PHOTOREFRACTIVE PROPERTIES AND APPLICATIONS OF KNO₃ CRYSTALS

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PHOTOREFRACTIVE PROPERTIES AND APPLICATIONS OF KNbO₃ CRYSTALS

Abstract
Both doped and nominally pure KNbO₃ single crystals exhibit extraordinarily attractive photorefractive properties. These crystals possess simultaneously favorable photoconductivity and also large electro-optic effects. These properties can be finally shaped to measure by different consecutive procedures after crystal growth, according to various applications and their needs. It was found that the electrochemical reduction is an extremely suitable method.

It was one aim of the present work to examine the photorefractive properties of pure and doped KNbO₃ crystals before and after such treatments. It is of special interest to obtain more information about the defect centers active in the photorefractive process. In doing this, optical spectroscopy, photoconductivity experiments, paramagnetic resonance technique, and photorefractive methods were applied.

In the introduction an overview of the phenomenological theory of the photorefractive effect, based on band models, is given. For this task, it was necessary to develop partly new approaches and special approximations, which were discussed in the interpretation of the experimental results obtained with KNbO₃.

Because of the exceptionally large electro-optic coefficients $r_{42}$ and $r_{51}$ of KNbO₃ an essential part of the present thesis deals with the discussion of the so-called anisotropic Bragg diffraction. In case of the special type of diffraction occurring in such photoinduced gratings, the polarization directions of the incident and diffracted linearly polarized light beams differ, i.e., they are orthogonal with respect to each other. As KNbO₃, being an optically biaxial crystal, possesses a relatively large birefringence, the incident and diffracted wave propagate with different velocities and have different absolute values of their wave vectors. This leads to very special diffraction conditions. The occurring diffraction geometries are discussed and the pertinent experimental results are presented. In a theoretical treatment based on the "coupled wave theory," an analytical solution of the diffraction efficiency for weak violations of the Bragg condition is deduced and discussed.

One of the objectives of the present work was the design and investigation of an optically addressable light deflector. It was found that the anisotropic
Bragg diffraction in a special, non-critical configuration is ideally suitable for such an application. This was made possible by varying the spacing of the photoinduced grating by changing the wavelength of the writing beams. In such a configuration the read-out beam entering the crystal at a constant incidence angle is deflected in different directions depending on the wavelength of the writing beams. Changes of up to 57.7 nm (457.9 nm < \lambda < 514.5 nm) yielded a deflection range of up to 5.67° for a HeNe-laser beam.

In a second application this type of light deflection is used for the design of an optically addressable fiber switch. In the configuration described above, a variably deflected laser beam can be coupled by a focussing lens behind the crystal into any selected fiber of a linear array. A prototype of a linear array containing 128 multimode fibers was used for a 1x128 fiber switch. In this device the information beam entering through a fiber can be deflected to any position of the 128 exit channels (fibers) by a variation of the control wavelength (produced by a Rhodamin 6G dye laser). Using reduced KNbO₃ single crystals as host of the variable grating, switching times of typically 1 ms and efficiencies of 1% can be reached (ratio of the intensities in the output and input fiber).

In a further application of anisotropic Bragg diffraction a special writing geometry is used. In these experiments the writing beams are diffracted by the self-induced grating. In contrast to the case of isotropic diffraction, the deflected beam differs both in direction and polarization with respect to the writing beams. This so called anisotropic self-diffraction is used for designing an optically addressable spatial light modulator or incoherent-to-coherent converter. In this application, in addition to the two writing beams a two dimensional image (information) is projected into the crystal. Thereby the photoinduced grating amplitude is changed in such a manner that in the bright zones of the projection the diffraction efficiency is decreased because of a local increase of the photoconductivity. In such a way the image can be transferred with inverted contrast from the incoherent beam onto the self-diffracted beam. A simple theoretical model describing the intensity transfer characteristics is proposed for this process, and the factors limiting the resolution are examined. Experiments with reduced KNbO₃ single crystals led to resolutions of up to 10 line pairs per millimeter and conversion times of the order of 1 ms.
PHOTOREFRAKTIVE EIGENSCHAFTEN UND ANWENDUNGEN VON K\textsubscript{2}N\textsubscript{3}O\textsubscript{9}-KRISTallen

Zusammenfassung

Sowohl dotierte als auch nominell reine K\textsubscript{2}N\textsubscript{3}O\textsubscript{9} Kristalle besitzen ausserordentlich attraktive photorefraktive Eigenschaften. Einerseits zeigen diese Kristalle grosse elektro-optische Effekte und sind andererseits gut photoleitend. Insbesondere eine nachträgliche Materialbehandlung eröffnet die Möglichkeit, K\textsubscript{2}N\textsubscript{3}O\textsubscript{9} Kristalle für die verschiedensten Anwendungen und Anforderungen "nach Mass zu schneidern". Dabei zeigt sich die elektrochemische Reduktion als äusserst geeignete Methode.


Zunächst wird in einer Einführung ein Überblick über die, auf Bandmodellen basierende, phänomenologische Theorie des photorefraktiven Effektes gegeben. Dabei werden teilweise neue Wege gegangen und spezielle Nähерungen hergeleitet und diskutiert, soweit sie zur Interpretation von experimentellen Resultaten gebraucht werden.

Aufgrund der besonders grossen elektro-optischen Koeffizienten $r_{42}$ und $r_{51}$ von K\textsubscript{2}N\textsubscript{3}O\textsubscript{9} besteht ein wesentlicher Teil der vorliegenden Arbeit in der Diskussion der sogenannten anisotropen Bragg Beugung. Bei dieser speziellen Art von Beugung an photoinduzierten Gittern unterscheiden sich die Polarisationsebenen von einfallendem und abgebeugtem linear polarisiertem Licht, sie sind orthogonal zueinander. Da nun K\textsubscript{2}N\textsubscript{3}O\textsubscript{9} als optisch zweifachsesiger Kristall eine im Vergleich zu andern hohe Doppelbrechung zeigt, breiten sich einfallende und abgebeugte Welle mit ganz verschiedenen Geschwindigkeiten aus und ihre Wellenvektoren haben verschiedene Beträge. Dies führt dazu, dass die Bragg Bedingung nur für ganz spezielle Beugungsgeometrien erfüllt werden kann. Diese Beugungsanordnungen werden diskutiert und mit dazugehörigen experimentellen Resultaten untermauert. In einer theoretischen Behandlung, basierend auf der "Theorie der gekoppelten Wellen", wird die analytische Lösung der Beugungseffizienz für Lichtwellen, welche die Bragg Bedingung zumindest beinahe erfüllen, hergeleitet und diskutiert.


