length $f = 100 \text{ mm}$, $l_y = 10 \text{ mm}$, $D_x = 5 \text{ mm}$, and the refractive index $n = 2.1$ the maximum shift between the input images is $\Delta x_{\text{max}} \approx 0.26 \text{ mm}$.

For the maximal allowed shift in the $y_0$ direction we obtain

$$\Delta y_{\text{max}} < \frac{0.9n\lambda f}{\theta_{\text{max}} l_z},$$

(4.16)

where $\theta_{\text{max}}$ is the maximum angle between the reference $R$ and the signal beam $S_m$. For the same parameter as above, and a maximal angle $\theta_{\text{max}} = 45^\circ$ we obtain a maximum shift between the input patterns of $\Delta y_{\text{max}} \approx 13 \mu\text{m}$. The shift invariance in this direction is therefore strongly reduced, and obviously connected with the maximal number of holograms that can be stored (see Eq. (3.9)). The effect of moving the Fourier plane outside the crystal (quasi Fourier plane holograms) is explicitly considered by the displacement $\Delta z$. The displacement is related to the shift invariance by the condition $\Delta z' < \lambda(f/2)^2/(D_x \Delta x_{\text{max}} + (D_y + 2h)\Delta y_{\text{max}})$, and therefore smaller than the maximal allowed hologram thickness $l_z$.

The noise generated by the simultaneous presence of multiple holograms can be calculated in a similar way as cross-talk noise in holographic storage (see section 3.1.3), and was calculated in Ref. [136].

As conclusion we have shown that while for thick volume holograms the Bragg condition strongly reduces the shift invariance in the $y_0$-direction, the system retains partially its shift invariance in the direction $x_0$ which is perpendicular to the plane of incidence.

Although the shift invariance is partially lost in one dimension, the utilization of volume holograms in the Vander Lugt correlator is favorable because multiple
matched filters can be stored. The correlation rate resulting from the number of correlations which are processed in parallel can be far above the correlation rate of any electronic system developed before 2000.

4.2 Correlation experiments in LiNbO₃ and LiTaO₃

A Vander Lugt type correlator was implemented based on the set-up for holographic storage (see Fig. 3.7). The set-up can work as correlator if the holograms are read out with the image beam instead of the reference beam. The reconstructed reference beams (plane wave) are Fourier transformed by the lens 8 in Fig. 3.7 and detected on CCD-camera 2, which corresponds to the output plane of the correlator in Fig. 1.1. The input pattern can be encoded on the image beam by amplitude or phase modulation. In amplitude modulation, because of self-focusing (see section 3.2.3), the Fourier plane was chosen 2 cm behind the crystal. The results of correlation with amplitude modulated images are presented in section 1.2.1.

In the correlation experiment, the image beam does not need to be detected as an image anymore, only the correlation output plane is of interest and phase modulated image can therefore be used. This has the additional advantage that it is possible to write Fourier plane holograms in the storage medium. The results of correlation based on phase modulated images, which exhibit also a much higher selectivity, are presented in section 4.2.2.

4.2.1 Correlation with amplitude modulated images

The performance of the correlator was tested using two different sets of 100 stored images. The first set is based on the Metamorphosis II pictures by Escher[140] (see Appendix F). The second set consists of 100 randomly generated bitmap images of 256 × 256 pixels, where each pixel was set randomly to 1 or 0 (see Fig. 1.6). These images produce an homogeneous intensity in the Fourier plane of the image and exhibit also components with the maximum spatial resolution that can be produced by our spatial light modulator. For both image sets the measured correlation plane is compared with the theoretically predicted correlation function. The correlation rate which has been achieved by detecting the output plane with a high frame rate CCD-camera of 10’000 images/s or 0.6 Gbit/s.

Escher-Images

Fig. 4.5 shows both the measured and theoretical predicted output for the case that image no. 76 is presented on the spatial light modulator. The measured output is in good qualitative agreement with the mathematical calculated correlation function. Image no. 76 can be correctly identified. As can be seen with the theoretical calculated curve, the inhomogeneous total number of bright pixels in the Escher images gives a smoothly changing correlation function. Therefore the correct identification of the image displayed on the SLM was not possible in every case.
4.2. Correlation experiments in LiNbO₃ and LiTaO₃

The experiment was repeated with the random image set. The result is reported in Fig. 4.7, where the correlation between the 100 random images and image no. 26 is shown. Image no. 26 can clearly be identified.

**Random Bitmap Pattern**

The correlation intensity between two non reciprocally shifted bitmap images is given by the square of the number of pixels which are in the 1 state in both images. The correlation between two identical random images is then given by \((n_{\text{pix}}/2)^2\) where \(n_{\text{pix}}\) is the total number of pixels. For two different images the correlation can be estimated to be \((n_{\text{pix}}/4)^2\) considering the probability 1/4 to find a pixel in the 1 state in both images. The correlation peak is therefore to be expected
4 time higher as the homogeneous background of the non matching images[141]. In the experiment the peak is found approximately two times the value of the background. This is probably due to image distortion by the self-focusing of the image beam, which is still present in the quasi Fourier plane holograms. Despite the contrast reduction, each one of the 100 images could be correctly recognized in our experiments. For amplitude modulated images the contrast can be further increased, presenting a reversed input image on the modulator. In this case the correlation peak corresponding to the matched filter will be minimal (equal to 0 in theory) compared to the peaks of the non matching images. By subtracting the correlator output for the reversed image from the output with the regular output, a much higher selectivity can be obtained.

A Vander Lugt correlator with quasi Fourier plane holograms does not have any shift invariance as predicted by Eq. (4.10). In fact, in the output plane the reconstructed reference beams are observed as a linear sequence of spots. If shift invariance is not required the output plane can also be read out by a linear CCD-array, which can show a much higher read-out rate well above 1000 lines/s. If in addition the number of matched filters is increased till 500 images, a correlation rate of 500'000 images/s or 32 Gbit/s can be reached. The correlation rate is then mainly limited by the spatial light modulator. With the newest commercially available high speed (3000 images/s) high resolution (1280 x 1024) ferroelectric spatial light modulator from Displaytech, correlation rates of 1’500’000 images/s or 2 Tbit/s should be feasible using the presented method.

Figure 4.7: Correlation of image no. 26 with the previously stored 100 random bitmap pictures.
4.2. Correlation experiments in LiNbO$_3$ and LiTaO$_3$

4.2.2 Correlation with phase modulated images

When phase modulated images are used, the 0th order peak in the Fourier plane of the input image is strongly reduced if the number of pixels in the 0 and 1 states are approximately the same. It is then possible to store the matched filters as Fourier plane holograms, and the shift invariance in the $x_0$ direction is preserved.

For phase modulated images the correlation presents also a much better selectivity because all pixels independently of their state are contributing to the generation of the correlation.

![Figure 4.8: Correlation output plane for image no. 33 with the previously stored 100 Escher-bitmap pictures. The plot represents a cut along the marked by the arrows in the image.](image)

The correlation output plane for the Escher-images read out with image no. 48 is shown in Fig. 4.8. Compared to previous results with amplitude images, the experiment shows a sharp correlation peak, 50 times higher than the background produced by diffraction at the non matching holograms. As can be best noticed in the enlargement of Fig. 4.8, the correlation peaks show also an extension in the $x_c$ direction, which represents the correlation between the input images reciprocally shifted in the $x_0$ direction. This shift invariance is introduced by the fact that the Fourier plane of the image are inside the crystal. All images could be correctly detected using the information coded in the phase. In Fig. 4.9 b) the results of the correlation with all the images are summarized in a single picture and compared with the theoretical correlation function in Fig. 4.9 a). The picture shows an excellent

\[\text{This in contrast to amplitude modulation where only the pixels in the 1 state are contributing to the correlation function. This can however partially be overcome by the additional correlation with the reversed image as mentioned previously.}\]
agreement between the experiment and theory. The experiments were also repeated with 100 random images (see Fig. 4.10 b)), giving even better results: no peaks of the non correlating image could be observed with the dynamic range (8 bit) of our CCD-camera.

It is also noteworthy that in correlation experiments the holograms can be read-out with much lower light power compared to the storage experiment. This because the light in the output plane is not distributed in a fully illuminated two dimensional region as during the read out of an image, but is concentrated in focused points. The advantage is that many correlations can be performed with a negligible erasure of the holograms. However the required detection time will determine the intensity of the input image beam during the read-out.

### 4.3 Potential applications

Correlation of a fixed pattern, as can be achieved with a photorefractive Vander Lugt type correlator is important in many industrial processes. For example the recognition of mechanical parts and their orientation on a production line, or for quality check of complicated structures. In all cases optimal results can only be achieved optimizing the geometry of the correlator, and choosing a suitable encoding of the information on the input pattern. For example, a gray scale image can be implemented as binary phase image, encoding the gray scale bits in a $n \times m$ rectangular region representing a pixel of the real image. In this way it is also possible to combine input images from different sources as for example cameras sensitive at different wavelength ranges. Additional features like scale or rotation
4.3. Potential applications

a) Theory

b) Experiment

Figure 4.10: Correlation of 100 random images. a) The theoretical correlation for unshifted phase images between the 100 images. b) Experimental results obtained by stacking of 100 slices of the output plane obtained by the correlation with the corresponding 100 images.

Invariance can be obtained by appropriate preprocessing techniques. Many different techniques[142, 143, 144, 145, 146] have been proposed based on diffractive optics or electronic preprocessing. In the next section we will shortly present two possible applications of the photorefractive Vander Lugt correlator.

4.3.1 Recognition of position and orientation of pattern

If the objects are at a fixed distance for example on a plane, no scale invariance is required. The only free parameters of an object are its position and orientation. The recognition of those parameters can be efficiently implemented with a photorefractive Vander Lugt type correlator. Shift invariance in one direction can be achieved by choosing the appropriate thickness of the holographic medium. The shift invariance in the second direction and the rotation invariance can be achieved by storing the reference hologram at different positions and orientations.

One might suggest to illuminate the scene with a laser and image it, by means of a lens system, on the holographic material. This is however not a practical solution. Much more flexibility is obtained capturing the input scene with a CCD-camera and reproducing the input image on a spatial light modulator in the correlator input plane. In this way it is also possible to preprocess and encode the image. The detection of the correlation peak can be performed by the use of a Dammann grating[147] or a second CCD-camera driven by dedicated hardware or a computer with frame grabber. The system could be made smaller than 40 x 40 x 20 cm³ included a diode pumped laser as light source. The response time of the system would mainly be given by the CCD-camera. Using high-frame rate cameras recognition and processing times of 1 ms are possible. The speed of such a system could therefore
outperform in execution time any all electronic device which calculate the cross correlation numerically by at least 3 orders of magnitude.

4.3.2 Key search for large databases

Large amounts of data are stored in databases. The information is mostly accessed performing complicated and time intensive queries. This queries can be implemented very efficiently based on a holographic memory operated as correlator. The number of stored pages in a holographic memory can be very high (5000 or more). Using large storage material it is possible to store several Gbyte of data. In the proposed system no data are stored, but only keys. For each record in an external database a particular region of one particular page of the holographic storage contains the key patterns which can be searched. Let’s call this part index region. Each page of the holographic memory will contain several hundreds of index regions all arranged in the same way across the stored pages. The keys are encoded using phase patterns with the three state scheme proposed in Fig. 4.11. The scheme offers the possibility

![Phase codes for tri state encoding of phase images for correlation applications.](image)

**Figure 4.11:** Phase codes for tri state encoding of phase images for correlation applications. Black and white represent 0 and 180° phase, respectively.

to store three different values: 0, -1, 1. The -1 and 1 pattern are used to encode the bit pattern of the key, whereas the 0 state can be used for not used bit in the index page and the bits which have not to be considered during the query. The key search can be implemented presenting a page with same search pattern for each index region. If a correlation peak is appearing, the corresponding index region can be extracted by a successively reducing the regions for which the input pattern is presented. With this system it would be possible to perform complex queries on multiple index keys for a database with more than 100'000 entries in less than 10 ms. The system is particularly attractive if large two dimensional patterns (for example fingerprints) have to be searched. Also in this application the total response time would be mainly given by the response times of the spatial light modulator and the detection system.

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*An additional property of this encoding scheme is that the resulting number of “on” and “off” pixels on the modulator are equal thus permitting the use of Fourier plane holograms.*
Chapter 5
Joint Fourier transform correlator

In the joint Fourier transform correlator (JFT) the object and scene patterns to be correlated are presented simultaneously at the input plane. Both are Fourier transformed either by a single lens (single-axis) or two lenses (dual-axis). The interference pattern of their Fourier transforms is recorded in a hologram in real time. The hologram is read-out by an additional plane wave. The light diffracted from the read-out beam contains the correlation function of the input patterns, which can be extracted performing one last Fourier transform. In the first part of this chapter the theory for the joint Fourier transformed correlator will be presented first for the case of thin grating (Raman-Nath regime) and later for the case of thick holographic media. In a subsequent section the pulsed interband photorefractive effect in tin hypothiodiphosphate (Sn$_2$P$_2$Se$_6$) is investigated. This is the optical nonlinearity used to build our JFT correlator. Afterwards a section presenting correlation experiments at high frame rate follows, and finally a section with examples of applications.

5.1 Principles of joint Fourier transform correlator

In the joint Fourier transform correlator proposed by Weaver and Goodman[18] two patterns with transmittance $s_1(x_0, y_0 + h)$ and $s(x_0, y_0 - h)$ are modulated on a collimated input beam in the input plane. The patterns are spatially separated: $s_1$ is positioned below the optical axis with its center at the point $(0, -h)$, and the second transmittance $s$ is positioned above the optical axis with its center at the point $(0, +h)$. The modulated patterns are Fourier transformed by lens 1 (see Fig. 5.1) which has its back focal plane exactly in the holographic medium. The light amplitude in the medium is then given by $\mathcal{F}\{s_1(x_0, y_0 + h) + s(x_0, y_0 - h)\}$, where $\mathcal{F}$ denotes the Fourier transform operator. The interference of the two input waves creates an intensity distribution

$$I(x, y) = |\mathcal{F}\{s_1(x_0, y_0 + h) + s(x_0, y_0 - h)\}|^2$$

$$= |S_1(x, y)|^2 + |S(x, y)|^2 + S_1(x, y) S^*(x, y) e^{-i\frac{2\pi}{\lambda} h y} + S^*_1(x, y) S(x, y) e^{i\frac{2\pi}{\lambda} h y}$$

where $S_1(x, y)$ and $S(x, y)$ are the Fourier transformed of $s_1(x_0, y_0)$ and $s(x_0, y_0)$, respectively, and $S^*$ indicates complex conjugation of $S$. 

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This intensity distribution which presents terms containing the products of the Fourier transformed of the input patterns is recorded either as amplitude or phase hologram in a thin real-time holographic material (for example a photographic-plate). In the following we explicitly assume that the hologram fixing process produces a change in refractive index $\Delta n$ (or an absorption change $\Delta \alpha$ for amplitude holograms) which is linear in respect to the intensity pattern described by Eq. (5.1).

The hologram is read-out by a plane wave having its wavevector perpendicular to the plane of the hologram. The diffracted light is Fourier transformed by lens 2 (see Fig. 5.1) and the correlator output plane can be found in the back focal plane of the same lens.

The output plane can be divided into three regions: At the coordinate $(x_c = 0, y_c = 0)$ the sum of the autocorrelation $s_1(x_0, y_0) \otimes s_1(x_0, y_0) + s(x_0, y_0) \otimes s(x_0, y_0)$ of the two input function is observed. The side regions at $(x_c = 0, y_c = -2h)$ and $(x_c = 0, y_c = +2h)$ coordinates corresponds to the last two terms of Eq. (5.1) and contain the cross-correlations $s_1(x_0, y_0) \otimes s(x_0, y_0)$ and $s(x_0, y_0) \otimes s_1(x_0, y_0)$ of the input patterns, respectively. The observed intensity distribution is proportional to the absolute square of the amplitude correlation function of the two scenes:

$$I(x_c, y_c \pm 2h) \propto |s_1(x_0, y_0) \otimes s(x_0, y_0)|^2 \propto \left| \int_{\text{aperture}} s_1(x_0 \pm x_c, y_0 \pm y_c) s^*(x_0, y_0) \, dx_0 \, dy_0 \right|^2$$

(5.2)

In the case of two equivalent images the cross-correlation has a maximum at $(x_c = 0, y_c \pm 2h)$. In the case of an object set consisting of several images the cross-correlation shows maxima at positions corresponding to the relative shifts between the similar portions of images in the input plane.

These properties are equivalent to the case of a Vander Lugt type correlator and can be used to identify the position of a smaller image inside a larger image, or to

![Figure 5.1: Schematic set-up for a single-axis joint Fourier transform correlator.](image)

$f$ is the focal length of the lenses 1 and 2.
find out if a certain object is present in a scene (multichannel JFT) as previously mentioned in Fig. 4.3.

Joint Fourier correlators have been proposed and implemented in many nonlinear optical materials: in photorefractive Bi$_{12}$SiO$_{20}$[148, 87, 149] and LiNbO$_{3}$[150], in GaAs and various others semiconducting materials[151], in multiple quantum wells[152], atomic vapors[141], and liquid crystal spatial light modulators[153]. The single-axis JFT technique, as presented in this section, is based on a scheme where the two data masks are encompassed by a single Fourier transform lens. Thus the lens is required to have a large numerical aperture when the signals of interest have a large space-bandwidth product (SBWP). The SBWP is defined as the area of the image divided by the achievable spatial resolution of a device, or in other words the effective number of pixels. Such a lens is difficult to produce in practice. A way to relax the requirements on the lenses is presented in section 5.1.2.

5.1.1 Joint Fourier transform correlator using volume holograms

The JFT correlator can also be realized using thick holographic material. The intensity distribution in the correlator output plane is then formed by Bragg diffraction of the read-out beam on the thick holographic medium, for example a photorefractive crystal. A first analysis based on Kogelnik’s results for diffraction from a volume hologram (Eqs. (2.6)-(2.10)) was presented by Nicholson et al.[154]. Because the theory is based on simple plane wave arguments, the analysis can only serve as a rough guide. Gheen et al.[151] presented a more detailed analysis, which takes into account the structure of the grating. Thereby the volume hologram is decomposed into a series of thin slices and the contributions from the composite thin holograms are added to get the final result. With this method the intensity distribution at the output plane can be calculated for the system sketched in Fig. 5.2. It is assumed that the beams are not significantly affected through scattering or absorption as
they pass through the material. Thus the result is only valid for small diffraction efficiencies.

The influence of thick grating in the JFT correlator can be derived based on the scalar diffraction theory in analogy to the calculation presented for the Vander Lugt type correlator in section 4.1.1.