1. Introduction

1.1 History of the Photorefractive Effect

In 1966 ASHKIN et al. at Bell Laboratories discovered that some ferroelectric crystals like LiNbO$_3$, LiTaO$_3$ and BaTiO$_3$ showed a change in the refractive index by illumination with intense blue or red light [1.1]. These index variations persisted in the dark, sometimes for many months, and could be erased by flooding the crystal with uniform illumination. Although the authors realized that this effect could be interesting in itself, it was considered at that time as highly detrimental to possible acousto-optic, electro-optic and nonlinear optic applications.

In 1968, however, CHEN et al. [1.2] proposed the use of this so called "optical damage" to develop erasable holographic memory devices. In their first experiments performed with LiNbO$_3$ they found very promising properties for such applications. Resolutions of up to 1600 lines per millimeter (this leads to storage densities in the order of $10^{12}$ bits per cm$^3$) and diffraction efficiencies up to 42% were observed. In the same work first annealing experiments in an oxygen-deficient environment at 900°C were reported which enhanced the efficiency and decreased the recording times.

In the past years photorefractive effects (as "optical damage" was called later) have been observed in a variety of electro-optic materials including, in addition to the materials mentioned above, KTa$_{1-x}$Nb$_x$O$_3$ (KTN) [1.3], KNbO$_3$ [1.4], Bi$_{12}$SiO$_{20}$ and Bi$_{12}$GeO$_{20}$ [1.5], Bi$_{12}$TiO$_{20}$ [1.6], Ba$_2$NaNb$_5$O$_{15}$ [1.7], Sr$_{1-x}$Ba$_x$Nb$_2$O$_6$ [1.8], PLZT-ceramics [1.9], GaAs [1.33] and InP [1.34].

Besides the high storage density mentioned above, it was soon recognized that photorefractive crystals used as media for three dimensional holographic recording (volume holograms) do possess exposure sensitivities comparable to that of the best photographic emulsions (at least 100μJ per cm$^3$). But, in contrast to conventional media, photorefractive crystals can be used for dynamic or "real-time" holography in which the writing and reading process occurs continuously and simultaneously. Several proposals for dynamic applications have been given, including optical image amplifiers [1.10], image correlators [1.11], edge enhancers [1.12], spatial light modulators [1.13], optical diodes [1.14] and laser beam deflectors [1.15]. All these concepts show clearly the
importance that photorefractive materials will have in the future of optical signal processing and optical computing.

A first explanation of the photorefractive effect was given by CHEN [1.16]. He considered the refractive-index change to be caused by a drift of photexcited carriers under the influence of an internal electric field. He assumed that there are electron traps in the material. Initially, some of the traps are filled (state of neutral charge) and they provide electrons upon photoexcitation. The others are empty and capture electrons. In addition, he assumed that there is an internal electric field in the direction opposite to that of the spontaneous polarization. The photoexcited electrons drift towards the positive side of the field leaving behind positive charges of ionized trapping centers. The photoexcited charges will be retrapped and reexcited until they finally drift out of the illuminated region and stay trapped. Therefore, a space-charge field is created between the positively ionized centers and the trapped negative charges. This space-charge field causes the spatial variation of the refractive index via the linear electro-optic effect in the sample. The need for the ad hoc assumption of an internal field in Chen’s model was removed by GLASS et al. [1.17] by introducing the concept of the bulk photovoltaic effect.

Another model has been proposed by AMODEI [1.18]. He has pointed out that the charge migration by diffusion is an important factor in holographic recording for sufficiently small grating periods. He has shown that even in the absence of internal or externally applied fields, the photoexcited electrons still can migrate out of the illuminated regions by thermal processes. For plane-wave holograms, in the case of initial and steady states of holographic recording, he has derived expressions for the electric field patterns generated through diffusion and through drift, respectively [1.19].

The model describing up to now most of the results of photorefractive experiments, was derived by KUKHTAREV et al. [1.20]. Basically it is a transport model, assuming the existence of photoexcitable centers. It takes into account the effects of externally applied fields, the bulk photovoltaic effect, and the recursive effects of the space-charge field on the distribution of the space charge itself.
The importance of defects and impurities for the photorefractive properties was realized very early. HOU et al. [1.21] found that the optical absorption edge of $\text{Bi}_2\text{SiO}_20$ single crystals was broadened by excitons and/or impurities. They also noted that in undoped and lightly doped crystals, electrons dominated the photocurrent while in heavily doped crystals holes were dominant.

The effect of doping was also found to be important in $\text{LiNbO}_3$ crystals [1.22]. Iron was proved to be very effective for the enhancement of photorefractive properties. Iron acts as a donor-acceptor trap via valency exchange effects such as $\text{Fe}^{3+} + e^- \leftrightarrow \text{Fe}^{2+}$. It was found, moreover, that Fe-doping improves the photorefractive properties of most oxygen-octahedra ferroelectrics. The space-charge build-up by the action of light is attributed to a charge re-distribution between divalent and trivalent impurities.

1.2 $\text{KNbO}_3$ - a nonlinear optical material

After the invention of the laser in 1960 a search for nonlinear optical materials started. The realization of many hypothetical devices depended on the discovery of an optical equivalent of the electronic Silicon, a material in which optical waves are able to interact strongly. Although there exists up to now a large number of known nonlinear optical materials, it seems that an "optical silicon" has not yet been found and therefore, the optical materials' research is still important and in rapid progress.

It was realized very soon that the class of ferroelectric oxides has a number of properties which make them attractive for use in optical devices [1.23]. They have wide band gaps ($>3\text{eV}$), large electro-optic and nonlinear optical coefficients, high static dielectric constants, and many of them exhibit a spontaneous polarization in suitable temperature ranges. Candidates were choosen mainly from three basic structures, the simple $\text{ABO}_3$ perovskite lattice, the trigonal $\text{LiNbO}_3$-type structure and the so-called tetragonally distorted tungsten bronze structure [1.23].

$\text{KNbO}_3$ is one of the ferroelectric oxide-perovskite crystals available in large single domain samples. By the method of top seeded solution growth
It is possible to obtain crystals which can be prepared to single domain samples up to a size of approximately 20×20×10 mm³ [1.24,1.25].

In the past years the basic physical properties of KNbO₃ were thoroughly studied and led more recently to device oriented investigations of electro-optical and nonlinear optical properties.

In this section I will review the physical properties of KNbO₃ as far as they are important for the present work.

**Physical properties of orthorhombic KNbO₃**

KNbO₃ undergoes the same series of ferroelectric transitions in the same sequence as BaTiO₃[1.31]. Table 1.1 shows the different phases and the corresponding transition temperatures of KNbO₃. All these transitions are of first order and hysteresis effects are readily observed.

The oxygen framework forms a rigid octahedral array and the displacement from cubic perovskite structure is dominantly parallel to the polar axis in each phase. In Fig.1.1 the structure of KNbO₃ and the direction of the polar axis in the room temperature phase is shown.

<table>
<thead>
<tr>
<th>Symmetry</th>
<th>Transition for increasing temp.</th>
<th>Transition for decreasing temp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rhombohedral 3m</td>
<td>-29°C</td>
<td>-50°C</td>
</tr>
<tr>
<td>Orthorhombic mm2</td>
<td>218°C</td>
<td>203°C</td>
</tr>
<tr>
<td>Tetragonal 4mm</td>
<td>436°C</td>
<td>426°C</td>
</tr>
<tr>
<td>Cubic m3m</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Fig.1.1 Structure of orthorhombic KNbO$_3$ (room temperature structure)

Many of the physical tensor properties of KNbO$_3$ show large anisotropies at room temperature. The clamped and free dielectric constants are [1.27]

$$\varepsilon_{11}^S = 37, \quad \varepsilon_{22}^S = 780, \quad \varepsilon_{33}^S = 24,$$
$$\varepsilon_{11}^T = 160, \quad \varepsilon_{22}^T = 1000, \quad \varepsilon_{33}^T = 55,$$

where $\varepsilon^S$ and $\varepsilon^T$ are the dielectric tensor elements at constant stress ($S$) and strain ($T$), respectively.

At optical frequencies the dielectric properties are characterized by the refractive indices which at room temperature are very well described by a 2-oscillator Sellmeier dispersion relation

$$n^2(\lambda) = \frac{S_1\lambda_1^2}{1 - (\lambda_1^2/\lambda^2)} + \frac{S_2\lambda_2^2}{1 - (\lambda_2^2/\lambda^2)} + 1 \quad (1.1)$$

In Tab.1.2 the parameters $S_1\lambda_1^2$, $S_2\lambda_2^2$, $\lambda_1^2$ and $\lambda_2^2$ are listed for KNbO$_3$ at 22°C.

The most relevant third rank tensors for this work are the electro-optic and nonlinear optic tensors. The values of all symmetry allowed elements for KNbO$_3$ are listed in Tab.1.3. They all are relatively large, and this is a
reason why KNbO₃ is an interesting material for nonlinear optics.

Tab.1.2 Parameters for the Sellmeier 2-oscillator fit of the refractive indices of KNbO₃ at room temperature [1.28]

<table>
<thead>
<tr>
<th></th>
<th>$S_1\lambda_1^2$</th>
<th>$\lambda_1^2(10^{-20}m^2)$</th>
<th>$S_2\lambda_2^2$</th>
<th>$\lambda_2^2(10^{-20}m^2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_1$</td>
<td>1.4392</td>
<td>5985156</td>
<td>2.3592</td>
<td>2302662</td>
</tr>
<tr>
<td>$n_2$</td>
<td>0.1362</td>
<td>11308698</td>
<td>3.8013</td>
<td>4087914</td>
</tr>
<tr>
<td>$n_3$</td>
<td>0.0875</td>
<td>10724149</td>
<td>3.3023</td>
<td>3130828</td>
</tr>
</tbody>
</table>

Tab.1.3 Electro-optic and nonlinear optic tensor elements of KNbO₃ at room temperature.

<table>
<thead>
<tr>
<th>Electro-optic tensor elements</th>
<th>Nonlinear optic tensor elements</th>
</tr>
</thead>
<tbody>
<tr>
<td>(10⁻¹²m/V) after Ref.[1.29]</td>
<td>(10⁻¹²m/V) after Ref.[1.30]</td>
</tr>
<tr>
<td>$r_{13}=28$, $r_{23}=1.3$, $r_{33}=64$</td>
<td>$d_{31}=-27$, $d_{32}=-18$, $d_{33}=-16$</td>
</tr>
<tr>
<td>$r_{42}=380$, $r_{51}=105$</td>
<td>$d_{24}=17$, $d_{15}=17$</td>
</tr>
<tr>
<td>(for $\lambda=633$nm)</td>
<td>(for $\lambda=1.06\mu$m)</td>
</tr>
</tbody>
</table>

The fact that the grating build-up in a photorefractive material is produced by a redistribution of ionized impurity centers suggests, that the nature and concentration of dopants and defects determine the response of the material under light illumination, i.e. the temporal evolution and the amplitude of the photoinduced space-charge field. Therefore, the knowledge of all the extrinsic properties of a photorefractive crystal is very important for a complete characterization. In KNbO₃ some work has already been done for identifying and characterizing the relevant centers [1.32] and it was one of the aims of this thesis to continue this work (Sect.3).

One of the main advantages of KNbO₃ is the possibility of obtaining "tailored" crystals for specific photorefractive applications. Doping the crystals with transition metal impurities is only one way to improve the photorefractive sensitivity of KNbO₃. Another way is the reduction of pure and doped KNbO₃ samples. Properties of reduced samples as well as the procedure itself
will be discussed in Sect.3. Figures 1.2 and 1.3 give a first illustration of such features. They show the intensity dependence of the refractive index change $\Delta n$ and the writing time constant $\tau$ for crystals with different Fe-concentration and chemical treatment.