Let’s consider the two input patterns \( s(x_0, y_0 - h) \) and \( s_1(x_0, y_0 + h) \), which are Fourier transformed by a lens (see Fig. 5.2). In the coordinate system of the holographic medium the amplitude \( S(x, y, z) \) produced by the input pattern \( s(x_0, y_0 - h) \) can be calculated in the same way as Eq. (4.8). We obtain

\[
S(x, y, z) = \frac{e^{2ki}}{i\lambda f} e^{in\lambda z} \int dx_0 dy_0 s(x_0, y_0 - h) e^{-i\frac{2\pi}{\lambda f}(x_0+y_0)} e^{-i\frac{\pi}{\lambda f} z} e^{\Delta x'(x_0^2+y_0^2)} \tag{5.3}
\]

where \( \Delta x' \), which was already defined in Eq. (4.8), is related to the distance between the Fourier plane of lens 1 and the crystal position. The amplitude \( S_1(x, y, z) \) produced by the second input pattern \( s_1(x_0, y_0 + h) \) has the same form as Eq. (5.3), but with \( s(x_0, y_0 - h) \) replaced with \( s_1(x_0, y_0 + h) \). The refractive index change produced by the interference of the two beams through the photorefractive effect can by approximated by* \( \Delta n \propto SS_1^* + c.c., \) \tag{5.4}

and is read-out by the plane wave

\[
R = R_0 e^{ik_r r}. \tag{5.5}
\]

The direction of the wavevector \( k_r = (k_{rx}, k_{ry}, k_{rz}) \) in the medium is chosen to satisfy the Bragg condition for the grating generated by the zero-order Fourier components of the two scenes. In paraxial approximation† we can rewrite the wavevector as

\[
k_{rx} = 0
\]

\[
k_{ry} = \frac{2\pi h}{\lambda f}
\]

\[
k_{rz} = -nk_0 \sqrt{1 - \left( \frac{k_{ry}}{nk_0} \right)^2}
\]

\[
\approx -\frac{2\pi h}{\lambda} + \frac{\pi}{n\lambda} \left( \frac{h}{f} \right)^2.
\tag{5.6}
\]

where \( k_0 \) is defined as \( k_0 = 2\pi/\lambda \). According to scalar diffraction theory the amplitude \( E \) of the light diffracted from the read-out beam \( R \) by the hologram \( \Delta n \) is given by

\[
E(r) \propto \int dr' e^{-i k_{\perp r'} R(r') \Delta n(r')}. \tag{5.7}
\]

*The influence of the complete photorefractive nonlinearity on the correlation process as been addressed in detail in Ref.[155]

†The paraxial approximation can be applied because the angle between the two input beam is limited by the numerical aperture of the lens.
where \( \mathbf{k}_d \) is the wavevector of the diffracted plane wave. Expressed in paraxial approximation the wavevector \( \mathbf{k}_d \) is given by

\[
\begin{align*}
\mathbf{k}_{d_x} &= +\frac{2\pi x_c}{\lambda f} \\
\mathbf{k}_{d_y} &= +\frac{2\pi y_c}{\lambda f} \\
\mathbf{k}_{d_z} &= -\frac{2\pi n}{\lambda} + \pi \frac{x_c^2 + y_c^2}{f^2}.
\end{align*}
\] (5.8)

where \((x_c, y_c)\) are the coordinates in the output plane (see Fig. 5.2). The intensity in the output plane is obtained evaluating Eqs. (5.3)-(5.8). The result is

\[
I_{corr}(x_c, y_c) \propto \int dx_0 dy_0 dx_1 dy_1 s(x_0, y_0 - h)s^*_1(x_1, y_1 + h)e^{-i\Delta z'}\left(\frac{\pi}{\lambda x f^2} x_1^2 + y_1^2 + x_0^2 + y_0^2 + h^2 - x_c^2 - y_c^2\right) e^{i\Delta z'}.
\] (5.9)

where \(l_x, l_y, l_z\) are the dimensions of the holographic medium in the \(x, y, z\) directions respectively and \(x_1, y_1\) are variables introduced by the integration.

The interpretation of Eq. (5.9) is similar as for the Vander Lugt type correlator (see Eq. (5.10)). In the case that \(l_x, l_y \gg l_z\), that is with a thin plate as holographic volume medium, the first two sinc functions can be replaced with a \(\delta\) function, and deliver the relations between the input coordinates and the coordinate in the output plane:

\[
x_c = x_1 - x_0 \\
y_c = y_1 - y_0 + h.
\] (5.10)

The argument of the third sinc function can be algebraically transformed using Eqs. (5.10)-(5.11). The resulting intensity in the output plane is

\[
I_{corr}(x_c, y_c) \propto \left| \int dx_0 dy_0 s(x_0, y_0 - h)s^*_1(x_0 + x_c, y_0 + y_c)\right|^2,
\] (5.12)

where \(\Delta z'\) was set to 0. The expression (5.12) is close to the desired correlation function (5.2). However, the Bragg effect has introduced an extra sinc-term. Note that the argument of the sinc-function is not symmetrical in \(x\) and \(y\). This can be verified considering that for two identical input patterns the correlation peak is centered at \((0, -h)\) in the output plane. Introducing the maximum spatial extents \(D_x\) and \(D_y\) in the \(x_0\) and \(y_0\) direction of the input patterns \(s(x_0, y_0 - h)\) and \(s_1(x_0, y_0 + h)\), it
is possible to evaluate the sinc-function at half energy \(\text{sinc}^2(x) = 1/2\) and derive following condition for the maximal grating thickness of the holographic medium

\[
l_z < \frac{0.9n\lambda f^2}{D_x^2 + D_y(D_y + 2h)}.
\]

(5.13)

Here the shift invariance can be considered as shift of a smaller pattern inside the input rectangle with the size \(D_x \times D_y\). A graphical representation of Eq. (5.13) is given in Fig. 5.3. For a pattern size \(D_x = D_y = 4\) mm, a distance \(2h = 44\) mm, a lens with focal length \(f = 250\) mm, a refractive index \(n = 3\), and the wavelength \(\lambda = 532\) nm we obtain a maximum thickness of 430 \(\mu\)m.

The influence of the position of the Fourier plane in respect to the holographic medium, described by \(\Delta z'\), is the same as in the case of the Vander Lugt type correlator and can be treated in a similar way as the argument of sinc function. For the maximum displacement we obtain

\[
\Delta z' < \frac{1}{4} \frac{n\lambda f^2}{D_x^2 + D_y(D_y + 2h)}.
\]

(5.14)

The error of positioning of the Fourier plane has therefore to be 4 times smaller than the thickness of the holographic grating.

The maximum number of pixels that can be processed in parallel, i.e. the space-bandwidth product is given by the width of the correlation peak in the output plane. For this purpose let’s consider two ideal input patterns, one consisting of a single point source and the second consisting of two point sources placed at a distance \(\delta_{12}\) between each other. The point sources can be identified if the corresponding correlation peaks in the output plane are separated. The width of the correlation peak is given by the first two sinc terms in Eq. (5.9). Evaluating the argument of
the sinc function we obtain the following condition for the minimal feature size in the input planes

$$\delta_{12} > \frac{\lambda f}{l_{x,y}}.$$  \hspace{1cm} (5.15)

where \(l_{x,y}\) denotes the crystal size either in \(x\) or \(y\) direction. The maximum space-bandwidth product \(N_{\text{max}}\) is obtained dividing the maximum image size in the input plane \(D_{\text{max}}\) by \(\delta_{12}\). The maximum image size \(D_{\text{max}}\) is limited by Eq. (5.13). Choosing a minimal \(h = D_{\text{max}}/2\) which leads to a maximum image size and setting \(D_{\text{max}} = D_x = D_y\), Eq. (5.13) can be solved with respect to \(D_{\text{max}}\). Dividing the obtained expression by Eq. (5.15) we obtain a maximum space bandwidth product of

$$N_{\text{max}} < 0.55 \frac{l_{x,y}}{\sqrt{\lambda f}}.$$  \hspace{1cm} (5.16)

In the case the grating is generated by interband photorefraction, the grating thickness \(l_z\) is determined by the absorption of the material. For an absorption of 100 cm\(^{-1}\) (as shown in Sn\(_2\)P\(_2\)S\(_6\) around 50°C) we obtain a grating thickness of less than 100 \(\mu\)m, resulting in a maximum space bandwidth product of 370 for a \(l_{x,y} = 5\) mm large crystal. Note however that for the input image size \(D_{x,y}\) used in the previous numerical examples the resolution is reduced to 125 x 125 pixels.

In conclusion we have shown that despite of their strong theoretical similarity, the impact of using thick holograms in a joint Fourier transform correlator is very different from the case of the Vander Lugt type correlator. In the latter the thick holograms allow an efficient correlation of multiple images, at the cost of the shift invariance. In the case of the JFT correlator in contrast the use of thick holograms limits the maximum usable image size in the input plane (and therefore the shift invariance), thus forcing the use of thinner holographic media.

A simple way to achieve thin hologram also in large sample, consists in writing the hologram with light that is strongly absorbed. This is fulfilled in the case of interband photorefraction (see section 2.3) and a material showing high sensitivities in the blue-green spectral region will be presented in section 5.2.

**Impact of read-out wavelength**

The wavelength \(\lambda_R\) of the read-out plane wave \(R\) can be chosen different from the wavelength \(\lambda\) of the hologram writing waves \(S\) and \(S_1\). The advantages are twofold: First the read-out beam does not interfere with the input beams and write parasitic holograms. The second advantage is that it is possible to select a wavelength \(\lambda_R\) where the material shows no sensitivity or absorption. The effect of a \(\lambda_R \neq \lambda\) can be calculated modifying appropriately Eqs. (5.6) and (5.8). Note that the angle \(\phi\) in Fig. 5.2 will have to be readjusted for matching the Bragg-condition of the grating written by the zero order Fourier components of the input beams. Also, the meaning of the output plane coordinate will change, resulting scaled by a factor of \(\lambda_R/\lambda\) in respect to the previously reported relations (5.10) and (5.11). Following the same method as presented above we obtain the following condition for the maximum
thickness $l_z$ of the material:

$$l_z < \frac{0.9\eta f^2}{D_x^2 + D_y(D_y + 2h) + \left|1 - \frac{\Delta}{\lambda x^2}\right|(6h^2 + D_y^2 + h(5D_y + 3D_x))}.$$  \hspace{1cm} (5.17)

For $\lambda_R \neq \lambda$ the maximum thickness $l_z$ is therefore additionally reduced. For the parameters stated above and a read-out wavelength $\lambda_R = 633$ nm we obtain a reduction of 20% of the maximum crystal thickness.

### 5.1.2 Dual-axis joint Fourier transform correlator

![Optical architecture for a dual-axis joint Fourier transform correlator](image)

**Figure 5.4:** Optical architecture for a dual-axis joint Fourier transform correlator. $f$ is the focal length of the lenses.

In the dual-axis joint Fourier transform correlator the two input signals are Fourier transformed by two different lenses (Fig. 5.4), thus greatly reducing the numerical aperture requirements on the lenses. Although the dual-axis JFT correlator relieves the stringent requirements on the FT lens, it does introduce the problem that the two Fourier planes have a relative tilt with respect to each other. A first tentative analysis of the impact of the tilted Fourier planes on the correlator output was proposed by Lee\[156\]. The method is based on a plane wave argument for calculating the broadening of the correlation peak in the case of tilted Fourier planes.

An exact solution in the paraxial approximation can be obtained if the amplitude $S(x,y,z)$ of the input beams in the plane of the holographic material defined in Eq. (5.3) is replaced by

$$S = \frac{e^{i2k_f}}{i\lambda f} e^{i\kappa k z} \int dx_0 dy_0 s(x_0, y_0) e^{-\frac{\pi}{\lambda f}(x x_0^2 + y y_0^2)} e^{-i\frac{\pi}{\lambda f}(z y_0 + n - \Delta z')} e^{-i\frac{\pi}{\lambda f}(x y_0 + \Delta x')}$$  \hspace{1cm} (5.18)

where $\theta$ is the half angle between the axis of the correlator (see Fig. 5.4).
5.2. \textit{Sn}_2\textit{P}_2\textit{S}_6 as fast real time correlation material

Repeating the calculation leading to Eq. (5.9) and approximating $\sin \theta \approx h/f$ we find that the effect of the tilted Fourier planes can be neglected if the lateral crystal size $l_y$ in the $y$ direction is

\begin{equation}
l_y < \frac{0.9n\lambda f^2}{D_x^2 + D_y(D_y + 2h)/h} n f.
\end{equation}

The major impact of the tilted Fourier planes is therefore a reduction of the usable crystal width in $y$ direction. This can be understood considering that the separation between the tilted Fourier planes is a linear function of the $y$ coordinate and presents its maximum at the $y$ boundaries. For the same parameters as used above we obtain $l_y < 5.7$ mm.

Therefore the restrictions imposed by the tilted Fourier planes in the dual-axis JFT correlator are therefore not limiting the technical realization. The use of two separate lenses increases, however, the requirement on the relative positioning of the components.

5.2 \textit{Sn}_2\textit{P}_2\textit{S}_6 as fast real time correlation material

Tin hypothiodiphosphate ($\text{Sn}_2\text{P}_2\text{S}_6$) single crystals is a ferroelectric material below $T_c = 66^\circ$C. In this phase the crystal is monoclinic with an optical absorption edge of $E_B = 2.3$ eV ($\lambda_B = 533$ nm) at room temperature. This makes the material very well suited for interband photorefractive (see section 2.3) in the blue-green spectral region. Experimental results of continuous wave photorefractive interband measurements at a wavelength of 488 nm and 514 nm, are presented in Appendix C. In this section we present a complete characterization of $\text{Sn}_2\text{P}_2\text{S}_6$ at a wavelength of 532 nm. The material was characterized by absorption and photoconductivity measurements, and photorefractive diffraction experiments in a 50 ns pulsed regime. The photorefractive diffraction was measured as function of the grating spacing, the crystal temperature, and the fluence of the laser pulse. The section ends with a characterization of the diffraction efficiency at high repetition rate which is of crucial importance for the temporal performance of a joint Fourier transform correlator.

5.2.1 Absorption measurement

The absorption of $\text{Sn}_2\text{P}_2\text{S}_6$ was measured by means of a Perkin-Elmer $\lambda$-9 spectrophotometer, in a crystal cut along the crystallographic axes with the spontaneous polarization nearly parallel to the OX-axis (See Appendix D.4 for the definition of crystal axes). The size of the sample is $X \times Y \times Z = 5.6 \times 5.9 \times 1.61$ mm$^3$ with the polished surfaces perpendicular to the $Z$ direction. The absorption was calculated from the intensity transmission $T$ delivered by the spectrometer. In the case multiple reflections at the crystal surfaces are considered, following expression for the absorption $\alpha$ can be derived

\begin{equation}
\alpha = -\frac{1}{d} \ln \left( \frac{\sqrt{(1-R)^4 + 4R^2T^2} - (1-R)^2}{2R^2T} \right), \tag{5.20}
\end{equation}
where $R$ is the Fresnel intensity reflection at the crystal surface, and is related to the refractive index $n(\lambda)$ by

$$ R = \left( \frac{n(\lambda) - 1}{n(\lambda) + 1} \right)^2. \quad (5.21) $$

The wavelength dependence of the refractive indeces of Sn$_2$P$_2$S$_6$ is reported in Appendix I.1.

The resulting absorption are reported for room temperature in Fig. 5.5 b) for two different linear polarizations of the incident light$^4$. The measured absorption

![Figure 5.5: Optical absorption measurements for light polarized parallel and perpendicular to the polar X-axis in a Z-cut Sn$_2$P$_2$S$_6$ crystal. a) The absorption is plotted as a function of the crystal temperature for a wavelength $\lambda = 532$ nm. b) As function of the wavelength at room temperature.](image)

coefficients are 23 cm$^{-1}$ at 532 nm and 0.5 cm$^{-1}$ at 633 nm with the polarization (defined here parallel to the direction of the electric field $E$) parallel to the $X$-axis. For $E$ parallel to $Y$ we have 7 cm$^{-1}$ at 532 nm and 0.5 cm$^{-1}$ at 633 nm, respectively.

The absorption for a wavelength of 532 nm is strongly dependent on the temperature. The crystal was therefore temperature stabilized in a controlled oven, and the transmittance and reflection change measured in function of the temperature. The resulting absorption is reported in Fig. 5.5b). Changing the temperature from 20$^\circ$C to 55$^\circ$C, the absorption coefficient can be tuned from 7 till 120 cm$^{-1}$. This corresponds to a penetration depth $\delta_\alpha = 1/\alpha$ ranging from 8 till 142 $\mu$m. This is an important property which allows to adjust the effective thickness of photorefractive interband gratings. As mentioned in section 5.1.1 the grating thickness is of fundamental importance for the performance of a JFT correlator.

$^4$Note that monoclinic crystals are optically biaxial with the optical axis rotated with respect to the crystal axes.
5.2. Photoconductivity

Absorption measurements reveal the spectral region where light is interacting with the material. The generation of free charge carriers necessary for the photorefractive effect, can be investigated by photoconductivity measurements. In these measurements the conductivity change is measured as a function of the incident light intensity (that is the light intensity incident on the material after deducing the reflection losses at the material surface). The light was incident on the surface perpendicular to the \( Z \) axis, and an external field was applied by means of two silver painted electrodes on the \( X \)-faces. This direction is nearly parallel to the polar axis and therefore the relevant direction for writing photorefractive grating (see also Appendix D.4). Photoconductivity measurements were performed under continuous wave illumination at a wavelength of 532 nm. The electric current \( I_{el} \) was measured for different light intensities under an external field of \( E = 200 \) V/cm. By applying the method presented in section 2.3.3 the photoconductivity reported in Fig. 5.6 was calculated. In this figure the photoconductivity is reported for 27 and 50°C. At the higher temperature and for a light intensity larger than 2 mW/cm\(^2\) the photoconductivity follows a straight line with a slope of 1/2. In the double logarithmic representation of Fig. 5.6 this corresponds to a square root dependence on the intensity, as expected in the case of interband photoexcitation (see section 2.3.2). At 27°C the absorption is much smaller and the photoconductivity is partially affected by the presence of trapping levels in the band gap. The conductivity generated by the light illumination is very high, compared to ferroelectric oxide like \( \text{KNbO}_3 \) and \( \text{LiNbO}_3 \), because of the high absorption, which generates a large number of free charges.

![Figure 5.6: Photoconductivity \( \sigma_{ph} \) measurement for 27 and 50°C in a Sn\(_2\)P\(_2\)S\(_6\) crystal. The photoconductivity was obtained from photocurrent measurement at \( \lambda = 532 \) nm by the method described in section 2.3.3.](image-url)
The dark conductivity \( \sigma_{\text{dark}} \) was also measured and found to be strongly dependent on the previous light illumination. Maximal \( \sigma_{\text{dark}} \) was measured after preillumination with green light (532 nm). In this condition we measured \( 7 \cdot 10^{-9} \ \Omega^{-1}\text{m}^{-1} \) at room temperature, and \( 9 \cdot 10^{-8} \ \Omega^{-1}\text{m}^{-1} \) at 50°C. \( \sigma_{\text{dark}} \) is therefore much smaller than the photoconductivity at a light intensity of 20 \( \mu\text{W/cm}^2 \). The fact that the dark conductivity depends on the previous illumination indicates that multiple traps level which can be populated by visible wavelength are present in Sn\(_2\)P\(_2\)S\(_6\) crystals also if they are not intentionally doped. Note that the lifetime of these metastable light populated levels is longer than several weeks at room temperature.

The sign of the majority charge carriers was determined by beam coupling experiments and was found to be electrons at a wavelength of 633 and 532 nm at room temperature. At 50°C and 532 nm, the method would require thin samples. In addition two beam coupling gain is also expected in the case of pure interband photorefraction, where the effect is driven by the mobility difference between electrons and holes.