![Diffraction efficiency and refractive index change vs. laser intensity of different KNbO$_3$ samples](image1.png)

**Fig.1.2** Diffraction efficiency and refractive index change vs. laser intensity of different KNbO$_3$ samples [1.4].

![Intensity dependence of the time constant $\tau$ for KNbO$_3$ :Fe](image2.png)

**Fig.1.3** Intensity dependence of the time constant $\tau$ for KNbO$_3$ :Fe [1.4].
Because of the exceptionally large electro-optic coefficients $r_{42}$ and $r_{51}$ of KNbO$_3$, an essential part of the present thesis deals with the discussion of the so-called anisotropic Bragg diffraction. In case of the special type of diffraction occurring in such photoinduced gratings, the polarization directions of the incident and diffracted linearly polarized light beams differ, i.e., they are orthogonal with respect to each other. As KNbO$_3$, being an optically biaxial crystal, possesses a relatively large birefringence, the incident and diffracted wave propagate with different velocities and have different absolute values of their wave vectors. This leads to very special diffraction conditions. In this thesis the occurring diffraction geometries are discussed and the pertinent experimental results are presented. In a theoretical treatment based on the "coupled wave theory", an analytical solution of the diffraction efficiency for weak violations of the Bragg condition is deduced and discussed.

One of the objectives of the present work was the design and investigation of an optically addressable light deflector. In Sect. 6 we show that the anisotropic Bragg diffraction in a special, non-critical configuration is ideally suited for such an application. In a second application, discussed in Sect. 7, this type of light deflection is used for the design of an optically addressable fiber switch.

A further application of anisotropic Bragg diffraction using a special writing geometry is discussed in Sect. 8. In these experiments the writing beams are diffracted by the self-induced grating. In contrast to the case of isotropic diffraction, the deflected beam differs both in direction and polarization with respect to the writing beams. This so-called anisotropic self-diffraction is used for designing an optically addressable spatial light modulator or incoherent-to-coherent converter. In this application, in addition to the two writing beams, a two-dimensional image (information) is projected into the crystal. Thereby the photoinduced grating amplitude is changed in such a manner that in the bright zones of the projection the diffraction efficiency is decreased because of a local increase of the photoconductivity. In such a way the image can be transferred with inverted contrast from the incoherent beam onto the self-diffracted beam. A simple theoretical model describing the intensity transfer characteristics is proposed for this process, and the factors limiting the resolution are examined. Experiments with reduced KNbO$_3$ single crystals led to resolutions of up to 10 line pairs per millimeter and conversion times of the order of 1 ms.
2. The Photorefractive Effect

2.1 Band Model with one type of carriers

The model able to describe most of the experimental results was derived by KUKHTAREV et al. [1,20]. It is a charge transport model assuming that photo-excited electrons (or holes) are ejected from filled (or empty) donor sites into the conduction (or valence) band. There they migrate by diffusion, under the action of an electric field (drift) or due to the photovoltaic effect to dark regions and are subsequently retrapped by the empty (or filled) donors. For inhomogeneous illumination the migration of charge carriers leads to regions of unneutralized charge, that can persist after the light has been removed. The charge separation results in a space charge-field which modulates the refractive index via the linear electro optic effect. The whole process is summarized in Figs. 2.1 and 2.2.

![Fig.2.1 Schematic representation of the band structure of a simple photorefractive material. The acceptors $N_A$ are assumed to be completely ionized. They do not contribute to the charge field build up due to a light induced redistribution of the empty donor centers.](image-url)
Fig. 2.2 Formation of a refractive index grating due to the photorefractive effect.
For the first set of basic equations we assume the existence of only one photoactive species (holes or electrons). In such a case the band transport model is described by

(a) The continuity equation for the mobile carriers with charge \( q \)

\[
\frac{\partial n}{\partial t} = g - r - \frac{1}{q} \nabla \cdot \mathbf{J} \tag{2.1}
\]

where \( n \) is the number density of free charge carriers, \( t \) the time, \( g \) the rate at which free carriers are created and \( r \) the rate at which these carriers are retrapped. \( q \) is the charge of the free carriers (negative for electrons) and \( \mathbf{J} \) the electronic current density.

The current density \( \mathbf{J} \) is the sum of diffusion, drift and photovoltaic components given by

\[
\mathbf{J} = e \mu n \mathbf{E} - q D \nabla n + \mathbf{J}^{ph} \tag{2.2}
\]

where \( e \) is the elementary charge, \( \mu \) the mobility and \( \mathbf{E} \) the electric field. \( D \) is the diffusion constant and \( \mathbf{J}^{ph} \) the photovoltaic current.

(b) The continuity equation for the immobile ionized donors (rate equation):

\[
\frac{\partial N_D^i}{\partial t} = g - r \tag{2.3}
\]

\( N_D^i \) is the density of ionized donors.

The generation rate is linearly proportional to the number of occupied donors:

\[
g = (N_D - N_D^i) \cdot (sI + \beta) \tag{2.4}
\]

where \( N_D \) is the total number density of donors, \( s \) the normalized cross section for photoionization, \( I \) the light intensity and \( \beta \) the probability rate of thermal excitation.
The retrapping rate is linearly proportional to the number of ionized donors:

\[ r = \gamma_R N_D^I n \]  

(2.5)

\( \gamma_R \) is the carrier recombination constant.

(c) Poisson's law (\( \nabla \cdot D = \rho \)):

\[ \nabla (\varepsilon \varepsilon_0 E) = q(n - N_D^I + N_A) \]  

(2.6)

where \( \varepsilon \) is the static unclamped dielectric tensor and \( N_A \) is the density of acceptors.

The density \( N_A \) is the number density of charges that compensate for the charge of \( N_D^I \) under dark conditions (it is assumed that \( N_D \gg N_A \)). These compensative acceptor levels are filled completely and are not involved in phototransitions. They enable part of the donors to be ionized even in the dark.

Neglecting the photovoltaic effect, the charge transport in a photorefractive material is described by the following equations:

\[ \frac{\partial N_D^I}{\partial t} = (N_D - N_D^I)(s I + \beta) - \gamma_R N_D^I n \]  

(2.7)

\[ \frac{\partial n}{\partial t} = \frac{\partial N_D^I}{\partial t} + \nabla \cdot (D \nabla n - \frac{E}{\varepsilon} \mu n E) \]  

(2.8)

\[ \nabla \cdot (\varepsilon \varepsilon_0 E) = q(n - N_D^I + N_A) \]  

(2.9)

Equations (2.7)-(2.9) together with appropriate initial and boundary conditions describe the material response on an inhomogeneous illumination.

The light intensity distribution is created by a light field \( \bar{E} \) which is present in the crystal. The interaction of this light field with the photorefractive material is described by the wave equation:

\[ \Delta E(r,t) - \nabla (\nabla \cdot E(r,t)) - \mu_0 \varepsilon_0 \varepsilon \frac{\partial^2 E}{\partial t^2} = 0 \]  

(2.10)
where it is assumed that the medium is non-magnetic ($\mu=1$) and source free ($\rho \approx 0$, $J \approx 0$).

We assume that the light field $\mathbf{E}$ is represented by a sum of plane waves

$$\mathbf{E}(r, t) = \sum_j \mathbf{E}_j(r) e^{-i(k_j r - \omega_j t)}$$

(2.11)

$\mathbf{E}_j$ is the amplitude of the $j$-th plane wave with a wave vector $k_j$ and an angular frequency $\omega_j$. The dielectric tensor $\mathbf{e}$ (at optical frequencies) in general is spatially modulated by the photorefractive effect and therefore acts as the coupling term of the different wave components $\mathbf{E}_j$.

2.1.1 Purely sinusoidal illumination in the steady state

The photorefractive material is illuminated by two coherent plane waves with wave vectors $k_{+1}$ and $k_{-1}$ (Fig. 2.3).

The light field in the crystal is then given by

$$\mathbf{E}(r, t) = \mathbf{E}_{+1}(r) e^{-i(k_{+1} r - \omega t)} + \mathbf{E}_{-1}(r) e^{-i(k_{-1} r - \omega t)}$$

(2.12)
The intensity distribution created by the interference of these two plane waves is

\[ I = \langle |\vec{E}|^2 \rangle_t = I_0 \left[ 1 + m \cos(K_g r) \right] \tag{2.13} \]

\( \langle \rangle_t \) is the time average. \( I_0 = |E_+|^2 + |E_-|^2 = I_{+1} + I_{-1} \) is the average intensity and \( m = 2 \cdot \sqrt{I_{+1} I_{-1}} / I_0 \) is the modulation ratio. The fringe pattern wave number \( K_g \) is given by

\[ K_g = k_{+1} - k_{-1} \tag{2.14} \]

\[ |K_g| = 2\pi / \Lambda = 4\pi \sin(\Theta') / \lambda_0 \] where \( \Lambda \) is the fringe spacing of the intensity grating, \( 2\Theta' \) the angle between the two writing beams (outside the crystal) with a vacuum wavelength \( \lambda_0 \).

We assume the number density of the free charge carriers to be at any time much smaller than the trap density \( n \ll N_A \) and than the density of unionized donor centers \( n \ll (N_D - N_A) \). Moreover we assume a linear generation and recombination rate (i.e. \( N_D \approx N_A \)). Then, in the steady state, Eq.(2.7) reduces to

\[ n = n_0 \left[ 1 + M \cos(K_g y) \right] \tag{2.15} \]

where \( K_g \) is assumed to point into the y-direction (Fig.2.3) and \( n_0 \) is the mean charge carrier density

\[ n_0 = g_0 \tau_R \tag{2.16} \]

where the generation rate \( g_0 \) is given by

\[ g_0 = (N_D - N_A) (s \lambda_0 + \beta) \tag{2.17} \]

and the free carrier lifetime

\[ \tau_R = (N_A \gamma_R)^{-1} \tag{2.18} \]

\( M \) is the reduced fringe contrast

\[ M = \frac{m}{1 + \beta / (s\lambda_0)} \tag{2.19} \]

which is reduced due to the presence of free carriers even in the dark.
In the steady state (2.8) reduces to

$$\frac{\delta j}{\delta y} = \phi (D \frac{\delta n}{\delta y} - \frac{e}{q} \mu n E) = 0 \quad (2.20)$$

The integration of this equation together with Eq.(2.15) gives finally

$$E = \frac{j}{e \mu n_o} \cdot \frac{1}{1 + M \cos(K_y)} - \frac{q DK_g}{e} \cdot \frac{M \sin(K_y)}{1 + M \cos(K_y)} \quad (2.21)$$

where \( j \) is the free carrier current density.

For a boundary condition of constant applied voltage \( V \) over the crystal length \( L \) we must have

$$E_o = \frac{V}{L} = \frac{1}{L} \int_0^L E dy \quad (2.22)$$

By integration of Eq.(2.21) over a large number of grating periods, we are able to find an expression for the current density \( j \)

$$j = \sqrt{1 - M^2} e \mu n_o E_o \quad (2.23)$$

The conductivity for sinusoidal illumination is reduced by a factor \( \sqrt{1 - M^2} \) compared to the conductivity for uniform illumination at the same average intensity.

Finally we can rewrite Eq.(2.21) in the form

$$E = E_o \frac{\sqrt{1 - M^2}}{1 + M \cos(K_y)} - \frac{M \sin(K_y)}{1 + M \cos(K_y)} \quad (2.24)$$

\( E_d \) is the diffusion field given by

$$E_d = \frac{q DK_g}{e} = \frac{q}{e} \left( \frac{k_g T}{e} \right) K_g \quad (2.25)$$
where the last equation is obtained by the Einstein relation, $k_B$ is the Boltzmann constant and $T$ the temperature.