The wavelength dependence of the photocurrent \( I_{\text{ei}} \) is reported in Ref. [157]. The photocurrent presents two peaks at photon energies \( \nu h \) of 2.20 eV \( \equiv \) 563 nm and 2.39 eV \( \equiv \) 519 nm. The first peak presents an energy which is clearly below the photon energy of our laser (2.3 eV). This indicates that beside the transitions from trapped charge carriers in the bands, also direct band-to-band transitions are present in our measurements. Which one of these two photoexcitation mechanisms is dominating, is finally dependent on the light intensity. At low intensities both effect are present contemporaneously, whereas at high intensity the trap level can be neglected because they are either completely filled up or emptied by the high density of free electrons and holes produced by the interband photoexcitation.

The dynamics of the build-up of the photoconductivity under continuous wave illumination was also considered. The results are reported in Fig. 5.7 where the experiment (solid line) was fitted by the theoretical curve (dashed line) obtained by numerical integration of Eq. (2.64). The theoretical curves follows the experimental point very precisely for the first 3 ms. The difference after 3 ms is probably given by the influence of traps within the band gap. Note that the crystal was not intentionally doped but showed a strong photorefractive effect at 633 nm. In order to reduce the effects of traps the measurement was performed at a temperature of 60°C and at a moderate light intensity of 200 mW/cm\(^2\). Analyzing the time dependence of the build-up, it is possible to estimate the interband recombination constant \( \gamma_{\text{dir}}^{\text{cond}} \) and the sum of the mobilities \( \mu_{\text{e}}^{\text{cond}} + \mu_{\text{h}}^{\text{cond}} \). The obtained parameters are \( \gamma_{\text{dir}}^{\text{cond}} = 8 \cdot 10^{-20} \ \text{m}^3/\text{s} \) and \( \mu_{\text{e}}^{\text{cond}} + \mu_{\text{h}}^{\text{cond}} = 1.7 \cdot 10^{-4} \ \text{cm}^2/\text{Vs} \), where we have introduced the exponent “cond” to identify that the values were obtained by photoconductivity measurements. The value of the measured photoconductivity and the interband recombination constant have to be considered as lower limit because there were calculated assuming the quantum efficiency \( \Phi = 1 \) and mainly because of the influence of shallow traps which are weakly trapping the free charge carriers in the bands. The shallow traps were shown to have an considerable impact on the charge transport in photorefractive oxides[158, 159, 160] and semiconductors[161], where mobility reduction of 4 orders of magnitude were observed. The influence of
shallow traps can be avoided characterizing the charge transport by optical methods as described for example in Ref. [57].

Photoconductivity measurements under pulsed illumination however were not possible, because of the presence of a strong and fast (shorter than 20 ns) current peak due to the pyroelectric effect, which was hiding the 1000 times lower and slower current peak of the photoconductivity change.

### 5.2.3 Diffraction efficiency

The photorefractive properties of Sn$_2$P$_2$S$_6$ were characterized by diffraction experiments. The set-up is presented in Fig. 5.8. Holographic gratings were written by the interference of two plane waves produced by a a frequency doubled Nd:YAG laser in Q-switch operation ($\lambda = 532$ nm and 50 ns pulse length). The grating was tested under Bragg-angle by a third beam with a wavelength of 633 nm and therefore only weakly absorbed by the material. The Q-switched laser produced horizontal polarized light and was triggered simultaneously with the data acquisition system by a frequency generator. The chosen symmetric configuration is such that the generated grating vector is parallel to the $X$-axis (polar axis). The diffracted beam is separated from the writing beams by a dichroic filter and detected by a photomultiplier. The Sn$_2$P$_2$S$_6$ crystal is held in a temperature controlled oven which allows to change the temperature of the crystal in a range of 20-60°C.

\[\text{Even if the pyroelectric effect is very strong in } \text{Sn}_2\text{P}_2\text{S}_6 \text{ its contribution to the build-up of the holographic grating (see section 5.2.3) was shown to be negligible following the arguments presented in Appendix C.}\]
Figure 5.8: Set-up for pulsed interband photorefractive measurements in Sn$_2$P$_2$S$_6$ crystal. The photorefractive grating is written by the green laser, and read out with a weakly absorbed red beam.

A typical curve for the dynamics of the diffraction efficiency $\eta$ under pulsed illumination is shown in Fig. 5.9. The pulse fluence was 100 $\mu$J/cm$^2$, the crystal temperature 50°C, and the grating spacing $\Lambda_g = 0.8 \mu$m. The diffraction efficiency presents a strong peak with a full length at half maximum of 3 $\mu$s. The pulse length of the laser for this experiment was 50 ns and therefore much shorter than the diffraction efficiency transient. In Fig. 5.9 we also show a simulation for the squared amplitude of the space charge field $|E_{sc}|^2$ which is expected to be proportional to $\eta$ in the case of infinitesimal thin grating (thickness $d \ll 1/\alpha$). The parameter used for the calculation are $\gamma_{dir} = 6.8 \cdot 10^{-19}$ m$^3$/s, $\mu_e = 2.4 \cdot 10^{-3}$ cm$^2$/Vs, and $\mu_h = 8.0 \cdot 10^{-3}$ cm$^2$/Vs and where obtained by analyzing the first 500 ns of the build-up. The resulting sum of the mobilities $\mu_e + \mu_h = 10^{-2}$ cm$^2$/Vs, which has to be considered a raw estimation, is approximately two orders of magnitude higher than the value obtained by the photoconductivity measurement in 5.2.2, whereas the direct band-to-band recombination constant $\gamma_{dir}$ is approximately one order of magnitude higher in the pulsed experiment. The difference of the parameters can be explained considering the influence of shallow traps[163] and was also observed in many others photorefractive material like BaTiO$_3$[160], Bi$_{12}$SiO$_{20}$[160], KNbO$_3$[58, 59]. Note that the band mobility measured in the ns time scale after photo excitation with ps laser pulses, is expected to be larger by at least 2-3 orders of magnitude, as observed in other materials[162, 163, 164, 54, 165], unless the mobility in Sn$_2$P$_2$S$_6$ would be strongly reduced by the formation of polarons[54]. The simulation presented in Fig. 5.9 is in qualitative good agreement with the experiment. Note that the dynamics of the diffraction efficiency can not be directly related to the space charge field, because the intensity of the writing beam is exponentially decreasing as function of the depth inside the crystal. For the build-up of the space charge field this means that the
5.2. $\text{Sn}_2\text{P}_2\text{S}_6$ as fast real time correlation material

Sn2P2S6 as fast real time correlation material

\section*{Diffraction efficiency measurement}

\section*{Numerical calculation of $|E_{sc}|^2$}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.9.png}
\caption{Diffraction efficiency $\eta$ (solid curve) of a Sn2P2S6 crystal under pulsed illumination. The pulse fluence is 100 $\mu$J/cm$^2$ the crystal temperature is 50°C and the grating spacing is $\Lambda_g = 0.8 \mu$m. The dashed line shows a numerical calculation for the square of the space charge field $E_{sc}$, calculated with $\gamma_{dir} = 6.8 \times 10^{-19}$ m$^3$/s, $\mu_e = 2.4 \times 10^{-3}$ cm$^2$/Vs, and $\mu_h = 7.6 \times 10^{-3}$ cm$^2$/Vs.}
\end{figure}

Figure 5.9: Diffraction efficiency $\eta$ (solid curve) of a Sn2P2S6 crystal under pulsed illumination. The pulse fluence is 100 $\mu$J/cm$^2$ the crystal temperature is 50°C and the grating spacing is $\Lambda_g = 0.8 \mu$m. The dashed line shows a numerical calculation for the square of the space charge field $E_{sc}$, calculated with $\gamma_{dir} = 6.8 \times 10^{-19}$ m$^3$/s, $\mu_e = 2.4 \times 10^{-3}$ cm$^2$/Vs, and $\mu_h = 7.6 \times 10^{-3}$ cm$^2$/Vs.

build-up time, the amplitude, and the phase of $E_{sc}$ will depend on the depth. The diffracted beam at the back side of the crystal is formed by the coherent sum of all the amplitudes of the diffracted light at different depths.

The diffraction efficiency transient presents a build-up time which is in the same order of magnitude as the successive grating decay time. This is a strong indication that the grating decay is driven by the quadratic interband recombination term $-n_p\gamma_{dir}$ (see Eqs.(2.58)-(2.62)). In conventional photorefractivity the decay in the dark would be driven by the dark conductivity and therefore be in a milliseconds till seconds time scale for Sn2P2S6. That is 3-5 orders of magnitude slower than the observed effect. Based on this fact, on the photoconductivity measurements of section 5.2.2, and on the estimation of the pyroelectric contribution presented in Appendix. C, we conclude that the diffraction efficiency transient observed in this work are driven by band-to-band photoexcitation.

\section*{Pulse fluence and grating spacing dependence}

The maximum diffraction efficiency $\eta_{max}$ and the length of the diffraction efficiency transient at full length half maximum $\tau_\eta$ were measured for three different grating spacings $\Lambda_g = 0.8$, 1.6, and 6.6 $\mu$m, and by changing the pulse fluence from 2 to 100 $\mu$J/cm$^2$. The results are reported in Fig. 5.10, where the dependence of the square
Figure 5.10: The square root $\sqrt{\eta_{\text{max}}}$ of the maximum diffraction efficiency, is plotted as function of the pulse fluence of the green writing beams, and for three different grating spacings $\Lambda_g = 0.8 \, \mu\text{m}, 1.6 \, \mu\text{m},$ and $6.6 \, \mu\text{m}$.  

The square root of the diffraction efficiency is plotted as function of the fluence in a logarithmic scale. Considering the exponentially decreasing fluences inside the crystal and under the crude assumption that the maximum refractive index modulation $\Delta n_{\text{max}}$ of the holographic grating is not dependent on the fluence of the writing beams, the maximum diffraction efficiency can be approximated by

$$\sqrt{\eta_{\text{max}}} = \frac{\pi}{\lambda \cos \theta_2} \frac{\Delta n_{\text{max}}}{\alpha} \ln(F/F_{\text{min}}),$$

(5.22)

where $F_{\text{min}}$ is the minimal fluence producing a refractive index change $\Delta n_{\text{max}}$. Relation (5.22) results in straight lines for the representation coordinates chosen for Fig. 5.10. In fact, the maximum diffraction efficiency follows straight lines for fluences larger than 10 $\mu\text{J/cm}^2$. For lower fluences, the fluence dependence of the square root of the diffraction efficiency is much weaker, thus indicating two distinct effects which are dominating one over the other in the two different fluence regimes.

Considering the grating spacing dependence we see that the $\sqrt{\eta_{\text{max}}}$ is higher for shorter grating spacings. By numerical simulations of Eqs. (2.87)-(2.89) it can be shown that this grating spacing dependence is predicted for a photorefractive process which presents a charge carrier life time $\tau_{\text{dir}}$ which is smaller than the diffusion time $\tau_{D,h/e}$ (See section 2.4.2). The diffusion time is proportional to $\Lambda_g^2$ and therefore much longer for large grating spacings. In this case the charge carrier will recombine before the diffusion process reaches the maximum possible space charge field, thus the resulting space charge field will be smaller for larger grating spacings.
The experiments therefore indicates that in Sn$_2$P$_2$S$_6$ the assumption leading to the solution (2.95-2.98) are not fulfilled and a solution of the pulsed interband equations (2.87-2.89) has to be found by numerical integration.

The impact of the carrier life time which is shorter than the diffusion time is also confirmed by the grating spacing dependence of the diffraction efficiency pulse length $\tau_\eta$. From the experiment we obtained $\tau_\eta(\Lambda_g = 0.8 \, \mu m) = 4.5 \, \mu s$, $\tau_\eta(\Lambda_g = 1.6 \, \mu m) = 6.5 \, \mu s$, and $\tau_\eta(\Lambda_g = 6.6 \, \mu m) = 8.0 \, \mu s$. The grating spacing dependence is much weaker than the $\Lambda_g^2$ dependence predicted for a purely diffusion driven process. Note that, the length $\tau_\eta$ of the diffraction efficiency transient is practically independent on the pulse fluence for a fluence range of 20 – 110 $\mu J/cm^2$.

**Temperature dependence**

The crystal temperature is influencing several relevant parameters of the pulsed photorefractive grating build-up: the absorption (see Fig. 5.5) which is responsible for the grating thickness and the efficiency of the photoionization process, the electro-optic coefficient $r_{\text{eff}}$ and the dielectric permittivity $\epsilon_{\text{eff}}$ which are supposed to be strongly temperature dependent near the phase transition at 66°C.

Measurements for the maximum diffraction efficiency $\eta_{\text{max}}$ are reported as function of the temperature and for three different grating spacings in Fig. 5.11. The experiments were performed under the same condition as described above with a pulse fluence of 110 $\mu J/cm^2$.

![Figure 5.11: Diffraction efficiency $\eta_{\text{max}}$ vs. crystal temperature $T$ for three different grating spacings (pulse fluence 110 $\mu J/cm^2$).](image)

The interpretation of the measurements is complex. For shorter grating spacings ($\Lambda_g < 3 \, \mu m$), $\eta_{\text{max}}$ decreases by around one order of magnitude between 20 and
35°C and stays practically constant at higher temperatures. Assuming that the only effect of the absorption on $\eta_{\text{max}}$ is a change on the grating thickness as suggested by the fluence dependency measurements of section 5.2.3, the decrease of $\eta_{\text{max}}$ in the temperature range can be explained qualitatively. At higher temperatures the effect is partially compensated by the increase of the electro-optic effect. For the second order phase transition observed in Sn$_2$P$_2$S$_6$ at $T_c = 66°C$, the electro-optical coefficient is predicted to change with

$$\varepsilon_{\text{eff}}(T) \propto P_s(T)\varepsilon(T) \propto (T - T_c)^{-1/2}$$  \hspace{1cm} (5.23)

where $P_s$ denotes the spontaneous polarization and $\varepsilon$ is the dielectric permittivity.

At large grating spacings ($\Lambda_g > 5 \mu m$) the diffraction efficiency $\eta_{\text{max}}$ is only weakly dependent on temperature and showing a minimal $\eta_{\text{max}}$ around 35°C. Considering the time dependence of the diffraction efficiency at large grating spacings reported in Fig. 5.12, we note that after the first fast peak with a length of 3-12 $\mu$s, a second much slower peak with a length around 1-2 ms appears. The second peak is observed only at large grating spacings and can be explained considering the interband model with one additional trap level presented in section 2.4.3.

A particularly interesting temperature is 38°C, where the diffraction efficiencies are almost the same for all measured grating spacings. This is an important property which can be exploited to increase the grating spacing range accessed by the correlator and therefore its space bandwidth.

The length $\tau_{\eta}$ of the diffraction efficiency transient is reported for the three grating spacings in Fig. 5.13. As general tendency for all grating spacings the response

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure512.png}
\caption{Diffraction efficiency of the reading beam vs. time for different temperatures, pulse fluence of 110 $\mu J/cm^2$, and $\Lambda_g = 6.6 \mu m$}
\end{figure}
Figure 5.13: Pulse length of the diffraction peak (FWHM) \( \tau_n \) as function of the crystal temperature and for different grating spacings \( \Lambda_g \), a pulse fluence of 110 \( \mu J/cm^2 \), and a pulse length of 50 ns.

Pulse length \( \tau_n \) becomes shorter for higher temperature. This can be qualitatively explained by the higher absorption at higher temperature which are producing a higher band-to-band photoexcitation rate (see Eq. (2.63)). It is noteworthy that at a temperature of 50°C, \( \tau_n \) is as short as 2.5 \( \mu s \) and approximatively the same for all grating spacings.

**High Repetition Rates**

In the pulsed diffraction experiments presented so far the repetition rate was held small (less than 100 Hz) to avoid cumulative effects. In contrast, in this section we want to investigate the maximum possible correlation rate of a \( \text{Sn}_2\text{P}_2\text{S}_6 \) based JFT correlator under pulsed illumination. At high correlation rates two fundamental limitations are observed. First the time between the pulses is not long enough to allow the grating written by the previous pulse to decay, and cumulative effects are produced. The problem can be avoided by introducing a slowly moving mirror (for example mounted on a piezo-electric actuator) in the set-up which is slowly changing the reciprocal phase of the writing beam, or by electronic filtering (high pass) of the detection signal. The second limitation observed at high correlation rates is given by the maximum allowed average intensity of the writing beam. It was found in experiments at 488, 514, and 532 nm, that the signal starts to show instabilities if the mean intensity is higher than 100-200 mW/cm\(^2\). This is probably related to mechanical deformations produced by the temperature gradient near the surface were the light is strongly absorbed. A typical time dependence of the diffraction efficiency at a
repetition rate of 10 kHz and a pulse fluence of 10 µJ/cm² is shown in Fig. 5.14. At 10 kHz the build-up and decay of the diffraction efficiency of the holographic grating are still fast enough to allow Sn₂P₂S₆ to be used as correlator. The dependence of the maximum diffraction efficiency change \( \eta_{\text{max}} \) on the fluence is shown in Fig. 5.15. For fluences larger than 20 µJ/cm² the diffraction efficiency start to fall down, and a distortion of the diffracted beam can be observed. The effect can be observed also at other repetition rates, and in all cases the signal starts to degrade when a mean intensity of 200 mW/cm² is reached. The maximum diffraction efficiency, which can be reached without distortions of the diffracted beam, as function of the repetition rate, is summarized in Fig. 5.16. The diffraction efficiency strongly diminishes for repetition rates higher than 3 kHz. However correlation rates of 10 kHz are still possible as we will show in section 5.3.

The aim of section 5.2 was to identify the relevant properties of Sn₂P₂S₆ to be used as joint Fourier transform correlator. We have found that the photorefractive properties of Sn₂P₂S₆ allow to build a correlator working at a wavelength of 532 nm with a response time comparable to correlators based on semiconductor materials. The response time is shorter than 3 µs with a pulse fluence of 1-10 µJ/cm², with sensitivities as high as \( S_1 = 2.5 \cdot 10^{-9} \text{m}^3/\text{J} \), \( S_2 = 7 \cdot 10^{-6} \text{m}^2/\text{J} \) which are comparable with the sensitivity of KNbO₃ in the blue (see Appendix B). The maximum repetition rate of 10 kHz which can be achieved is limited by the maximum mean intensity of 200 mW/cm² above which the response degrades. By fixing the crystal temperature at 50°C a 100 µm thick grating is achieved, allowing...
5.3. Correlation experiments in $\text{Sn}_2\text{P}_2\text{S}_6$

A dual axis joint Fourier transform correlator was implemented using a $\text{Sn}_2\text{P}_2\text{S}_6$ crystal as correlation material. The fast image sequence produced by the holographic storage set-up presented in section 3.4 is used as input for the correlator, whereas the second image comes from a ferroelectric liquid crystal spatial light modulator. A schematic representation of the set-up is shown in Fig. 5.17. The right upper part of the set-up in Fig. 5.17 corresponds to the holographic storage system operated with image plane holograms (see Fig. 3.7), whereas in the left lower part the two arms of the dual axis JFT correlator can be recognized. As the storage set-up was already described in detail in section 3.4, we will present only the JFT correlator part and the eventual modification of the storage set-up. The reconstructed image beam of the storage system is used as input beam for the correlator. The beam is deflected by the mirror pair 2-3 and Fourier transformed by lens 11 (focal length 250 mm). The second input beam of the correlator is derived from the original image beam of the storage system by beam splitter 1. After deflection at mirror 4 the beam is also Fourier transformed by lens 10. The paths length of the two arms are adjusted to reach a space-bandwidth (number of pixels) of 75 for each mm of lateral crystal size. At the temperature of 40°C the diffraction efficiency depends only slightly on the grating spacing in a range of $\Lambda_g = 0.8 \cdots 6.6 \, \mu$m.