The final equation is an expression for the photoinduced space charge field

$$E_{sc} = E - E_0 = E_0 \left( \frac{\sqrt{1 - M^2}}{1 + M \cos(K_g y)} - 1 \right) - E_d \frac{M \sin(K_g y)}{1 + M \cos(K_g y)}$$

(2.26)

Fig.2.4 Electric field distribution along one grating period for different modulation ratios $m$: a) for diffusion  b) for drift
Due to the presence of dark conductivity even for a modulation ratio $m=1$ the reduced fringe contrast $M$ is always smaller than unity (see Eq. 2.19). Therefore the space-charge field $E_{sc}$ physically can never reach infinity.

Figure 2.4 shows examples of Eq. (2.26) for the two cases where diffusion is dominant ($E_O \ll E_d$) and where the external field $E_O$ is much larger than the diffusion field ($E_O > E_d$).

For later investigations of first order Bragg diffraction by such photoinduced phase gratings it is useful to expand the space-charge field Eq. (2.26) as a Fourier series

$$E_{sc} = 2 \sqrt{E_o^2 + E_d^2} \sum_{j=1}^{\infty} \left[ \sqrt{\frac{1}{M^2} - 1} - \frac{1}{M} \right]^j \cos(jK_g y - \varphi)$$

with

$$\tan(\varphi) = \frac{E_d}{E_o}$$

From Eq. (2.27) it seems possible that the space-charge field could reach arbitrarily large values for increasing applied fields $E_O$ or decreasing fringe spacing $A$ (increasing $E_d$). In reality the Poisson law Eq. (2.9) limits the space charge field due to the fact that only a finite number of traps are available.

Although a general analytic solution of the system of coupled differential equations (2.7) - (2.9) is not possible for sinusoidal illumination, a solution can be found in a linear approximation. For small modulation ratios $m$ such an approximation is justified as can be seen from Eq. (2.27). We can write in this case

$$I = I_o \left[ 1 + \text{Re}(m e^{iK_g y}) \right]$$

$$n = n_o \left[ 1 + \text{Re}(m e^{iK_g y}) \right]$$

$$E = E_o + \text{Re}(E_{sc} e^{iK_g y})$$

$$N_{D} = N_{D_o} + \text{Re}(N_{D_i} e^{iK_g y})$$

Setting these expressions into the differential equations (2.7) - (2.9) we
obtain a formula for the space charge field $E_{sc}^0$ by equating the constant terms and the coefficients of $e^{K_x Y}$

$$E_{sc}^0 = \frac{m}{1 + \beta/s_0}$$

\[ \left[ \left( s_0 + \beta \right) \frac{N_D}{N_D^o} \frac{1}{Y_R} + 1 \right] \frac{1}{iE_o - E_o} + \frac{1}{iE_q} \tag{2.33} \]

where $E_d$ is the diffusion field given by (2.25) and $E_q$ is given by

$$E_q = \frac{q}{\varepsilon e_o} \frac{1}{K_g} \frac{N_A}{N_D} (N_D - N_A) \tag{2.34}$$

With the same assumptions leading to Eq.(2.15) ($N_D^o \approx N_A, n << N_D, n << N_N - N_A$) and assuming moreover that the thermal excitation is much smaller than the photonic excitation ($\beta << s_0$) we find

$$E_{sc}^0 = m \frac{E_q (iE_d - E_o)}{(E_q + E_d) + iE_o} \tag{2.35}$$

If no external field is applied this expression reduces to the well known formula

$$E_{sc}^0 = i m \frac{E_q E_d}{E_q + E_d} \tag{2.36}$$

From here it is obvious why $E_q$ is also called the "limiting" field. $E_{sc}^0$ can never become larger than $E_q$ which is proportional to the acceptor concentration $N_A$.

For an applied electric field which is much smaller than the possible internal fields ($E_o << (E_q + E_d)$) we find

$$E_{sc}^0 = -m \frac{E_o E_q^2}{(E_q + E_d)^2} + i m \frac{E_d E_q}{E_d + E_q} \tag{2.37}$$
For typical photorefractive parameters Fig. 2.5 illustrates the dependence of the photoinduced space-charge field on an externally applied electric field $E_0$ Eq. (2.35).

![Graph showing dependence of the photoinduced space-charge field on applied electric field $E_0$.]

$\Lambda = 10 \mu m$
$N_A = 10^{16} \text{cm}^{-3}$
$m = 0.1$

![Graph showing dependence of the photoinduced space-charge field on applied electric field $E_0$.]

$\Lambda = 1 \mu m$
$N_A = 10^{16} \text{cm}^{-3}$
$m = 0.1$

Fig. 2.5 Dependence of the photoinduced space-charge field on an externally applied electric field $E_0$ for typical photorefractive parameters.
2.1.2 Dynamic behaviour of the photoinduced space-charge field

Although a general analytic solution of the KUKHTAREV-equations (2.7)-(2.9) for the time evolution of the photoinduced space-charge field is not obtainable, solutions can be found under certain assumptions.

If the light intensity is spatially constant then \( N_D, n \) and \( E \) must also be spatially constant by symmetry reasons. Eq.(2.9) then gives

\[
\frac{\partial n}{\partial t} = (N_D - n - N_A) \cdot (s_1 + \beta) - \gamma_R (n + N_A) n
\]  

(2.38)

Substituting this equation into Eq.(2.7) leads to

\[
\frac{\partial n}{\partial t} = (N_D - N_A) \cdot (s_1 + \beta) - \gamma_R n N_A
\]  

(2.39)

With the assumption \( n \ll (N_D - N_A) \cdot N_A \) stated already in section 2.1.1 we find the differential equation

\[
\frac{\partial n}{\partial t} = (N_D - N_A) \cdot (s_1 + \beta) - \gamma_R n N_A
\]  

(2.40)

The initial conditions

\[
l = 0 \quad t < 0 \\
l = l_o \quad t > 0
\]

lead to the solution

\[
n = \tau_R (N_D - N_A) \cdot \left[ \beta + s_1 (1 - e^{-t/\tau_R}) \right]
\]  

(2.41)

where \( \tau_R \) is the recombination time already given in Eq.(2.18).