Figure 5.15: The maximum diffraction efficiency is plotted as function of the pulse fluence for a repetition rate of 10 kHz. For fluences larger than 20 $\mu$J/cm$^2$ the diffraction efficiency decreases because of thermal effects driven by the mean intensity of 200 mW/cm$^2$. 

5.3 Correlation experiments in $\text{Sn}_2\text{P}_2\text{S}_6$

A dual axis joint Fourier transform correlator was implemented using a $\text{Sn}_2\text{P}_2\text{S}_6$ crystal as correlation material. The fast image sequence produced by the holographic storage set-up presented in section 3.4 is used as input for the correlator, whereas the second image comes from a ferroelectric liquid crystal spatial light modulator. A schematic representation of the set-up is shown in Fig. 5.17. The right upper part of the set-up in Fig. 5.17 corresponds to the holographic storage system operated with image plane holograms (see Fig. 3.7), whereas in the left lower part the two arms of the dual axis JFT correlator can be recognized. As the storage set-up was already described in detail in section 3.4, we will present only the JFT correlator part and the eventual modification of the storage set-up. The reconstructed image beam of the storage system is used as input beam for the correlator. The beam is deflected by the mirror pair 2-3 and Fourier transformed by lens 11 (focal length 250 mm). The second input beam of the correlator is derived from the original image beam of the storage system by beam splitter 1. After deflection at mirror 4 the beam is also Fourier transformed by lens 10. The paths length of the two arms are adjusted to reach a space-bandwidth (number of pixels) of 75 for each mm of lateral crystal size. At the temperature of 40°C the diffraction efficiency depends only slightly on the grating spacing in a range of $\Lambda_g = 0.8 \cdots 6.6 \, \mu$m.
in order to image the pattern of the spatial light modulator exactly overlapping on the surface of the Sn$_2$P$_2$S$_6$ crystal. In order to produce an interference pattern, the optical path length in the two arms has to be equal. This is achieved by introducing a thick glass plate (25 mm) which compensates the 10 mm thick LiNbO$_3$ crystal. The gratings written by the input beam are read out with the expanded and collimated beam of a He-Ne laser ($\lambda_R = 633$ nm and 10 mW light power). The incidence angle of the read-out beam was adjusted by the mirror pair 5-6 to match the Bragg condition of the grating written by the zero-order Fourier transform component of the input beams. The diffracted read-out beam is separated from the input beams using a dichroic beam splitter, Fourier transformed by lens 12 (focal length 200 mm) and imaged on the output plane of the correlator. The intensity distribution on the output plane can be detected by a CCD-camera or the central peak is selected by an aperture and detected with a photomultiplier. The system is operated in two steps: First the images are stored in the LiNbO$_3$ crystal. In a second step the object beam is stopped by shutter 2, and the image to be correlated with the stored images is loaded in the spatial light modulator. The correlation between the input wave from the spatial light modulator and the read-out wave from the holographic memory will build up in the output plane.
5.3. Correlation experiments in $Sn_2P_2S_6$

5.3.1 Correlation of simple objects

The correlator was first tested using two lines as amplitude modulated input images (transmission masks). The lines are 50 $\mu$m thick and 1 mm long and were positioned parallel and perpendicular to each other. The theoretical correlation pattern for this input image is shown in Fig. 5.18. In the case where the lines are placed parallel, the correlation gives also a line with maximum intensity in the center. When the lines are placed perpendicularly the correlation pattern is a square with an intensity that is 400 times smaller than the maximum height of the correlation peak in the case of the parallel arrangement. For this reason picture b) is plotted with a different grey scale than picture a). The experimental result obtained with a pulse fluence of the input beams of 320 $\mu$J/cm$^2$ and a repetition rate of 1 kHz is shown in Fig. 5.19. The sensitivity and frame rate of the CCD camera was too low to capture the response of one single pulse, therefore integration over several pulses, where the same input pattern are presented was necessary. At a repetition rate of 1 kHz cumulative effects (see section 5.2.3) are getting important increasing the light intensity on the camera. The form and shape of the correlated images fits well with the theoretical prediction of Fig. 5.18. To quantify the correlation function, a pinhole (diameter 0.5 mm) was
placed in the back focal plane of lens 12 (see Fig. 5.17) and the passing intensity was measured with the photomultiplier. The maximum amplitude of the diffraction efficiency of the correlation with two horizontal lines is $3.2 \cdot 10^{-6}$, and for two lines perpendicular to each other $4.3 \cdot 10^{-7}$. The amplitude is therefore $7.5 \times$ higher for two parallel lines than with a vertical and a horizontal line, which allows clear distinction between the two cases.

![Figure 5.18](image)

**Figure 5.18:** Theoretical intensity distribution in the output plane of the correlator for two lines with orientation a) both vertical b) perpendicular.

![Figure 5.19](image)

**Figure 5.19:** Correlation output for two thin lines: a) parallel horizontally, b) perpendicular, c) parallel vertically

### 5.3.2 Shift invariance

According to the theoretical predictions (see section 5.1.1), the output images should also shift if the input images are shifted one with respect to the other. This behavior is demonstrated in Fig. 5.20 where one of the two input patterns consisting each of a horizontal thin line is moved by a distance of 1 mm to the left or to the right. Also if the intensity of the correlation peak is slightly reduced for the non centered images, the shift invariance property of the correlator can be clearly proved.

### 5.3.3 High repetition rates

High repetition image sequences were produced by reading 10 phase modulated Escher-images (image number 0, 10, 20, ..., 90 of Appendix F) at a frame rate of 10'000 frames/s from the holographic memory and monitoring the intensity of the correlation peak, which was selected using a pinhole. The time dependence of the
5.3. Correlation experiments in Sn$_2$P$_2$S$_6$

![Image of correlation experiments](image.png)

**Figure 5.20:** Shift invariance properties of the correlation: a) Image shifted 1 mm to the left. b) Image centered in the middle. c) Image shifted 1 mm to the right.

![Graph of correlation efficiency](graph.png)

**Figure 5.21:** Correlation of 10 Escher-images (0,10,20,..90) with image number 30. The correlation was performed at a correlation rate of 10’000 frames/s.

The correlation peak is shown in Fig. 5.21. The image sequence was correlated with image number 30, which exhibited also the highest peak and can therefore be correctly identified. The height of the peaks of the non correlating image is much higher than theoretically expected. The reason is that for the used Fourier lenses 10,11 with a focal length $f = 250$ mm, the maximum extension of the Fourier transformed of the input patterns in the plane of the crystal is 8 mm. The used Sn$_2$P$_2$S$_6$ crystal however showed a maximum usable aperture of 4x4 mm$^2$. The spatial resolution is therefore reduced by a factor 2. The height of the correlation peak was in fact particularly low for input images showing high spatial frequency like image number 0 of the Escher-images (see Appendix F).

In order to increase the diffraction efficiency which was as low as $10^{-7}$ the absorption of Sn$_2$P$_2$S$_6$ was reduced to 18 cm$^{-1}$ changing the polarization of the writing beam from horizontal to vertical polarization. A resulting grating thickness of the diffraction grating of 560 $\mu$m is obtained which is still thin enough to allow the correlation of the whole image size in the input plane.
The total energy of the image sequence was 200 nJ per image, whereas the input image pulse from the SLM had a 800 nJ. This results in a switching energy of 20 pJ/pixel for the resolution of our SLM. This switching energy is a figure of merit for light driven nonlinear spatial light modulator, and our switching energy is comparable to the values found in the literature for experiment in semiconductors, multiple quantum wells, and interband operated KNbO₃[85].

Beside the required pulse energy of the hologram writing beams, also the required light power of the read-out beam has to be considered. For a diffraction efficiency of $10^{-7}$ one can show that an input power of 5 mW is require in order to detect 1000 photons in the correlation peak, in 10 $\mu$s. The same result could be achieved by the use of pulsed laser with a pulse energy as low as 50 nJ synchronized with the writing beam.

In conclusion our aim to demonstrate a high frame rate of 10'000 frames/s in a joint Fourier transform correlator based on Sn$_2$P$_2$S$_6$ crystals is therefore fulfilled. To our knowledge this is the highest correlation rate ever demonstrated in a photorefractive JFT correlator at visible wavelength.

The correlation has been clearly demonstrated with 10 simple images. The output correlation function captured with a CCD-camera agrees qualitatively with the theoretical output and also the shift invariance has been demonstrated. The spatial resolution has however to be improved with larger (at least 10x10 mm$^2$) and higher quality Sn$_2$P$_2$S$_6$ crystals.

### 5.4 Potential applications

Compared to the Vander Lugt correlation discussed in Chapter 4 the joint Fourier correlator shows a lower correlation rate, but presents the advantage to be a fully shift invariant correlator. The system compares also favorable to conventional electronics. Calculating a 1024 pixel Fast Fourier Transform (FFT) with the fastest digital signal processor (DSP) available in 1999 (see for example Texas Instrument TMS320C80 or Sharp LH9124) requires 80 $\mu$s. To calculate the 2-d FFT therefore 2 x 1024 cycles or 164 ms are required. To obtain the correlation two FFT operation are required, thus one obtains a maximal repetition rate of 3 Hz. This is more than 3 orders of magnitude slower than the Sn$_2$P$_2$S$_6$ based photorefractive JFT correlator, which is in principle independent of the number of pixel. The maximal number of pixel is however limited by the requirements imposed to the numerical aperture of the Fourier transform lenses and the Sn$_2$P$_2$S$_6$ crystal size. Note that for the DSP the time requirement for a 2-d FFT scales with $N^2 \log_2 N$ where $N$ is the number of pixels. This behavior is reported in Fig. 5.22 where optics and electronics is compared. Note that already for the relatively low resolution (256 x 256 pixels) of the SLM used in this work, the optical system is 140 time faster than the corresponding electronic system.

The shift invariance of the JFT correlator is a particularly interesting property to detect changes and movements in the input scene. Similarly to the case of the Vander Lugt type correlator the optimal encoding and preprocessing of input scene is of crucial importance for the performance of the correlator.
5.4. Potential applications

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Figure 5.22: Time required for correlation of two images with \( N_{px} \times N_{px} \) pixels, for digital calculator with 1 GHz clock rate and joint Fourier transform correlator.

5.4.1 Pattern and position recognition

Pattern recognition is a classical application for correlators. An object present in the input scene has to be identified and the position of the object (for example bacteria observed by a microscope) to be tracked. A very efficient detection of the correlation peak can be achieved by Dammann gratings\[147\] which are able to encode the peak position in a digital coordinate. In this way an object can in principle be tracked with a response time of less than 100 \( \mu s \). This however requires fast spatial light modulators and CCD-cameras which can operate at maximum repetition rates around 1000 frames/s, thus reducing the system response time to the order of 1 ms.

5.4.2 Tracking of an unknown moving object

Consider an input scene which is changing in time, take the scene at time \( t = 0 \) as one input image (reference image), and correlating with the actual image, new correlation peaks will appear in the correlator output plane in the places corresponding to the displacements of the moving objects. If the intensity distribution in the correlation plane is subtracted by the auto-correlation intensity of the reference image, the peaks due to moving objects can be enhanced. By tracking the peaks the displacement of the unknown objects can be identified. If required, the object can be additionally visualized using a novelty filter\[166\].
Chapter 6

Conclusions

In this work the potential of the photorefractive effect for optical parallel processing applications was investigated. Particular attention was devoted to optical correlators and a holographic storage system based on pulsed read-out.

The requirements to the photorefractive nonlinearity imposed by those applications are very different and can be achieved only by a proper material selection: KNbO₃, LiNbO₃ and LiTaO₃ were considered as holographic material using conventional photorefraction, whereas Sn₂P₂S₆ was chosen as fast dynamic holographic material based on pulsed interband photorefraction.

6.1 Material characterization

The theory for conventional photorefraction based on the single trap level model has been described. The standard characterization technique based on two wave mixing was discussed for different geometries and extended by photorefractive z-scan experiments. The z-scan technique is a newly developed method which allows to determine photorefractive properties for samples as small as 200 × 200 × 200 μm³. The parameters which can be determined with this technique are the dark intensity (intensity which produces a photoconductivity change equal to the dark conductivity), the ratio of photoionization constants between the scanning beam and an homogenous background illumination, and the photogalvanic field constant.

Based on these methods several KNbO₃, LiNbO₃, and LiTaO₃ crystals were investigated at a wavelength of 532 nm to be used as storage material. Among these materials KNbO₃ showed the highest sensitivity which was about 1-3 orders of magnitude higher than in LiNbO₃ and LiTaO₃. In Ce doped KNbO₃ a high sensitivity of \( S_1 = 25 \cdot 10^{-11} \text{m}^3/\text{J} \) and \( S_2 = 78 \cdot 10^{-8} \text{m}^2/\text{J} \) which are comparable to Fe doped KNbO₃ crystals has been demonstrated. Besides sensitivity also the storage time in the dark was considered. Storage times of more than 24 hours were demonstrated in Ce, Mn, and Ni doped KNbO₃, whereas much longer storage times ranging from weeks to years are expected for LiNbO₃ and LiTaO₃. Tomographic scattering measurements showed that for some particular dopants like Mn, KNbO₃ shows less scattering than LiNbO₃ or LiTaO₃. The impact of scattering is particularly important in LiNbO₃ and LiTaO₃, where combined with the strong photogalvanic effect a strong beam fanning can be observed.
Photorefractive effect based on direct band-to-band photoexcitation was theoretically considered and extended for the case of pulsed illumination. A new method allowing the determination of the photoconductivity also in samples which are much thicker than the penetration depth of the incident light has been presented. The method is based on the derivative of the photo current with respect to the light intensity and requires only one additional parameter (the linear absorption) to be known. For the case of pulsed interband photorefraction an analytical solution was derived for the case that the diffusion times of the holes and electrons are much shorter than the recombination times. The general case including the influence of traps was addressed by numerical solution of the complete (time and space dependent) equations.

Interband photorefractive effects were measured for the first time in Sn$_2$P$_2$S$_6$ crystals at 488, 514, and 532 nm. Under continuous illumination build-up times as short as 3 µs at 1 W/cm$^2$ and a diffraction efficiency of $10^{-4}$ have been observed. The material was fully characterized under pulsed illumination at a wavelength of 532 nm in order to evaluate its potential as correlator material. At this wavelength the absorption can be varied from 7 to 120 cm$^{-1}$ by changing the crystal temperature from 20 to 55°C. This allows to achieve a thickness of the holographic grating ranging from 80 to 1400 µm. The photoconductivity at 55°C was found to be $10^{-4}$ (Ω m)$^{-1}$ for a light intensity of 100 mW/cm$^2$. From the build-up of the photoconductivity the interband recombination constant $\gamma_{\text{dir}} = 8 \cdot 10^{-20}$ m$^3$/s and the sum of the electron and hole mobilities $\mu_e + \mu_h = 1.7 \cdot 10^{-4}$ cm$^2$/Vs which are the fundamental parameters for interband photorefraction were estimated and found to be strongly influenced the presence of shallow traps. Measurements of the diffraction efficiency using 50 ns pulses were performed as a function of the pulse fluence, crystal temperature, and grating spacing. The typical diffraction efficiency for a fluence of 110 µJ/cm$^2$ is around $10^{-4}$ whereas the length of the diffraction peak response varies from 3-12 µs at room temperature. For fluences larger than 10 µJ/cm$^2$, the diffraction efficiency is strongly dependent on the fluence. At high repetition rates the maximum allowed fluence is limited by the maximum mean intensity. For mean intensities higher than 100-200 mW/cm$^2$ the diffraction efficiency degrades and becomes instable. A repetition rate of 10 KHz leads to a diffraction efficiency which is 4 times smaller than at low repetition rates. The diffraction efficiency is however still large enough to allow the crystal to be operated as a correlator.

### 6.2 Holographic memory with high read-out frame rate

A holographic storage system based on LiNbO$_3$, LiTaO$_3$, or KNbO$_3$ was built. 100 images with a resolution of 256 × 256 pixels were successfully stored and retrieved in all three materials. The signal to noise of the read-out images was as high as 45, it was however not possible to recognize all pixels because the CCD-camera was not pixel-matched with the spatial light modulator. In LiNbO$_3$ and LiTaO$_3$, compared to KNbO$_3$, the diffraction efficiency of the stored hologram was found to be 14 times larger. This is due to the strong photogalvanic effect in these materials,
which is however also responsible for the self-focusing effect, which can dramatically decrease the image quality as was shown in this work. The system was not optimized for maximum storage density. This would require spatial light modulators with much higher resolution (for example 1200 × 1000 pixels) and specially designed lens systems. The aim of the system was to demonstrate high read-out frame rate. The maximum achieved frame rate of 50'000 frames/s was limited by the repetition rate of our laser. This is to our knowledge the fastest pulsed image sequence generated by a holographic storage system.

6.3 Vander Lugt correlator based on photorefractive LiNbO$_3$

The theory for a Vander Lugt type correlator based on photorefractive thick grating, which explicitly considers the shift invariance in two directions was presented. The results show that in the case a large number of matching filters have to be stored, the crystal thickness has to be increased, and the shift invariance in both transversal directions is actually lost.

The holographic memory set-up was slightly modified to be operated as Vander Lugt type correlator. Using phase modulated images a highly selective correlator was achieved. The correlator was tested with Escher-images and 256 × 256 pixels random bitmaps giving excellent results. By detecting the correlation image with a fast frame rate CCD-camera and storing 100 images in the crystal a correlation rate of 10'000 images/s was achieved. The correlation can be easily increased till 500'000 images/s (which corresponds to a throughput of 32 Gbit/s) by replacing the CCD-camera with a high speed line scan camera operating at 1000 images/s and by increasing the number of stored images to 500. Higher rates are possible by increasing the resolution of the spatial light modulators. For ferroelectric modulator with a resolution of 1280 × 1200 pixels correlation rates of 1'500'000 images/s (2 Tbit/s) should be possible.

6.4 Joint Fourier transform correlator based on Sn$_2$P$_2$S$_6$

The theory for a JFT correlator based on photorefractive thick grating was presented. The theory takes into account the grating thickness, the position error of the Fourier planes with respect to the crystal, and the influence of the read-out wavelength. The case of dual axes JFT correlator was also addressed.

A joint Fourier transform correlator was successfully built. The correlator receives the first input image from the high frame rate holographic memory and the second image from a spatial light modulator. The correlator was first tested with simple images, showing a qualitative agreement with the theoretical correlation image and a confirmation of the shift invariance of the correlator. Correlation at 10'000 frame/s has been demonstrated with a total of 10 phase modulated Escher-images. This is to our knowledge the highest correlation rate demonstrated in a
photorefractive JFT correlator. The selectivity of the correlator was reduced by the limited size and quality of the available crystal. Highly selective correlation should be possible with a 10 mm$^2$ high quality Sn$_2$P$_2$S$_6$ crystal.

### 6.5 Material selection

An important part of this work was devoted to the application oriented investigation of different materials. The investigated materials present very different characteristics which make them suitable one over the other depending on the targeted application. In the following section we will give a short summary of the most important photorefractive properties and the application fields of the investigated materials at visible wavelength.

#### 6.5.1 Storage materials

For storage applications long storage times are required. Storage times ranging from some weeks till years are observed in LiNbO$_3$ and LiTaO$_3$. In this materials the photorefractive sensitivity is typically 3-4 orders of magnitude lower than in the best photorefractive materials. Low sensitivity can be an advantage in holographic storage because it allows to read the memory for many times only slightly erasing it. Another characteristic which makes LiNbO$_3$ and LiTaO$_3$ attractive for holographic storage is the strong asymmetry (5-10 times) between the writing time and the erasing time produced by the photogalvanic effect. LiNbO$_3$ most notably can be easily grown in large size, quantities, and good optical quality.

#### 6.5.2 Real-time holographic materials

For many simple applications like laser beam clean-up, novelty filters, holographic interferometry, dynamic Bragg-reflectors, and light induced waveguides, response times in the order of some ms are fast enough. A very attractive material for this kind of applications is KNbO$_3$. Especially cerium doped KNbO$_3$ crystals exhibit a very high sensitivity comparable to the best photorefractive materials. In addition these crystals exhibit a very low dark conductivity which allows to realize the applications cited above at sub-$\mu$W laser power.

#### 6.5.3 Fast photorefractive effects

The photorefractive effect can be made fast (in the order of some $\mu$s) increasing the efficiency of photoexcitation process. This can be achieved by interband photorefraction. A material which presents interband photorefraction at visible wavelength is Sn$_2$P$_2$S$_6$. In interband photorefraction, due to the strong absorption, thin gratings are obtained. Thin gratings are particularly interesting for building optical correlators or incoherent-to-coherent optical converters. At a wavelength of 532 nm, which can be produced by compact diode pumped lasers, Sn$_2$P$_2$S$_6$ presents the interesting properties that the thickness of the photorefractive grating can be changed in a wide range by tuning the crystal temperature.
Appendix A

Beam propagation in photorefractive materials

The photorefractive effect not only allows to transfer energy or produce phase changes between multiple beams, but also acts on a single beam itself. The effect can be exploited to produce light induced dynamical lenses or light induced waveguides. The later called spatial solitons are of particular interest from a theoretical point of view, because they allow to observe beams that are not changing their shape during propagation. The effect is not always desirable, if the beam carries information like in holographic storage the self lensing effect can produce a strong distortion of the images. In the following section we present first a complete theory for beam propagation in photorefractive material in the case of one-transverse-dimensional beams. In section A.2 the theory is applied to the case of photorefractive spatial soliton. The section terminates with what we claim to be the first experimental observation of spatial soliton in KNbO₃ crystals.

A.1 Theory of beam propagation in photorefractive media

In the paraxial approximation, under the influence of the photorefractive nonlinearity, the propagation of an optical beam which is invariant in the transversal y direction is governed by the equation

\[
i \frac{\partial}{\partial x} A(x, z) + \frac{1}{2k_0n} \frac{\partial^2}{\partial z^2} A(x, z) - \frac{k_0}{2} \text{r}^{\text{eff}} \tilde{E}_0 \nu(x, z) A(x, z) = 0, \quad (A.1)
\]

where \(x\) is the beam propagation coordinate, \(k_0 = 2\pi/\lambda\) is the wavenumber of the wave with the wavelength \(\lambda\) in vacuum, \(n\) is the refractive index of the medium, and \(A(x, z)\) is the complex scalar amplitude of the optical wave. The last term in Eq. (A.1) describes the optical nonlinearity due to the photorefractive effect, with \(r^{\text{eff}}\) being the effective electro-optic coefficient relevant to the experimental geometry[60]. Finally, \(\nu\) is the normalized light-induced space-charge electric field.
\( E_{sc} \) defined by the relation

\[
\nu(z) \equiv \frac{e}{k_B T k_D} E_{sc}(z) \equiv \frac{E_{sc}}{E}. \tag{A.2}
\]

Here \( k_D \equiv (e^2 N^*/\varepsilon_{eff} \varepsilon_0 k_T) \) is the Debye wave-vector, \( e \) the elementary charge, \( \varepsilon_0 \) the permittivity of vacuum, \( \varepsilon \) the relative dielectric constant, \( k_B \) the Boltzmann constant, and \( N^*_\text{eff} \) the effective density of traps defined below. In the framework of the Kukhtarev-Vinetskii model \([63]\) with a single impurity level and charge transport in the conduction band alone the equation for the normalized space-charge field \( \nu(x, z) \) has the form

\[
(n_0 + \kappa \nu_{ph}) \frac{I_D}{I + I_D} - \psi \nu_{ph} + I + I_D - \frac{\psi}{\chi} + \frac{1}{\chi^2 k_D^2} \frac{\partial \nu}{\partial z} - \frac{\psi}{\chi} \frac{\partial I / \partial z}{I + I_D} = 0. \tag{A.3}
\]

where

\[
\chi(z) \equiv 1 + \frac{1}{k_D \xi_0} \frac{\partial \nu}{\partial z}, \tag{A.4}
\]

and

\[
\psi(z) \equiv \xi_0 + (1 - \xi_0) \chi(z), \tag{A.5}
\]

Here \( I(x, z) \propto |A(x, z)|^2 \) is the intensity of light, and \( I_D \) is the dark intensity. In general, \( I_D \) is proportional to the spatially homogeneous component of the number density of carriers that make a transition from a non-mobile to a mobile state. It can be written as \( I_D \equiv I_{D,em} + I_{D,th} \), where \( I_{D,th} \) is the equivalent intensity corresponding to the number of thermal transitions, and \( I_{D,em} \) is the equivalent intensity of a homogeneous background illumination. The parameter \( \kappa \) describes the fraction of this electromagnetic illumination as

\[
\kappa \equiv I_{D,em} / I_D. \tag{A.6}
\]

The reduction state of the crystal is considered explicitly and is expressed by the parameter

\[
\xi_0 \equiv \frac{N_D^+}{N_{\text{eff}}^*} \approx \frac{N_A}{N_{\text{eff}}^*}, \tag{A.7}
\]

where the effective density of traps is defined as \( N_{\text{eff}}^* \equiv N_D^+(N_D - N_D^+)/N_D \), the quantity \( N_D \) is the total density of donors, and \( N_D^+ \approx N_A \) is the density of ionized donors in the dark (acceptors). The optimum reduction state of the crystal corresponds to \( N_D^+ = N_D - N_D^+ \) and \( \xi_0 = 2 \).

Eq. (A.3) also explicitly contains terms describing the photogalvanic effect. The normalized photogalvanic field is defined as

\[
\nu_{ph} \equiv \frac{E_{ph}}{E} \equiv \frac{1}{E} \left( \xi_0 N_{\text{eff}}^* \gamma_L I_{ph} \right) / \mu_e, \tag{A.8}
\]
A.1. Theory of beam propagation in photorefractive media

where \( \gamma_e, \mu_e, \) and \( I_{ph} \) are defined in section 2.2. As defined here, \( E_{ph} \) is the field that would have to be applied in order to measure a voltage-induced photocurrent equal to the photogalvanic current induced by \( I_{D,em} \). For most photorefractive materials such as LiNbO\(_3\) and KNbO\(_3\), the photogalvanic current measured along the positive c-axis of the crystal is negative, thus \( \nu_{ph} \) is also negative. The final quantity to be defined in Eq. (A.3) is \( \nu_0 \equiv E_0/E \), i.e., the applied electric field \( E_0 \) with the usual normalization. In the experimental practice, the applied field \( E_0 \) seen in the bulk of the crystal may differ from the externally applied field \( E_{ext} \) \([167]\) due to charge screening effects, which can be accounted for by introducing a screening factor \( \xi \) defined as \( \xi \equiv E_0/E_{ext} \).

Note, that the terms proportional to \( \partial \nu/\partial z \) in the expressions (A.4) and (A.5) ensure that Eq. (A.3) remains valid even when the lateral spatial extent of the propagating beam \( W \) is comparable to the Debye screening length \( 2\pi/k_D \), which is usually of the order of 1 \( \mu m \) or less in most photorefractive samples.

In most previously published literature the assumption \( N_A \ll N_D \) \((N_A \approx N_{eff}^* \), or \( \xi_0 = 1 \)) is made. This approximation gives \( \chi(z) = 1 + (\partial \nu/\partial z)/k_D \equiv \tilde{\chi}(z) \) (Eq. (A.4)), \( \psi(z) = 1 \) (Eq. (A.5)), and transforms Eq. (A.3) to the form given for instance in Ref.\([\text{73}]\]

\[
(v_0 + \nu_{ph}) \tilde{\chi}^2 \frac{I_D}{I + I_D} - \tilde{\chi}^2 \nu_{ph} - \tilde{\chi} \nu + \frac{1}{k_D^2} \frac{\partial^2 \nu}{\partial z^2} - \frac{\tilde{\chi}}{k_D} \frac{\partial I/\partial z}{I + I_D} = 0. \tag{A.9}
\]

Note that Eq. (A.9) is no longer dependent on \( \kappa \).

The opposite case is the one for which \( N_A \approx N_D \gg (N_D - N_A) \approx N_{eff}^* \). Then \( \xi_0 \gg 1, \chi(z) = 1 \) and \( \psi(z) = \tilde{\chi}(z) \), so that Eq. (A.3) reads

\[
(v_0 + \nu_{ph}) \frac{I_D}{I + I_D} - \tilde{\chi} \nu_{ph} + \frac{1}{k_D^2} \frac{\partial^2 \nu}{\partial z^2} - \frac{\tilde{\chi}}{k_D} \frac{\partial I/\partial z}{I + I_D} = 0. \tag{A.10}
\]

A comparison of Eqs. (A.9) and (A.10) shows that the role of the correction quantity \( \tilde{\chi}(z) \) is slightly different in the two limiting cases. As \( 1 \leq \xi_0 \leq \infty \) one has always \( \chi \leq \tilde{\chi} \). Therefore, if the beam width \( W \) largely exceeds the Debye length \( 2\pi/k_D \), the terms due to charge diffusion (the last two terms in Eq. (A.3)) can be neglected and Eq. (A.3) reduces to the simplified form \([168, 169, 81]\)

\[
(v_0 + \nu_{ph}) \frac{I_D}{I + I_D} - \nu_{ph} - \nu = 0 \tag{A.11}
\]

for all values of the reduction state parameter \( \xi_0 \) and of the dark intensity factor \( \kappa \).

Eqs. (A.1) and (A.3) give a complete description of beam propagation in photorefractive material. They can be solved by numerical methods. A special class of solutions which maintains their spatial profile during propagation (spatial solitons) is discussed in the next section, whereas the influence of photorefractive self-focusing on beams containing modulated images was discussed already in section 3.2.3.
A.2 Photorefractive self-focusing and spatial solitons

A spatial solitary wave \([170]\) is a beam that maintains its shape while propagating in a nonlinear optical medium. Natural diffraction of the beam is exactly counterbalanced by optical nonlinearity. Steady-state self-focusing of light beams and formation of spatial solitary waves (in the following referred to as spatial solitons) produced by means of the photorefractive effect \([19]\) have witnessed some considerable interest in the last few years due to the low light intensities that are required to reach the nonlinearity necessary for beam trapping \([73, 171, 172, 168, 169, 173, 80, 174, 175, 176, 81, 177, 178, 179, 180, 181]\). While a complete zoology of names has been created to distinguish between slightly different cases, the basic physical mechanism underlying photorefractive self-focusing and the formation of photorefractive solitons is conceptually very simple. It is based on the local screening of the photocurrent produced either by an external applied field or the photogalvanic effect \([19]\). In both cases the screening is achieved by the field induced by space-charges redistributing during the soliton formation and getting trapped at the borders of the beam \([73]\).

A full two-transverse-dimensional \((2+1)D\) theory of photorefractive self-focusing and spatial soliton formation that has been developed in Ref.\([73]\) and Refs.\([171]\) -\([172]\), respectively, is rather involved due to the inherently anisotropic and nonlocal nature of photorefractive nonlinearity. Fortunately many salient features of this formation can be investigated in the case of one-transverse-dimensional \((1+1)D\) stripe beams \([168, 169]\).

In the following sections we investigate theoretically and experimentally the formation of a spatial soliton starting from a Gaussian beam. Particular emphasis is posed on the spatial transients (changes of waveguide width) occurring during the soliton formation. The spatial transient are affecting the phase velocity of the self-guided beams, which is of critical importance if the light induced waveguide has to be used for nonlinear optics. This could be particularly interesting in Knbo3 which exhibits large second order nonlinear optical coefficients, and in addition, due to its large birefringence, is highly phase matchable for frequency conversion applications \([182]\).

A.2.1 Transition from Gaussian beams to solitons

In this section we study the propagation of 1D optical beams and the transition from a Gaussian input beam to a soliton in nonlinear photorefractive materials using the set of equations presented in section A.1. While the optimum condition for launching a spatial soliton would be to produce an input intensity profile already equal to the soliton profile, experimentally it is much easier to use a Gaussian input beam. This input beam will have to reshape to the soliton profile, and this process will require a certain propagation distance within the crystal.

The analysis is carried out by numerical solution of Eqs. (A.1) and (A.9) for the
input boundary conditions corresponding to a Gaussian beam

\[ A_{\text{Gauss}}(x=0,z) = \frac{I_0^{1/2}}{(1 + i2 \ln(2) l_w \lambda / \pi d^2)^{1/2}} \exp \left[ -\frac{2 \ln(2) z^2}{d^2 (1 + i2 \ln(2) l_w \lambda / \pi d^2)} \right] \]  

(A.12)

where \( d \) is the full width at half maximum (FWHM) diameter, \( I_0 \) is the maximum intensity in the beam waist, and \( l_w \) is the position of the waist with respect to the input crystal face \( x = 0 \). Positive values of \( l_w \) correspond to the beam converging at the input face of the crystal (the waist lies inside) and vice versa.

Eq. (A.1) is solved using a Crank-Nicholson-type (CRN) scheme [183]. The equation for the space-charge grating (A.9) is solved iteratively using a CRN-type routine as well. All calculations presented here correspond to material parameters of photorefractive crystals of K\(\text{Nb}_3\)O\(_5\) in a geometry for which the \( r_{333} \) electro-optic coefficient is active, with \( n = 2.227 \), \( r_{\text{eff}} = 56 \ \text{pm/V} \), \( k_D = 4 \ \mu\text{m}^{-1} \), \( \lambda = 488 \ \text{nm} \), an electric field of \( E_{\text{tot}} = E_0 + E_{\text{ph}} = 2.6 - 0.6 \ \text{kV/cm} = 2.0 \ \text{kV/cm} \), and a maximum propagation length of 100 mm. If the width of the beam is much larger than the inverse Debye wavenumber \( (W \gg 2\pi/k_D) \), the results of the simulations can be applied also to other materials, wavelengths, or electric fields by simple rescaling of the beam width \( W \) and the propagation distance \( x \) according to the relations

\[
\frac{W_{\text{mat}}}{W_{\text{KN}}} = \left[ \frac{n^2}{\lambda} \sqrt{r_{\text{eff}} E_{\text{tot}}} \right]_{\text{KN}} / \left[ \frac{n^2}{\lambda} \sqrt{r_{\text{eff}} E_{\text{tot}}} \right]_{\text{mat}} = 3.40 \times 10^4 \ \text{m}^{-1} / \left[ \frac{n^2}{\lambda} \sqrt{r_{\text{eff}} E_{\text{tot}}} \right]_{\text{mat}},
\]

(A.13)

and

\[
\frac{x_{\text{mat}}}{x_{\text{KN}}} = \left[ \frac{1}{\lambda} n^3 r_{\text{eff}} E_{\text{tot}} \right]_{\text{KN}} / \left[ \frac{1}{\lambda} n^3 r_{\text{eff}} E_{\text{tot}} \right]_{\text{mat}} = 253 \ \text{m}^{-1} / \left[ \frac{1}{\lambda} n^3 r_{\text{eff}} E_{\text{tot}} \right]_{\text{mat}}
\]

(A.14)

where the subscripts “KN” and “mat” refer to K\(\text{Nb}_3\)O\(_5\) and some other material, respectively. In SBN, for example, with the same applied field, we obtain a corresponding propagation length of 29 mm instead of 100 mm.

Figure A.1 shows typical evolution of an input Gaussian beam inside the photorefractive medium and its convergence to a soliton solution at large propagation distances. The parameters of the run are \( d = 18.5 \ \mu\text{m} \), \( I_0/I_D = 1 \), and \( l_w = 0 \). For the material parameters of the K\(\text{Nb}_3\)O\(_5\) listed above, a stripe soliton with the peak intensity \( I = I_D \) has exactly the same FWHM diameter \( W_{\text{tot}} = 18.5 \ \mu\text{m} \) (see, e.g., Ref. [81]). Figure A.1 demonstrates that even though the intensity difference between the input Gaussian beam and the soliton never exceeds 2% of the maximum intensity, reshaping produces beam width variations of more than 10% along propagation. As expected, stronger reshaping occurs in the initial part of propagation. The beam bending due to the out-of-phase component of the space-charge field caused by charge diffusion can be also clearly recognized in Fig. A.1 by the position of the intensity maxima. In Fig. A.2 we show the beam width (FWHM) \( W(x) \) and the maximum intensity \( I_{\text{max}}(x)/I_D \) as a function of \( x \) for the simulation presented in Fig. A.1. We notice that the Gaussian beam with maximum intensity \( I_0/I_D = 1 \) and the 18.5 \( \mu\text{m} \) width converges towards a soliton with slightly different
Figure A.1: Numerical calculation of spatial soliton formation in a photorefractive KNbO₃ crystal. A Gaussian input beam \( (d = 18.5 \ \mu \text{m}, I_0/I_D = 1) \) is launched at \( z=0 \), the total electric field is \( E_{\text{tot}} = E_0 + E_{\text{ph}} = (2.6 - 0.6) \ \text{kV/cm} \). The intensity profile \( I(z) \) is plotted after different propagation distances \( x = 0, 3, 11, 50, \) and \( 100 \ \text{mm} \). The beam profile at the input \( (x = 0) \) and after \( 100 \ \text{mm} \) are very similar, however strong reshaping is observed in the first \( 10 \ \text{mm} \) of propagation.

The peak intensity of \( \hat{I}/I_D = 0.94 \) and width \( W_{\text{sol}} = 18.7 \ \mu \text{m} \), which exceeds slightly the value expected for \( \hat{I}/I_D = 1 \). Except for the first \( 10 \ \text{mm} \), the beam width evolution can be well described by a damped oscillation

\[
W(x) = W_{\text{sol}} + \Delta W \sin(2\pi x/\Lambda + \phi_0) \exp(-x/L) \quad (A.15)
\]

around the final soliton width \( W_{\text{sol}} \). In the following, the amplitude \( \Delta W \), the spatial period \( \Lambda \), and the spatial damping length \( L \) of the damped oscillation will be investigated as a function of the incoming beam width, the waist position \( l_w \) along the \( x \)-axis, and the maximum intensity of the resulting soliton.