A measurement of the time dependence of the photocurrent \( j = \varepsilon \mu n E \) delivers experimental data of the recombination time \( \tau_R \). Typical values of \( \tau_R \) found are \( 10^{-5}s \) for BSO and \( 10^{-9}s \) for BaTiO_3 [2.1].

For sinusoidal illumination a temporal solution can be found in a linear approximation similar to Eqs.(2.29) - (2.32)

\[
l = l_o \left[ 1 + Re(\varepsilon \mu n_0(t) e^{i \omega t}) \right]
\]  

(2.42)

\[
n = n_o(t) + Re(n(t) e^{i \omega t})
\]  

(2.43)
\[ E = E_0 + \text{Re}(E_{sc}(t)e^{iK_0y}) \]  
\[ N_0 = N_0(t) + \text{Re}(N_1(t)e^{iK_0y}) \]

It is assumed that the spatially constant quantities are time independent in Eqs. (2.42)-(2.45). This means that for low power c.w. operation the excitation of the carriers occurs on a much shorter time scale than the emergence of the grating (for high power pulsed operation this assumption is no longer valid). The same argument makes it possible to neglect the time derivative of \( n_0 \) in Eq. (2.40) \( (\partial n_0/\partial t \approx 0) \). These assumptions are also called the "KUKHTAREV's quasi steady state approximations".

Substituting Eqs. (2.42)-(2.45) into Eqs. (2.7)-(2.9) together with the above mentioned assumptions, we obtain equations in which we can neglect all terms of quadratic or higher order in the oscillating increments. This is justified if we assume the magnitude \( m \) of the optical modulation to be much smaller than unity \( (m \ll 1) \).

The zero-th order free-carrier density is then

\[ n_0 = \frac{N_D - N_0}{\gamma R N_0} (s l_0 + \beta) \]  
(2.46)

By algebraic manipulation we get a second order differential equation for the time evolution of the space-charge field \( E_{sc}(t) \)

\[ \frac{\partial^2 E_{sc}}{\partial t^2} + \frac{\partial E_{sc}}{\partial t} \left[ \frac{1}{\tau_{dl}} + i \frac{e}{q \tau_E} + \frac{1}{\tau_D} + \frac{1}{\tau_I} + \frac{1}{\tau_R} \right] + E_{sc} \left[ i \frac{e}{q \tau_E} + \frac{1}{\tau_D \tau_I} + \frac{1}{\tau_{dl} \tau_I} + \frac{1}{\tau_{dl} \tau_R} \right] = i(N_D - N_A) \frac{\text{sm} l_0}{K g \varepsilon_0} \left[ \frac{e}{q \tau_E} + \frac{1}{\tau_D} \right] \]

(2.47)

where

\[ \tau_{dl} = \frac{\varepsilon\varepsilon_0}{e\mu n_0} \quad \text{(dielectric relaxation time)} \]  
(2.48)

\[ \tau_E = \frac{1}{K g \mu E_0} \quad \text{(drift time)} \]  
(2.49)
\[ \tau_D = \frac{e}{\mu k_B T k_g^2} \quad \text{(diffusion time)} \]  
\[ \tau_R = \frac{1}{\gamma_R n_A} \quad \text{(recombination time)} \]  
\[ \tau_I = \frac{1}{s l_o + \beta + \gamma_R n_o} \quad \text{(inverse of production and recombination rate)} \]

With
\[ \frac{1}{T_1} = \frac{1}{\tau_{dl}} + \frac{e}{q} \frac{1}{\tau_E} + \frac{1}{\tau_D} + \frac{1}{\tau_I} + \frac{1}{\tau_R} \]  
\[ \frac{1}{T_2} = \frac{e}{q} \frac{1}{\tau_E \tau_I} + \frac{1}{\tau_D \tau_I} + \frac{1}{\tau_{dl} \tau_I} + \frac{1}{\tau_{dl} \tau_R} \]

\[ C = i(N_D - N_A) \frac{s m l_o q}{k_g e e_o} \left[ \frac{e}{q} \frac{1}{\tau_E} + \frac{1}{\tau_D} \right] \]

Eq. (2.47) reduces to
\[ \frac{\delta^2 E_{sc}}{\delta t^2} + \frac{1}{T_1} \frac{\delta E_{sc}}{\delta t} + \frac{E_{sc}}{T_2} = C \]

In the stationary case \((\delta E_{sc}/\delta t=0)\) the solution of Eq. (2.55) corresponds to Eq. (2.33) and Eq. (2.35). The general time dependent solution of Eq. (2.55) is
\[ E_{sc} = E_1 e^{t/\tau_1} + E_2 e^{t/\tau_2} + CT_2^2 \]

where
\[ E_{sc}(0) - T_2^2 C - \tau_{2,1} \frac{\delta E_{sc}}{\delta t} \bigg|_{t=0} \]
\[ E_{1,2} = \frac{1}{1 - \tau_{2,1}/\tau_{1,2}} \]

and the exponential time constants are given by
\[ \frac{1}{\tau_{1,2}} = \frac{1}{2} \left[ \frac{1}{\tau_1} + \sqrt{\frac{1}{\tau_1^2} - \frac{4}{T_2^2}} \right] \]
\[ E_{sc}(0) \text{ and } \left. \frac{\partial E_{sc}}{\partial t} \right|_{t=0} \text{ are the initial conditions:} \]

\[ E_{sc}(0) = 0 \quad \text{(for writing)} \]
\[ E_{sc}(0) = E_{sc}^0 \quad \text{(for erasure)} \]

The time derivative of the space-charge field at time \( t=0 \) can be obtained through Eq. (2.8) and Eq. (2.9)

\[ \left. \frac{\partial E_{sc}}{\partial t} \right|_{t=0} = \frac{1}{\tau_{dl}} \left[ E_{sc}(0) + E_{sc}^0 \frac{n_1}{n_0} - i E_d \frac{n_1}{n_0} \right] \quad (2.59) \]