We first investigate how the input beam width influences the convergence to a soliton. Since a soliton solution is fully characterized by its power, we keep the power

\[
P_{\text{Gauss}} = \int_{-\infty}^{+\infty} |A_{\text{Gauss}}|^2 dz = I_0 d \sqrt{\frac{\pi}{\ln(2)}} \quad (A.16)
\]

of the input Gaussian beam constant while changing its width \( d \). Under the assumption that only a small part of the optical power is lost by radiative scattering of light, we expect to obtain the same soliton width at steady state, independent of the chosen width of the input Gaussian. In Fig. A.3 we show the results of the
simulation of the beam width as a function of the propagation distance $x$. The input width $d$ varied between 11 and 25 $\mu$m. The constant input power was taken to be equal to that used for Figs. (A.1) and (A.2). The previously described damped oscillation behavior can be observed for the whole range of $d$. Note that the amplitude of the oscillation is minimized for an input beam width around 21 $\mu$m, which is approximately 12% larger than the formed soliton width. The width after 100 mm is nearly constant for a wide range of $d$ and approaches $W_{\text{sol}}$, confirming that the soliton solution is working as an attractor for the input beam with the appropriate power. A more detailed analysis shows that the asymptotic soliton profile varies only between 18.9 $\mu$m and 18.7 $\mu$m for $15\mu$m $\leq d \leq 25\mu$m. To describe the spatial stability of the beam towards the end of the propagation region, one may use the semi-heuristic quantity $\Delta W_{70-100}$ giving the average amplitude of the damped oscillations in the last 30 mm. We find that $\Delta W_{70-100}$ always lies between 0.08 $\mu$m for $d = 21\mu$m and 0.6 $\mu$m for $d = 15\mu$m, showing again that the former value of $d$ (12% above $W_{\text{sol}}$) gives the optimum launching conditions for a spatial soliton of intensity $I/I_D \approx 1$.

Second we investigate how the soliton formation is influenced by the position of the crystal surface with respect to the waist of the Gaussian input beam. The simulations of Fig. A.3 were repeated, changing the position of the waist $l_w$ from -1 to 1 mm with respect to the entrance face at $x = 0$.

The calculations show that the oscillations present a minimum for the waist located exactly on the entrance face and $d = 21 \mu$m as above. The spatial transients
are strongly influenced by the waist position. For a waist located 300 μm from the crystal and an input beam width of 21 μm, we obtain a doubled amplitude of the spatial oscillations with respect to the optimum conditions. Experimentally a waist positioning error < 100 μm is easily achieved by appropriate techniques (see experimental section). The waist position will therefore be held at the entrance face in all following investigations.

Up to now we have considered only solitons with a normalized intensity very close to $\hat{I}/I_D = 1$. The soliton formation strongly depends on $\hat{I}/I_D$. For small intensities $\hat{I}/I_D \ll 1$, Eqs. (A.1) and (A.11) lead to a nonlinear Schrödinger equation with a Kerr-type nonlinearity proportional to the light intensity. Therefore the refractive index change for $\hat{I}/I_D \ll 1$ is much smaller than for $\hat{I}/I_D = 1$. On the other hand, for $\hat{I}/I_D \gg 1$ the nonlinearity is strongly saturated, thus producing only small refractive index changes in the central part of the beam. In both limiting cases the focusing effect is weaker than for $\hat{I}/I_D \approx 1$, suggesting that a longer propagation distance is needed in order to generate a soliton [73, 171, 172]. We extended the simulations presented in Fig. A.3 for $\hat{I}/I_D$ ranging from 0.1 to 50. The simulations are analyzed considering the evolution of the FWHM $W(x)$ along the propagation path as before. In the intensity range from $\hat{I}/I_D = 0.5$ to 20, the width is found to follow the damped oscillations of Eq. (A.15) after the first 10 mm of propagation.
The damping length $L$, the oscillation period $\Lambda$, and the normalized amplitude of deviation from the soliton width $\Delta W_{\text{rel}} \equiv \Delta W_{70-100}/W_{\text{sol}}$ in the propagation region between 70 and 100 mm are reported in Fig. A.4 as a function of $I/I_D$. The curves in Fig. A.4 were obtained as follows: for each $I/I_D$, runs were made using different input widths $d$, and only the values $L$, $\Lambda$ and $\Delta W_{\text{rel}}$ corresponding to the optimum value of $d$ (minimum oscillations) have been plotted in the figure. The corresponding width $d$ is also shown in Fig. A.1 as a function of $I/I_D$ in the form of the relative quantity $d_{\text{rel}} = d/W_{\text{sol}}$ (top horizontal axis scale). The strongest damping of the oscillation is found for $I/I_D = 2$ with a damping length of $L = 34$ mm. The shortest period of the spatial oscillation is found for $I/I_D = 5$ with $\Lambda = 15$ mm. The smallest deviation from a soliton $\Delta W_{\text{rel}} = 0.2\%$ is found for $I/I_D = 1$. It should be noted that in view of applications such as phase matched frequency conversion, the best conditions are those for which $\Delta W_{\text{max}}$ [defined as $\Delta W_{\text{max}} \equiv \max(|W(x) - W_{\text{sol}}|)$] and $L$ are as small as possible. Furthermore, the ratio $L/\Lambda$ should be small.

For $I/I_D > 20$, the beam width evolution is no longer approximated by the damped oscillation of Eq. (A.15) but shows a more complicated, undamped, periodical shape. For small normalized intensities $I/I_D < 0.2$, the damping length $L$ and
Appendix A. Beam propagation in photorefractive materials

the period Λ are much longer than the simulated propagation length of 100 mm and can be determined only by considering longer propagation lengths. For experimental soliton observation this means that even though soliton solutions exist for a wide range of $I/I_D$, the physical length of the available crystals limits their observation to the range $I/I_D = 0.2 \ldots 20$. This range does not depend on the chosen value for the applied field.

As their effect can be important, we look more closely at the amplitude of the oscillations. In Fig. A.5 a) we report the normalized amplitude $\Delta W_{70-100}/W_{\text{sol}}$ of the oscillations in the last 30 mm of propagation. The amplitudes are plotted as function of relative width $d_{\text{rel}}$ of the Gaussian input beam with respect to the soliton. The curves present the normalized intensity $I/I_D$ as parameter. For all values of $I/I_D$, the amplitude $\Delta W_{70-100}/W_{\text{sol}}$ presents a minimum at a particular relative input width

![Figure A.5: a) Mean amplitude of the normalized beam width oscillations $\Delta W_{70-100}/W_{\text{sol}}$ in the last 30 mm of propagation as a function of the relative width $d_{\text{rel}}$ of the input wave. b) Normalized maximum deviation from the soliton profile $\Delta W_{\text{max}}/W_{\text{sol}}$ throughout the whole simulation region of 100 mm as a function of $d_{\text{rel}}$. In a) the smallest amplitude $\Delta W_{70-100}/W_{\text{sol}} = 0.2\%$ is obtained for $I/I_D = 1$ and an input beam 12% wider than the formed soliton. In b) the smallest maximum deviation $\Delta W_{\text{max}}/W_{\text{sol}} = 3\%$ is found for $I/I_D = 5$ and an input beam 1% narrower than the formed soliton. The horizontal line in b) shows the upper limit above which a self-focused beam cannot be considered as an (experimental) soliton according to the definition in the text.](image-url)
The smallest amplitude $\Delta W_{70-100}/W_{\text{sol}} = 0.2\%$ is obtained for $I/I_D = 1$ and $d_{\text{rel}} = 1.12$. The input beam width that minimizes $\Delta W_{70-100}/W_{\text{sol}}$ is smaller than the soliton width for $I/I_D > 3$ and larger for $I/I_D < 3$. The optimal beam width $d_{\text{rel}}$ is shown as a function of $I/I_D$ in Fig. A.4. An alternative way to analyze the changes in the beam width along propagation is to consider the maximum difference $\Delta W_{\text{max}}$ (defined above) between the propagating beam and the formed soliton. As already mentioned, it is important to keep $\Delta W_{\text{max}}$ small when the generated self-trapped beam is used as a waveguide and a constant group velocity is required. The normalized quantity $\Delta W_{\text{max}}/W_{\text{sol}}$ is plotted in Fig. A.5 b) as a function of the relative width $d_{\text{rel}}$ and for different $I/I_D$. Similarly to Fig. A.5 a) one finds a minimum for all values of $I/I_D$. The smallest deviation is $\Delta W_{\text{max}} \approx 0.03 W_{\text{sol}}$ for $I/I_D = 5$ and $d_{\text{rel}} = 0.99$. This can be explained by the fact that, for $I/I_D = 5$, the profile of the soliton closely resembles a Gaussian (see for instance Refs. [80] and [81]).

To summarize, if one aims at the best matching of the output beam and the soliton profile (minimum oscillations close to the output face of the crystal), the optimum parameters for soliton launching are $I/I_D = 1$ and a Gaussian input beam $\approx 12\%$ wider than the soliton width. If, on the other hand, the criterion is to limit the maximum width discrepancy with respect to the soliton along the whole propagation path, then the optimum conditions are $I/I_D \approx 5$ with a Gaussian input beam width $d$ equal to the soliton width.

### A.2.2 Self-trapping and soliton experiments in KNbO$_3$

The set-up used for spatial soliton observation was the following: The spatially filtered beam at 488 nm of an Ar$^+$-ion laser is focused by means of a cylindrical lens on the surface of the photorefractive KNbO$_3$ crystal. Three different cylindrical lenses with focal lengths $F = 60$, 100, 150 mm were used, producing the FWHM beam waist diameters of 22, 36.5, and 54.6 $\mu$m, respectively. The used photorefractive KNbO$_3$ crystal was nominally undoped with the dimensions $a \times b \times c = 5.22 \text{ mm} \times 11.35 \text{ mm} \times 3.80 \text{ mm}$. The soliton was propagated in the direction of the $b$-axis, and the $c$-axis was chosen parallel to the focusing direction of the cylindrical lens ($z$-axis). The beam polarization and the applied electric field $E_{\text{ext}}$ were also along the $c$-axis.

After exiting the KNbO$_3$ crystal, the beam was imaged on a CCD camera using a microscope lens. The background conductivity was changed using an additional inhomogeneous illumination $I_{D,\text{em}}$. This light was generated from the same laser and was made incoherent to the signal beam by rotating its polarization parallel to the $a$-axis. For the presented geometry, the refractive index $n = 2.227$ and the effective electro-optic coefficient $r_{\text{eff}} = 56 \text{ pm/V}$ seen by the signal beam are the same as used for the simulations in the previous section.

We first investigated the self-focusing effect for the fixed ratio $I_0/I_D = 1$ and the input beam diameter $d = 54.6 \mu$m while changing the external field in the range $E_{\text{ext}} = 0 \ldots 6.2 \text{ kV/cm}$. The resulting intensity profiles detected with the CCD camera are presented in Fig. A.6. A strong self-focusing effect is observed. The output beam width decreases from $W = 78 \mu$m without applied field to $W = 10 \mu$m for $E_{\text{ext}} = 6.2 \text{ kV/cm}$. In addition, a smoothing effect of the beam profile for higher
Figure A.6: Experimental observation of self-focusing in KNbO₃. The output intensity profile is plotted for different applied fields $E_{\text{ext}}$ (input width $d = 54.6 \ \mu\text{m}$, intensity ratio $I_0/I_D = 1$, crystal length $= 11.35 \ \text{mm}$). A strong self-focusing effect and a smoothing of the intensity profile can be observed for large $E_{\text{ext}}$. The output width $W$ is plotted as function of the applied electric field $E_{\text{ext}}$ in Fig. A.7. Data for two different input beam diameters are shown. In the same graph we also show the numerically calculated predictions as a function of the bulk field $E_{\text{tot}} \equiv E_0 + E_{\text{ph}}$. The screening factor $\xi = 0.5 \pm 0.1$ and a photovoltaic field $E_{\text{ph}} = (-0.6 \pm 0.2) \ \text{kV/cm}$ are determined through a fit of the experimental results and numerics. The value for $\xi$ is consistent with independent interferometric electro-optic measurements performed in the same set-up. Note that for the sample investigated here, even for the largest fields, no formation of stripes due to transverse modulation instability previously reported in Refs. [178] for SBN could be observed. However, two additional KNbO₃ samples showed such effects for $E_{\text{ext}} \geq 4 \ \text{kV/cm}$ and $I_{D,\text{em}} \geq 25 \ \text{mW/cm}^2$, probably because of a larger level of scattering noise.

Figure A.8 shows the dependence of the output beam profile on the intensity ratio $I_0/I_D$ for the fixed applied field $E_{\text{ext}} = 5.3 \ \text{kV/cm}$. The input beam width was $d = 36.5 \ \mu\text{m}$. Different ratios $I_0/I_D$ are produced by changing the background intensity $I_{D,\text{em}}$. The strongest focusing effect can be observed for $I_0/I_D \approx 2$. This value is very close to the one for which a bright spatial soliton has minimum width [168, 68, 80, 81].

Finally, in Fig. A.9 we show the output width $W$ of the self-focused beam as a function of $I_0/I_D$ for different values of the externally applied field $E_{\text{ext}}$. For each value of $E_{\text{ext}}$, the width $W$ goes through a minimum for $I_0/I_D \approx 1 - 3$ and increases again for larger intensities. In the same plot we also show the numerical simulations for the same input width $d = 36.5 \ \mu\text{m}$. The simulations and experiments are in
A.2. Photorefractive self-focusing and spatial solitons

Figure A.7: Output width $W$ of the self-focused beam as a function of the external field $E_{\text{ext}}$ (upper abscissa). Full circles: input width $d = 54.6 \, \mu m$, open circles: $d = 36.5 \, \mu m$. The solid and dashed lines show the corresponding numerical simulations of $W$ as a function of the total bulk field $E_{\text{tot}} \equiv E_0 + E_{\text{ph}}$ (lower abscissa). The best fit for the relationship between the two x-axes delivers a screening factor $\xi = 0.5 \pm 0.1$ and a photovoltaic field $E_{\text{ph}} = (-0.6 \pm 0.2) \, kV/cm$.

good qualitative agreement. The difference between experiment and theory can be understood considering the uncertainty of the ratio $I_0/I_D$ and $E_0$ produced by the intensity dependent screening effect [167].

Applying the results of the previous section to the measurement of Fig. A.9 allows us to put forward a practical criterion distinguishing the soliton-like beam propagation from a more general self-focusing case. A soliton is a mathematical concept requiring an exactly unchanged profile of the beam along propagation. In practice, however, due to small natural inhomogeneities (e.g., of the crystal refractive index) and finite experimental accuracy, it is difficult to ascertain observation of an exact soliton. Therefore we need a somehow more relaxed definition of a soliton that can account for the precision of common experimental apparatus. In our case, we consider the experimental propagating beam as a bright spatial soliton if (a) the beam width converges to a constant for a very long crystal, and (b) the maximum relative deviation $\Delta W_{\text{rel}}$ of the beam width from the width at infinity never exceeds 10% in any position in the sample.

The first condition (a) limits the range of valid intensity-to-dark-intensity ratios $\hat{I}/I_D$ approximately to the range $0.5 \ldots 20$, as discussed above. Although in general
Appendix A. Beam propagation in photorefractive materials

The intensity ratio for the soliton $I/I_D$ differs slightly from the one for the input Gaussian beam $I_0/I_D$, this difference is usually small close to the soliton regime, and the above condition can also be written as $I_0/I_D \approx 0.5\ldots20$. Requirement (b) contains the necessary condition that the width of the outcoming beam has to be within $\pm10\%$ of the input diameter $d$. The two above necessary conditions are shown by the shaded area in Fig. A.9. The parameter region for the existence of solitons (according to our above definition) can be restricted further by considering the calculations presented in Fig. A.5 b). The above condition (b) puts additional constraints on the input diameter $d_{\text{rot}}$ relative to the asymptotic soliton. For each value of $I/I_D$ the valid range of $d_{\text{rot}}$ can be extracted from Fig. A.5 b). Using this procedure, we can identify the circles in Fig. A.9 as representing solitons as defined by our above definition. It should be noted that in Fig. A.9 the two triangle for $I/I_D = 0.1$ and $I/I_D = 50$ do not represent solitons even though their output width corresponds to their input width. Using the same kind of procedure as above, we have identified beams that satisfy the conditions for a soliton also using other sets of parameters, as for instance with an input diameter $d = 22 \mu$m, an external field $E_{\text{ext}} = 5.3 \text{ kV/cm}$, and a ratio $I_0/I_D = 10$. All the measurements are consistent with our calculations, confirming their validity and usefulness for the interpretation of the self-focusing and soliton experiments.

In conclusion we have presented a numerical analysis and experimental demonstration of photorefractive one dimensional spatial solitons in KNbO$_3$. The numerical simulations clearly show that the transition from an input Gaussian beam to a soliton occurs through variations of the beam width that can be described mathematically by damped oscillations. The best condition for launching a beam that is as close as possible to a soliton is found for $I/I_D \approx 5$, in which case the width oscillation amplitude never exceeds $3\%$ of the asymptotic soliton width. Our self-trapping
Figure A.9: Spatial soliton as a special case of self-focusing. The experimental width of the outcoming beam is plotted as function of the intensity ratios $I_0/I_D$ for different applied fields $E_{\text{ext}}$ (input beam $d = 36.5 \, \mu\text{m}$). The measurements are denoted by points (squares: $E_{\text{ext}} = 2.6 \, \text{kV/cm}$, stars: $E_{\text{ext}} = 3.9 \, \text{kV/cm}$, triangles: $E_{\text{ext}} = 5.3 \, \text{kV/cm}$). Numerical predictions for the output widths are also shown, with $\xi = 0.5$ and $E_{\text{ph}} = -0.6 \, \text{kV/cm}$ (Fig. A.7) they correspond to: $E_{\text{ext}} = 2.3 \, \text{kV/cm}$ (solid line), $E_{\text{ext}} = 3.3 \, \text{kV/cm}$ (dash-dotted line), $E_{\text{ext}} = 4.3 \, \text{kV/cm}$ (dashed line), and $E_{\text{ext}} = 5.3 \, \text{kV/cm}$ (dashed-double-dotted line). The grey rectangle shows the region where the two necessary conditions for soliton observation are fulfilled (see text). The spatial solitons, i.e., the self-focused beams, that fulfill our quantitative conditions are highlighted by a circle.

Experiments in a KNbO$_3$ crystals show a good agreement with the numerical predictions and demonstrate the existence of (1+1)D-spatial solitons in this material. The beam reshaping oscillations discussed here are expected to occur also for input profiles different from a Gaussian beam in a qualitatively similar manner.
Appendix B

Photorefractive properties of Ce doped KNbO₃

Doping of photorefractive KNbO₃ crystal can increase dramatically the photosensitivity and photorefractive response time. Here we demonstrate experimentally that cerium doped KNbO₃ shows a high photorefractive sensitivity at 488 nm. The photorefractive performance of Ce doped, KNbO₃ is comparable to iron doped KNbO₃ which is considered as the dopant which highest sensitivity at this wavelength.

The boules of KNbO₃:Ce were drawn from a melt containing 1 atomic ppm of Ce. The segregation coefficient for KNbO₃:Ce is very high, and very low concentration of Ce are needed in the melt (compared to other dopants like Fe, Cu, Ni, Rh) to produce strongly doped crystal. Optical quality samples were prepared from the boules of KNbO₃:Ce. The samples are polished perpendicularly to the a-axis and the crystal total size is 2.77 x 4.21 x 2.44 mm³.

The crystals are characterized by the absorption spectrum, effective trap densities, and photorefractive two beam coupling experiments.

B.1 Absorption and photoconductivity

Figure B.1 a) shows the absorption for wavelengths ranging from 380 nm to 1500 nm as measured with a spectrometer (Perkin-Elmer Λ9). The absorption constant is very low in the near infrared and around 3 cm⁻¹ for a wavelength λ = 488 nm. Only a very weak anisotropy in the absorption is observed. The electrical conductivities reported in Fig. B.1 were measured with illumination intensities ranging from zero to 4 W/cm² at 488 nm. An electric field between −250 V/cm and +250 V/cm was applied along the c-axis and the current through the sample was detected with a galvanometer. Within the experimental uncertainty of ±10%, the current increases linearly with voltage and illumination intensity. The conductivity was measured at each intensity and its value at zero intensity is the dark conductivity σ₅₉₉. The increase of the conductivity per incident intensity unit is the photoconductivity σₚ₉. The photoconductivity is found to be σₚ₉ = 250·10⁻¹² Ω⁻¹ cm⁻¹ for a light intensity I₀ = 1 W/cm² whereas the conductivity in the dark was σ₅₉₉ = 24·10⁻¹⁸ Ω⁻¹ cm⁻¹ (extrapolated from the decay time). The photogalvanic currents density J₉ is the current measured at 1 W/cm² without an applied external field and was J₉ =
Appendix B. Photorefractive properties of Ce doped KNbO₃

Figure B.1: Absorption and photoconductivity measurements for Ce doped KNbO₃. The Absorption is plotted as function of the photon energy $\hbar\nu$ and the wavelength $\lambda$. The photoconductivity was measured at $\lambda = 532$ nm.

10 nA/cm². From the absorption constant $\alpha$ and from $\sigma_{ph}$ the mobility-lifetime product $\phi\mu\tau = \frac{\sigma_{ph}\hbar\nu}{\epsilon\mu_e} = 2.1 \cdot 10^{-14}$ m²/V was calculated, where $\phi$ is the probability that an absorbed photon produces a photoionized charge carrier. The values are reported and compared to Fe doped reduced KNbO₃ in Tab. B.1.

B.2 Two beam coupling

The photorefractive properties were investigated with two beam coupling experiments (see section 2.2.3). The laser source was an Ar-ion laser at a wavelength of 488 nm. The grating wavevector laid parallel to the $c$-axis. In the copropagating geometry the beams entered the crystal on the face perpendicular to the $a$ direction, whereas for the counterpropagating geometry this was achieved entering the samples through opposite faces of the polar $c$-axis. The results are reported in Fig. B.2 where the build-up time $\tau_{pr}$ and the exponential gain $\Gamma$ are reported as function of the grating spacing $\Lambda_g$. The solid lines in Fig. B.2 are obtained by a fit based on Eq. (2.25) and Eq. (2.32) respectively. We obtain $N_{eff}^* = (3.6^{+0.1}_{-0.5} \cdot 10^{22}$ m⁻³). Based on this measurement we also determined the photorefractive sensitivities $S_1 = 25 \cdot 10^{-11}$m³/J, $S_2 = 78 \cdot 10^{-9}$m²/J (see Eqs. (2.11)-(2.12)). These are better than in gas reduced Fe doped KNbO₃ crystals (see also Tab. C.1). Note that the Ce doped sample are nominally not reduced. The photorefractive properties are therefore expected to improve after an additionally post growth reduction treatment.

The sign of the dominant photoexcited charge carriers was determined from the direction of energy transfer. It was found that electrons are the dominant charge carriers for the investigated grating spacing range. The wavelength dependence of the
B.2. Two beam coupling

Table B.1: Comparison of absorption $\alpha$ and photorefractive parameters of Ce doped KNbO$_3$ with 1000 ppm Fe doped reduced KNbO$_3$. The parameters are the conductivity in the dark $\sigma_{\text{dark}}$, the photoconductivity $\sigma_{\text{ph}}$, measured at a light intensity of 1 W/cm$^2$, the effective trap density $N_{\text{eff}}^*$, the Maxwell dielectric relaxation time $\tau_{\text{Di}}$, and the diffusion length $L_D$. The largest observed sensitivities per absorbed and incident unit energy density, $S_1$ and $S_2$, respectively, are shown in the last two rows. The wavelength was $\lambda = 488$ nm.

<table>
<thead>
<tr>
<th></th>
<th>1 ppm Ce not treated</th>
<th>1000 ppm Fe gas reduced</th>
<th>1000 ppm Fe electrochemical red.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$ (cm$^{-1}$)</td>
<td>3</td>
<td>0.5 - 1</td>
<td>4 - 7</td>
</tr>
<tr>
<td>$\sigma_{\text{dark}}$ (m$^{-1}$)</td>
<td>$2.4 \cdot 10^{-15}$</td>
<td>$&lt; 10^{-10}$</td>
<td>$2.0 \cdot 10^{-10} - 1.6 \cdot 10^{-7}$</td>
</tr>
<tr>
<td>$\sigma_{\text{ph}}$ (m$^{-1}$)</td>
<td>$2.5 \cdot 10^{-8}$</td>
<td>$(2.0 - 3.7) \cdot 10^{-9}$</td>
<td>$1.0 \cdot 10^{-10} - 1.6 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>$\Phi_{\text{FR}}$ (m$^2$/V)</td>
<td>$2 \cdot 10^{-14}$</td>
<td>$9 \cdot 10^{-15}$</td>
<td>$2 \cdot 10^{-12}$</td>
</tr>
<tr>
<td>$N_{\text{eff}}^*$ (m$^{-3}$)</td>
<td>$(3.1 - 4.6) \cdot 10^{22}$</td>
<td>$(2.8 - 3.6) \cdot 10^{22}$</td>
<td></td>
</tr>
<tr>
<td>$\tau_{\text{Di}}$ (ns)</td>
<td>25 - 35</td>
<td>90 - 130</td>
<td></td>
</tr>
<tr>
<td>$L_D$ (nm)</td>
<td>90 - 170</td>
<td>62</td>
<td></td>
</tr>
<tr>
<td>$S_1$ (m$^3$/J)</td>
<td>$2.5 \cdot 10^{-10}$</td>
<td>$1.7 \cdot 10^{-10}$</td>
<td>$1.2 \cdot 10^{-8}$</td>
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<tr>
<td>$S_2$ (m$^2$/J)</td>
<td>$7.8 \cdot 10^{-8}$</td>
<td>$4.4 \cdot 10^{-8}$</td>
<td>$5.0 \cdot 10^{-6}$</td>
</tr>
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</table>

Figure B.2: Two beam coupling experiments in Ce doped KNbO$_3$. On the left-hand side the build-up time $\tau_{\text{pr}}$ is plotted as function of $\Lambda_g$. The intensity was $I_0 = 1$ W/cm$^2$. Whereas on the right-hand side the steady state exponential gain $\Gamma$ is reported as function of the grating spacing $\Lambda_g$.

Photorefractive effect was also measured. Ce doped crystal show a very small photorefractive effect at 633 nm, whereas no effect could be observed in the near infrared. This could indicate that the trap level in those crystals is near the middle of the band gap. For such crystals a very long decay time in the dark is expected, because
trapped electrons can move only through thermal excitation, which is minimized by traps in the middle of the band gap. Basing on this property, recently BaTiO$_3$ crystals were found which exhibited extrapolated decay times of 2200 years$^{[184]}$. The dark decay time of Ce doped KNbO$_3$ was measured and found to be 31 hours (see Fig. B.3), which is very long compared to other doped KNbO$_3$ crystals (see Tab. 3.2). Due to their high sensitivities Ce doped crystals show also a very fast build-up time of around 10 ms for an intensity of $I_0 = 1$ W/cm$^2$ (see Fig. B.3 left-hand side). Note that KNbO$_3$ crystals with such a fast build-up generally show also a sub second decay time in the dark. The simultaneous presence of high photosensitivity and small dark conductivities makes Ce doped crystal particularly appealing for holographic storage, and photorefractive application at a very low light power level of less than 1 $\mu$W should be possible.

Figure B.3: Build up and decay in the dark of a photorefractive grating in a Ce doped crystal. Note the time scale which is ms for the build-up and hours for the decay. The build-up was obtained with a light intensity $I_0 = 1$ W/cm$^2$ at a grating spacing of $\Lambda_0 = 1$ $\mu$m.
Appendix C

Interband photorefraction in Sn$_2$P$_2$S$_6$

Tin hypothiodiphosphate (Sn$_2$P$_2$S$_6$), which was recently intensively investigated because of its strong photorefractive properties in the near infrared region [185, 186], presents a band gap energy of 2.3 eV. This corresponds to a visible wavelength of 532 nm. Sn$_2$P$_2$S$_6$ is therefore a very promising candidate for demonstration of interband photorefraction in the visible.

In this appendix we present a study of the build-up and decay of holographic gratings in Sn$_2$P$_2$S$_6$ at a wavelength of 514 and 488 nm. The gratings were probed using a He-Ne laser at 633 nm in the longitudinal geometry (see Fig. C.1). Based on the measurements we will demonstrate that the nature of the effect is given by the photorefractive interband effect.

C.1 Grating build-up and decay

The Sn$_2$P$_2$S$_6$ crystal under investigation is cut along the crystallographic axis with the spontaneous polarization nearly parallel to the OX-axis (see also appendix D.4). The size of the sample is $X \times Y \times Z = 5.6 \times 5.9 \times 1.61$ mm$^3$ with the surfaces perpendicular to the Z direction polished. High optical quality surfaces were achieved using colloidal fluids as polishing medium. The writing beams entered the crystal in a symmetric arrangement producing a grating with a grating vector parallel to the X-axis and a grating spacing $\Lambda = 1$ μm (see Fig. C.1). Two different lines of an argon ion laser (488 and 514 nm) were used to write the grating. The absorption of the Sn$_2$P$_2$S$_6$ crystal at these wavelengths were extrapolated based on Urbach’s rule for small absorption [187]. The results are reported in Tab. C.1. The grating written in the blue-green is read-out measuring the diffraction efficiency of a p-polarized beam of a helium neon laser (633 nm), which is entering the crystal under the Bragg-angle of the holographic grating.

In Fig. C.2 a) a typical grating build-up for s-polarized writing beams at a wavelength of 488 nm and a total intensity of 720 mW/cm$^2$ is presented. The writing beams were turned on for 1 ms using an acusto-optic modulator with a response time of less than 1 μs. Under the measured conditions the build-up time was 10 μs and the maximum diffraction efficiency was $1.1 \cdot 10^{-4}$. The small diffraction
efficiency can be explained considering the strong absorption of the writing beams ($\alpha \approx 2000 \text{ cm}^{-1}$). For such an absorption we obtain a penetration depth ($d = 1/\alpha$) of a few $\mu$m. The build-up times and diffraction efficiency for s- and p-polarization and a wavelength of 488 and 518 nm are also reported in table C.1.

C.2 Identification of the photorefractive interband effect

There are in principle many physical effects which can lead to a photoinduced diffraction grating. In the following we list the most important effects observed in other ferroelectric wide band gap materials, and estimate their eventual contribution to the grating build-up.

| Table C.1: Absorption constant $\alpha$, penetration depth ($d = 1/\alpha$), build-up time $\tau$, diffraction efficiency $\eta$ at 720 mW/cm$^2$ intensity of the writing beam, and calculated amplitude of the refractive index change $\Delta n \approx \lambda_633\sqrt{\eta}/(\pi d)$, for the given wavelength $\lambda$ and polarization ($s$ corresponds to $E \parallel Y$, and $p$ to $E \parallel X$). |
|---|---|---|---|---|---|
| $\lambda$(nm) | Pol. | $\alpha$ (cm$^{-1}$) | $d$ ($\mu$m) | $\tau$ ($\mu$s) | $\eta$ | $\Delta n$ |
| 514 | s | 98 | 102 | 25000 | $1.6 \times 10^{-3}$ | $7.5 \times 10^{-5}$ |
| 514 | p | 250 | 40 | 125 | $1.7 \times 10^{-5}$ | $1.9 \times 10^{-5}$ |
| 488 | s | a 2000 | 5 | 11 | $1.1 \times 10^{-4}$ | $4.0 \times 10^{-4}$ |
| 488 | p | a 4000 | 2.5 | 3 | $1.6 \times 10^{-5}$ | $3.0 \times 10^{-5}$ |

$^a$ Absorption value extrapolated by exponential Urbach behavior for small absorption. The value presents a large uncertainty of (-50 .. +200 %).
C.2. Identification of the photorefractive interband effect

Figure C.2: Build-up a) and decay b) of a holographic grating induced by band-to-band photoexcitation at 488 nm. The writing beams are s-polarized with an intensity of 720 mW/cm². The writing beam were turned on for 1 ms.

Contribution from the pyroelectric effect

Due to the strong light absorption of Sn₂P₂S₆ at 488 and 518 nm, the crystal temperature will locally change as function of the incident light intensity when the light of the writing beam is turned on. The temperature dependence of the spontaneous polarization (pyroelectric effect) will generate a space charge field, which will be revealed by the electro-optic effect as phase grating. The mechanism was investigated in detail by Ducharme[187] and Busé[189]. Following Ducharme the maximum of the diffraction efficiency produced by the pyroelectric effect is given by

\[
\eta \approx \left( \frac{\pi L}{\lambda \cos(\theta)} f_i F_0 \right)^2
\]

where \( L \) is the grating thickness, \( \theta \) is the incidence angle of the reading beam, \( \lambda \) is the wavelength of the reading beam, \( F_0 \) is the fluence of the writing beams, \( n \) is the refractive index seen by the read-out beam, \( r_{\text{eff}} \) is the effective electro-optic coefficient, \( P_s \) is the spontaneous polarization, and \( \bar{e} \) is the volume specific heat of the material. Note that the contributions due to the pyroelectric effect are observed only for high intensity produced by pulsed illumination. Thus we have reported Eq. (C.1) because it can be applied for the pulsed experiments presented in section 5.2.3. For \( \lambda = 532 \text{ nm}, F_0 = 100 \mu J/cm^2, L = 0.3 \text{ mm} \) and the material parameters[190, 191] \( P_s = 2 \mu/(m^2K) \) (at 66°C), \( \bar{e} = 0.83 \text{ J/(cm}^3\text{K}) \) we obtain a maximum diffraction
efficiency $\eta_{\text{pyro}} < 10^{-6}$. This is 2 orders of magnitude lower than the observed diffraction.

The response time of the pyroelectric effect is given by

$$\tau_{\text{pyro}} = \frac{c}{\kappa K_g^2}$$

where $K_g$ is the grating spacing vector and $\kappa$ is the coefficient for thermal conductivity which is $\kappa = 0.5$ W/(mK) for Sn$_2$P$_2$S$_6$[192]. For a grating spacing $\Lambda_g = 1$ $\mu$m the resulting response time is given by $\tau_{\text{pyro}} = 42$ ns and is therefore much shorter than the response time observed in section 5.2.3.

**Direct thermal effect**

Due to the strong light absorption of Sn$_2$P$_2$S$_6$ at 488 and 518 nm, the crystal temperature will locally change as function of the incident light intensity and produce a diffraction grating through the thermo-optic effect. Under pulsed illumination the local temperature change $\Delta T$ can be calculated by

$$\Delta T = \frac{1}{\tilde{c}} \alpha F_0$$

which for the parameters used in previous section results in a temperature change of $\Delta T = 3.6$ mK. Unfortunately there was no data available for the thermo-optic effect in Sn$_2$P$_2$S$_6$. The influence of thermal effects could be excluded by the measurement of the grating vector dependence of the diffraction. In fact rotating the grating vector parallel to the Z-axis of the crystal no diffraction was observed. This is in contradiction with the nature of thermal grating which should produce a diffraction which is independent on the grating vector direction.

**Absorption grating**

The absorption seen by the read-out beam can be dependent on the local light intensity of the writing beams. The effect can be produced by a change of the trapping level population and was observed in many photorefractive materials like BaTiO$_3$, KNbO$_3$, among others. Absorption gratings[70] can be excluded with the same experimental argument used for the direct thermal effect. No diffraction is observed for a grating vector parallel to the Z-axis, whereas an absorption grating would produce diffraction also for this geometry.

**Surface grating**

In surface gratings the diffraction is produced by the mechanical deformation produced by the writing beams. The effect can be excluded in our experiments because the thin surface grating would produce multiple diffraction orders (Raman-Nath regime), which were not observed.
Photorefractive effect

Sn$_2$P$_2$S$_6$ presents a strong photoconductivity in the 488-532 nm wavelengths range, which clearly indicates that charge transport is involved with the grating build-up. In addition, rotating the grating spacing vector in the crystal X-Z plane, the angular dependence predicted by the electro-optic tensor symmetry is confirmed. Moreover, the measured diffraction efficiencies are compatible with the refractive index changes predicted by the theory reported in section 2.4. Also, the measurements of the photocurrent as function of the wavelength reported in Ref. [157], and the strong absorption present in the 488-532 nm wavelength range, confirm that direct band-to-band photoexcitation is involved. We therefore conclude that the effects observed in Sn$_2$P$_2$S$_6$ in the blue-green wavelengths range is produced by the interband photorefractive effect.

C.3 Dynamics of the interband grating

The dynamics of the diffraction efficiency presented in Fig. C.2 cannot be directly related to the dynamics of the space charge field reported in Ref.[89], because the intensity of the writing beam is exponentially decreasing as function of the depth inside the crystal. For the build-up of the holographic grating this means that the build-up time, the amplitude, and the phase of the grating will depend on the depth. The diffracted beam at the back side of the crystal is formed by the coherent sum of all the amplitudes of the diffracted light at different depths. In addition, it was shown by Bernasconi[193] that the phase of the grating depends strongly on the intensities/depths if additional traps are present.

The build-up was therefore modeled with a semi-heuristic double exponential build-up function of the form $\eta = \left[ \sqrt{\eta_1}(1 - \exp(-t/\tau_1)) + \sqrt{\eta_2}(1 - \exp(-t/\tau_2)) \right]^2$. The function is plotted in Fig. C.2 as a solid line, and allows an accurate description of the data points. We obtain a fast time constant $\tau_1 = 11 \, \mu s$ and $\tau_2 = 120 \, \mu s$ for the slower. Plotting the curve in a double logarithmic scale however shows that the point are not well fitted for small times, where the measured diffraction efficiency grows faster than the double exponential function.

The decay in the dark of the holographic grating measured after 1 ms of illumination (presented in Fig. C.2 b)) shows also a fast ($\approx 10 \, \mu s$) and a slow component.

Note that due to the robustness of the interband gratings, the presence of the red read-out beam does almost not influence the grating written in the blue-green. This holds also for intensity of the red light which are much larger than the intensity of the blue-green record beams. However, for depths much larger than the penetration depth $d = 1/\alpha$ of the blue-green beams, a coupling of the diffracted and transmitted red beam can occur. This can be verified by repeating the experiment with opposite direction of the spontaneous polarization. In our experiment however, this effect was of practically no importance because conventional photorefraction induced by red light is much slower than the interband photorefractive effect.

The experiment was repeated for different wavelengths and polarizations (see Tab. C.1). At a wavelength of 514 nm and s-polarization, the absorption is only 98 cm$^{-1}$; in this situation the conventional photorefractive effect dominates, this is also
suggested by the very slow build-up time of 25 ms. The large diffraction efficiency for this case can be understood considering the much higher penetration depth. The fastest effect ($\tau = 3 \, \mu s$) was measured at a wavelength of 488 nm and p-polarization.

### C.4 Intensity dependence

The diffraction efficiency is strongly dependent on the intensity of the writing beams. This is reported in Fig. C.3, where the dependence of the square root of the diffraction efficiency, is plotted as function of the intensity in a logarithmic scale. Under the crude assumption that the amplitude of the holographic grating is not dependent on the intensity of the writing beams, the diffraction efficiency would follow the relationship $\sqrt{\eta} = \Delta n / \alpha \ln(I_0/I_{\text{ref}})$, where $I_{\text{ref}}$ is a reference intensity. This would give a straight line for the scales chosen in Fig. C.3. As can be seen in Fig. C.3

\[
\begin{array}{c}
\lambda = 514 \, \text{nm}, E // X \\
\lambda = 514 \, \text{nm}, E // Y \\
\lambda = 488 \, \text{nm}, E // X \\
\lambda = 488 \, \text{nm}, E // Y
\end{array}
\]

**Figure C.3:** Square root of the peak diffraction efficiency, which is proportional to the refractive index change times the effective thickness, is plotted as function of the total intensity of the writing beams, for two different wavelengths and polarizations.

The diffraction efficiency is more than linearly increasing as function of the intensity for all wavelengths and polarizations. This suggests that much higher diffraction efficiencies can be achieved by increasing the intensity. However unfortunately it is not possible to illuminate the crystal for longer times because the strong absorption produces high temperature gradients in the crystal which deteriorate the effect and can lead to crystal damage. A way to overcome this problem is to use pulsed lasers, as presented in section 5.2.
In conclusion we have demonstrated for the first time interband photorefraction in Sn$_2$P$_2$S$_6$ crystals. Fast effects with build-up times of 3 $\mu$s at a light intensity of 720 mW/cm$^2$ and a diffraction efficiency of $10^{-4}$ could be obtained. The values are comparable with the results obtained in KNbO$_3$ in the ultraviolet. Much higher diffraction efficiencies are possible in the transversal geometry where the read-out beam is entering the crystal from the face perpendicular to z-axis.
Appendix C. Interband photorefraction in Sn$_2$P$_2$S$_6$
Appendix D

Material parameters

D.1 KNbO₃

Potassium niobate has a perovskite crystal structure with an oxygen octahedron centered at the Niobium (Nb) atom. At room temperature the crystal is in the orthorhombic phase with the point group mm2. In this phase the spontaneous polarization \( P_s \), which defines the crystallographic \( c \)-axis, is given by the displacement of the Nb atom in the direction of a two-fold symmetry axis of the oxygen octahedra. The \( a \)-axis is parallel to another two-fold symmetry axis and the \( b \)-axis is perpendicular to the \( a \) and \( c \) and is parallel to a four-fold axis.

The dispersion and temperature dependence of the refractive indices have been determined by Zysset et. al.[194]. The room temperature values at some selected wavelength are given by

\[
\begin{array}{c|ccc}
\lambda (\text{nm}) & n_a & n_b & n_c \\
488 & 2.352 & 2.242 & 2.23 \\
514 & 2.333 & 2.395 & 2.212 \\
633 & 2.280 & 2.330 & 2.169 \\
\end{array}
\]

The values of the electro-optic coefficient \( r_{\text{eff}} \) to be used in photorefractive experiments are a combination of the clamped and unclamped ones and can be found in Ref.[65]. The dispersion and temperature dependence have been determined by Bernasconi et. al.[195]. Below we give the values for the non vanishing unclamped (suffix \( T \), constant stress) and for the clamped (suffix \( S \), constant strain) dielectric
Appendix D. Material parameters

\( \varepsilon_{ij} \) and electro-optic \( r_{ijk} \) tensors.

\[
\begin{align*}
\varepsilon^T_{ij} &= \begin{pmatrix}
150 & \cdot & \cdot \\
\cdot & 985 & \cdot \\
\cdot & \cdot & 44 \\
\end{pmatrix} \\
r^T_{mk} &= \begin{pmatrix}
\cdot & \cdot & 34 \\
\cdot & \cdot & 63 \\
120 & \cdot & \cdot \\
\end{pmatrix} \\
\varepsilon^S_{ij} &= \begin{pmatrix}
37 & \cdot & \cdot \\
\cdot & 780 & \cdot \\
\cdot & \cdot & 24 \\
\end{pmatrix} \\
r^S_{mk} &= \begin{pmatrix}
\cdot & \cdot & 20 \\
\cdot & \cdot & 7.1 \\
28 & \cdot & \cdot \\
\end{pmatrix}
\end{align*}
\]

where the units for the electro-optic coefficients are (pm/V) and we used the reduced form of the electro-optic tensor with the transformations \( ij \rightarrow mk: 11 \rightarrow 1; 22 \rightarrow 2; 33 \rightarrow 3; 23, 32 \rightarrow 4; 13, 31 \rightarrow 5; 12, 21 \rightarrow 6. \)

D.2 LiNbO\(_3\)

LiNbO\(_3\) is trigonal at room temperature and belongs to the point group 3\(m\). The spontaneous polarization is along the \(c\)-axis which exhibits three-fold rotation symmetry. The three equivalent \(a\)-axis of the conventional hexagonal unit cell are 120° apart and lie in a plane normal to the \(c\)-axis. The coordinate system used to describe the physical tensor properties is a Cartesian \(x, y, z\) system. The \(z\)-axis is chosen to be parallel to the \(c\)-axis. The \(x\)-axis is chosen to coincide with any of the equivalent \(a\)-axis. After the \(x\) and \(z\)-axes are selected, the \(y\)-axis is chosen such that the system is right handed. Thus the \(y\)-axis must lie in a plane of mirror symmetry.

The room temperature refractive indices at some selected wavelengths are given by [196]

<table>
<thead>
<tr>
<th>( \lambda ) (nm)</th>
<th>( n_0 )</th>
<th>( n_c = n_s )</th>
</tr>
</thead>
<tbody>
<tr>
<td>488</td>
<td>2.349</td>
<td>2.256</td>
</tr>
<tr>
<td>514</td>
<td>2.333</td>
<td>2.242</td>
</tr>
<tr>
<td>633</td>
<td>2.287</td>
<td>2.203</td>
</tr>
</tbody>
</table>

The values of the non vanishing unclamped (suffix \(T\), constant stress) and for the clamped (suffix \(S\), constant strain) dielectric \( \varepsilon_{ij} \) and electro-optic \( r_{ijk} \) tensors are
D.3 LiTaO$_3$

The crystal structure of LiTaO$_3$ is the same as LiNbO$_3$ if the Niobium (Nb) is replaced by Tantalum (Ta). Also the resulting optical properties of the material are similar.

The refractive indices at some selected wavelengths at room temperature are given by\cite{196}

<table>
<thead>
<tr>
<th>(\lambda) (nm)</th>
<th>(n_0)</th>
<th>(n_c = n_e)</th>
</tr>
</thead>
<tbody>
<tr>
<td>450</td>
<td>2.242</td>
<td>2.247</td>
</tr>
<tr>
<td>500</td>
<td>2.216</td>
<td>2.221</td>
</tr>
<tr>
<td>600</td>
<td>2.183</td>
<td>2.188</td>
</tr>
</tbody>
</table>

The values of the non vanishing unclamped (suffix \(T\), constant stress) and for the clamped (suffix \(S\), constant strain) dielectric \(\varepsilon_{ij}\) and electro-optic \(r_{ijk}\) are given by\cite{196}

\[
\varepsilon_{ij}^T = \begin{pmatrix} 54 & \cdot & \cdot \\ \cdot & 54 & \cdot \\ \cdot & \cdot & 43 \end{pmatrix}, \quad \varepsilon_{ij}^S = \begin{pmatrix} 42.6 & \cdot & \cdot \\ \cdot & 42.6 & \cdot \\ \cdot & \cdot & 42.8 \end{pmatrix} \tag{D.3}
\]

\[
r_{mk}^T = \begin{pmatrix} \cdot & -0.2 & 7.9 \\ \cdot & 0.2 & 7.9 \\ \cdot & \cdot & 27 \end{pmatrix}, \quad r_{mk}^S = \begin{pmatrix} \cdot & 1 & 8.4 \\ \cdot & -1 & 8.4 \\ \cdot & \cdot & 22 \end{pmatrix}
\]

D.4 Sn$_2$P$_2$S$_6$

Sn$_2$P$_2$S$_6$ presents a ferroelectric phase\cite{197} below \(T_c = 66^\circ C\). A spontaneous polarization appears at the phase transition because of the displacement of the Sn ions
from the centrosymmetric position occupied by them at temperatures $T > T_c$. In the ferroelectric phase the symmetry is monoclinic and belongs to the point group $C_s$ (symmetry class $m$). This symmetry class exhibits only one single symmetry plane. Following Dittmar [198], the coordinate system chosen to describe the physical tensor properties is a Cartesian $X,Y,Z$ system. The $Y$-axis is chosen perpendicular to the symmetry plane, and the $X$-axis is chosen parallel to the crystal axis $a$ (see Fig. D.1) and perpendicular to the $Y$-axis. The $Z$ axes is chosen perpendicular to the $X$ and $Y$ in such a way that the system is right handed. The results elementary cell is shown in Fig. D.1. In this coordinate system the direction of the spontaneous polarization results in the symmetry plane forming an angle of approximately $12^\circ$ with the $x$ axes.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Sn2P2S6_structure.png}
\caption{Schematic structure of Sn$_2$P$_2$S$_6$. The symmetry plane is parallel to the plane of the figure. The unit cell in the Dittmar notation is indicated by the dashed lines.}
\end{figure}
The refractive indices of Sn\(_2\)P\(_2\)S\(_6\) at room temperature is reported in Fig. D.2 as function of the wavelength. The data were taken from Refs. [199] and [200], whereas the solid line are the result of fit with single Sellmeier oscillator of the form

\[
n^2(\lambda) - 1 = \frac{s_0 \lambda_0^2}{1 - (\lambda_0/\lambda)^2}.
\]  

(D.4)

The values for the oscillator strength \(s_0\) and the corresponding oscillator wavelength \(\lambda_0\) for the three different refractive indices of Sn\(_2\)P\(_2\)S\(_6\) are

<table>
<thead>
<tr>
<th>(n_i)</th>
<th>(S_0 ) ((\mu)m(^{-2}))</th>
<th>(\lambda_0) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(n_1)</td>
<td>76 ± 4</td>
<td>292 ± 7</td>
</tr>
<tr>
<td>(n_2)</td>
<td>75 ± 5</td>
<td>284 ± 5</td>
</tr>
<tr>
<td>(n_3)</td>
<td>68 ± 3</td>
<td>313 ± 6</td>
</tr>
</tbody>
</table>

Note that the optical indicatrix is not diagonal in the crystal coordinates, but rotated in the \(X\)-\(Z\) plane. The rotation angle is also reported in Fig. D.2.

For the monocline crystal structure the electro-optic tensor has 10 independent elements. Unfortunately most of those elements are still unknown. The effective electro-optic coefficients relevant for writing a grating with grating vector parallel to the \(X\)-axis were indirectly determined in Ref.[201], by photorefractive two beam coupling experiments. The effective electro-optic coefficient in this geometry and horizontal polarization was found to be \(r_{\text{eff hor}} = (60 \pm 10) \) pm/V and for vertical polarization \(r_{\text{eff vert}} = (23 \pm 4) \) pm/V. The effective relative dielectric permittivity relevant for the same geometry as above, was estimated to be around \(\varepsilon_{\text{eff}} \approx 300\).
Appendix D. Material parameters
Appendix E

Electrical devices for optical computing

Parallel optical computing allows an efficient implementation of many basic mathematical operations. The input data to be processed and the output of the calculation are required to be in electronic form. An efficient way to transform digital data to modulated optical beams and back is therefore crucial in order to exploit the advantages optical computing.

Information is encoded on light beams by Spatial Light Modulators (SLM), which allow to modulate either the transmission amplitude or the phase of an incident light beam. Many physical effects can be used to build a SLM. In this section we will give a short overview of the most common SLMs for the visible wavelength and their key properties.

The detection of the information carried by a spatially modulated beam is achieved by one- or two-dimensional array of photodetectors. A widely used method, the Charge Coupled Device (CCD) allows a simple transformation of the input intensity pattern in a serial electronic signal. The characteristics of commercially available CCDs will be presented in section E.2.

In certain applications like angular multiplexed holographic memory, it is required to change the direction of the optical beam in a fast controlled way. This can be achieved by beam deflectors which are considered in the last section.

E.1 Spatial light modulators

A spatial light modulator (SLM) is a device capable of impressing information onto an optical wavefront. SLMs exist in many forms. Light modulation may be carried out by electro-optic, acousto-optic, and mechanical effects, among others. An overall review of the field can be found in Refs.[202] and [203]. In this section we will concentrate on SLM based on liquid crystal due to their large availability.

E.1.1 Twisted nematic spatial light modulators

Liquid crystal possess physical properties that are intermediate between conventional fluids and solids. They are fluid like, yet the arrangement of molecules within
them exhibit structural order. Liquid crystal can exist in three phases: nematic, cholesteric, and smectic. Nematic phase exhibits long-range molecular orientational ordering but possesses no positional ordering. The aligned nematic liquid crystal molecules, on the average, is characterized by one symmetry axes called the director. The director can be reoriented by an external electric field. A very attractive alignment of nematic liquid crystal is the so called twisted alignment. In this case the director is chosen parallel to the cell boundaries and rotated as function of the distance perpendicular to the cell boundaries, resulting in an helix-like structure. Applying a field perpendicular to the cell, the directors are reoriented parallel to the external field. In combination with an analyzer polarizer the liquid crystal can be used as amplitude or phase modulator, which work at low voltages of few Volts. Twisted nematic liquid crystal and its derivative can be found in many electronic devices like pocket TV’s, video projectors, and head-up displays. The response times of the nematic liquid crystals are typically in the 10 to 100 ms range.

E.1.2 Ferroelectric liquid crystal spatial light modulators

In contrast to nematic liquid crystals, ferroelectric liquid crystals (FLCs) have an electric polarization even in the absence of an externally applied electric field. Therefore the molecules respond directly to an applied electric field. A field of one polarity drives the FLC into what may be defined as on-state, and the FLC is switched off with a field in the opposite polarity. One desirable feature of FLCs is the fast switching (around some 10 $\mu$s) which results from the spontaneous polarization and from the ability to drive the FLC to its off state. FLCs exhibit a hysteresis, as do all ferroelectrics. If the hysteresis is sufficiently broad and the slope of the transition from one state to the other is sufficiently steep, the FLC remains in its prior state when the applied field is switched off to zero. The bistability is another of the desirable features of some FLCs.

FLCs can be obtained suppressing the helical structure in smectic C liquid crystal. This is accomplished by constraining the material to fill a region between two plates which is separated by a distance of less than a few times the helical pitch of the smectic C phase, typically 1 to 5 $\mu$m.

FLCs rely on birefringence to modulate light. The refractive index along the optic axis parallel to the molecular director is significantly different from the index along an optic axis which is perpendicular to the director. The difference, the birefringence ($\Delta n$) is quite substantial for FLC molecules, typically around 0.1.

FLCs are often driven by very large scale integration technology (VLSI) chips, which are bonded to transmissive substrates to form active backplane SLMs. Active backplane SLMs may be used in small head-up displays and can provide the performance required in optical computing and processing applications. Even with the limited frame rate of liquid crystal SLM ($< 1$ MHz) as compared with gallium arsenide multi-quantum-well modulators, for example, their high spatial resolution and large array sizes meet the needs of many high-performance applications. With current foundry lithography capabilities VLSI/FLC active backplane SLMs as large as 1280 $\times$ 1024 are feasible, running as a frame rate of 10 kHz with a contrast ratio of better than 100:1.
E.2 Imaging sensors

An individual photodetector registers the photon flux striking it as a function of the time. In contrast, an array containing a large number of photodetectors can register simultaneously the photon flux from many spatial points. Modern microelectronics technology permits the realization of arrays with a large number of individual semiconductor photodetectors. The accumulated electric charge separation generated by the individual detectors can be transferred into a Charge-Coupled device (CCD) read-out structure. The CCD transfer gate permits the charge to be transferred to a buried CCD channel at a specified time. Many different kinds of electrode structure and clocking schemes have been developed for periodically read-out the charge accumulated by each pixel and thereby generating an electronic data stream representing the image. Beside conventional CCD-detectors operating at standard video resolution and video rate, two main directions were pursued: First the number of pixels was increased obtaining resolutions as high as 16,000 x 16,000 pixels. Second the frame rate could be increased till 50,000 images/s by appropriate high speed data transfer technique.

An alternative way to achieve high speed and high sensitive detector is given by multichannel photomultiplier detectors. Arrays of 64 x 64 pixels are available. The advantage compared to the CCD-technology is the much higher light sensitivity which can be obtained which such devices.

E.3 Beam deflectors

Beam deflectors are important in optical applications, like optical interconnects, laser projection system, and profile scanning system. An obvious way to build a beam deflector is by a rotating mirror. Faster deflection angle changes can be obtained using acousto-optic or electro-optic effect.

E.3.1 Mechanical rotating mirrors

Mechanical rotating mirror exist in many forms. A particularly good compromise between angle precision and response time is obtained in galvanic scanners. In this devices a angle precision of 10 μrad can be obtained with a response time of < 1 ms for a 1° angle step. An advantage compared to acousto-optic deflectors is that much higher deflection angle (± 40°) can be obtained. The response time of such devices can be further increased be reducing the size the mirror.

E.3.2 Acousto-optic deflectors

Acousto-optic deflectors are based on optical diffraction on refractive index grating produced by a propagating acoustic wave. An acoustic wave with a frequency $f_0$ and a sound speed $v_s$ produces a beam deflection angle $\Delta \theta$ of

$$\Delta \theta = \frac{\lambda}{v_s} \Delta f$$  \hspace{1cm} (E.1)
where $\Delta f$ is the frequency change of the acoustic wave. The deflection $\Delta \theta$ can be controlled by frequency modulation (FM) of the acoustic wave. In this way it is possible to achieve scan angle of typically $2^\circ$, where the setup time is mainly given by the time required for the acoustic wave to propagate and is in the range from 0.1 to 10 $\mu$s. The maximum diffraction efficiency which can be achieved with acousto-optic deflectors ranges from 30 to 80%. A drawback of acousto-optic modulator is the Doppler shift induced by the diffraction on the moving grating. To perform interferometric experiments with the deflected beam, the frequency shift will have to be compensated in the second arm of the interferometer by a second deflector.
Appendix F

Stored images

Figure F.1: The images *Metamorphosis II* by M. C. ESCHER[140], no. 1-49
Figure F.2: The images *Metamorphosis II* by M. C. Escher[140], no. 50-100
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[58] I. Biaggio, “Photorefractive effects induced by short light pulses,” Ph. D., Diss. ETH No. 10009, Swiss Federal Institute of Technology ETH, Zürich, Switzerland, 1993. 13, 104


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Conference proceedings


- R. Ryf, G. Montemezzani, M. Zgonik, and P. Günter, “Route to spatial soliton in photorefractive KNbO₃ crystals”, European Conference on Applications of Polar Dielectrics (ECAP'D-3 ’96), Bled, Slovenia, August 1996


• R. Ryf, G. Montemezzani, and P. Günter, “Photorefractive Ce doped KNbO₃ as holographic storage material”, European Conference on Applications of Polar Dielectrics (ECAPD-4 ’98), Montreux, August 1998


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Curriculum vitae

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1987-1990  Study of electrical engineering at the Neu Technikum Buchs (NTB Buchs)
1990  Diploma in software engineering on “Specification of an access control system”
1990-1995  Study of Physics at the Swiss Federal Institute of Technology Zürich (ETH Zürich)
1995  Graduation with a diploma in experimental physics on “Application of integrated-optical difference interferometer based on ridge waveguides as sensor”
since 1995  Research and teaching assistant in the group of Prof. Dr. P. Günter, ETH Zürich
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