On the other hand it is easy to see with Eq. (2.58) that the real part of \( 1/\tau_1 \) is larger than \( 1/T_1 \). Therefore \( \tau_1 \) is smaller than any time constant occurring in Eq. (2.52) and is in particular smaller than \( \tau_R \) (\( \tau_i < \tau_R \)). According to our "quasi steady state approximation" we are not interested in such fast transients, and as a consequence we can neglect the second order derivatives in Eq. (2.55). For the slow time constant \( \tau_2 \) we then get

\[ \frac{1}{\tau_2} = -\frac{T_1}{T_2} = -\frac{1}{\tau} + i \omega \quad (2.60) \]

and

\[ E_{sc}(t) = E_2 e^{-t/\tau} e^{i\omega t} + C T_2^2 \quad (2.61) \]

With the assumption that the number density of the free charges \( n_0 \) is at any time much smaller than the trap density \( N_A \) (\( n_0 \ll N_A \)) and with the help of Eq. (2.46) it is easy to see that \( \tau_1 \gg \tau_R \). In Eq. (2.53) and Eq. (2.54), therefore, \( 1/\tau_1 \) can be neglected with respect to \( 1/\tau_R \). With this the real and imaginary part of Eq. (2.60) is

\[ \tau = \tau_{dl} \left[ \frac{1 + \frac{\tau_R}{\tau_{dl}} + \frac{\tau_R}{\tau_D}}{1 + \frac{\tau_R}{\tau_{dl}} + \frac{\tau_R}{\tau_D}} \right]^2 \quad (2.62) \]
Several features of Eqs. (2.62) and (2.63) are interesting. First, it is obvious that the time constant $\tau$ is a complicated function of the grating period, applied electric field strength $E_0$, temperature $T$ etc. In the following we will consider the case where an applied electric field $E_0$ is present and the case without any applied field.

i) With applied electric field $E_0$

The time constant $\tau_2$ has a complex value, i.e. the transient part of the space charge field moves with a velocity

$$v = \frac{\omega}{K_g}$$

(2.64)

The direction of the movement depends on the sign of the free charge carriers $e/q$.

After switching off the recording beams the whole space-charge field moves with the velocity $v$. To get the maximum amplitude of a photoinduced phase grating, it is necessary to move the intensity fringes with the calculated velocity.

The largest enhancement obtained by moving the fringes may then be interpreted as maximum interaction occurring when the grating velocity is equal to the fringe velocity [2.2].

As can be seen from Eq. (2.62) also the real part of the time constant is affected by an applied field. By analyzing this equation one finds that in general an applied field increases the value of the time constant $\tau$.

ii) Without applied electric field $E_0 = 0$

In this case Eqs. (2.62) and (2.63) reduce to

$$\tau = \frac{\tau_{dl} + \tau_D}{1 + \frac{\tau_{dl}}{\tau_D}}$$

(2.65)

$$\omega = 0$$

(2.66)
The time constant is given by the dielectric relaxation time only in the case where $\tau_R$ is much shorter than $\tau_{dl}$ (small diffusion due to low temperature or large grating period). Further one should keep in mind that the parameter $\tau_{dl}/\tau_1$ controls a separate regime for the erasure time. $\tau_{dl}/\tau_1$ is usually but not necessarily much less than unity. If $\tau_{dl}/\tau_1 \gg 1$ and $\tau_R/\tau_D \gg 1$ then $\tau \approx \tau_1$ and therefore $\tau$ is independent of $\tau_{dl}$.

Whereas the dielectric limit has soon been recognized to be a limit of the photorefractive time constant, the limiting $\tau_1$ has not been extensively taken into consideration up to now.

In the following we will discuss this limit given by the inverse of the production and recombination rate $\tau_1$ defined in Eq.(2.51)

$$\tau_1 = \frac{1}{s \lambda_o + \beta + \gamma_R n_o} \quad \text{(Inverse of production and recombination rate)} \quad (2.51)$$

We will assume the photoconductivity to be much higher than the dark conductivity and, therefore, we can neglect $\beta$ ($\beta \ll s \lambda_o$). Then Eq.(2.51) is reduced to

$$\tau_{lim} = \frac{1}{s \lambda + \gamma_R n_o} \quad (2.67)$$

which we will call "limiting time" $\tau_{lim}$.

The expression for the photoconductivity, to be discussed in Sect.3.3.1, is

$$\sigma_{ph} = e n o \mu = e \mu \frac{\alpha \lambda}{h \nu} \tau_R \quad (2.68)$$

$\alpha$ is the absorption constant and $\nu$ the light frequency. With Eq.(2.16) we find an expression for the cross section of photoionizations

$$s = \frac{\alpha \Phi}{h \nu N_D - N_A} \quad (2.69)$$
Together with Eq.(2.18) we find an expression for the limiting time

$$\tau_{\text{lim}} = \frac{\hbar \nu}{1 \alpha \Phi} \frac{N_A}{N_D} (N_D - N_A)$$

(2.70)

This time constant can be interpreted as the time needed to photoexcite $(N_D - N_A)N_A/N_D$ charge carriers. This is the number needed to build up the limiting space-charge field $E_q$ Eq.(2.34).

2.2 Band model with two types of carriers

The free charge carriers in photorefractive materials have been found to be electrons or holes depending on the material and its reduction state [1.21]. Experiments have been reported which lead to the conclusion that under certain conditions simultaneously electron and hole transport may be responsible for the photorefractive effect [2.3-2.5].

Theoretical descriptions of these phenomena have been given in form of an extension of the KUKHTAREV single species model [1.20] developed by KUKHTAREV [2.6], VALLEY [2.7] and STROHKENDL et al. [2.8].

In this Section I will discuss the two simplest two-species models schematically shown in Fig.2.6. In addition a third model with two different types of photorefractive active centers will be discussed.

Model 1

In this model, shown in Fig.2.6a, electrons are ionized from the filled traps $(N_D - N_D^+)$ into the conduction band, move by drift and diffusion, and recombine with empty traps $N_D^-$ while holes are photoexcited from acceptors $N_D^+$ and recombine with $(N_D - N_D^+)$. This model corresponds to the model developed by STROHKENDL et al. [2.8] and to the model 1 by VALLEY [2.7].

Corresponding to Eqs.(2.7)-(2.9) in this model we find

$$\frac{\partial N_D^+}{\partial t} = (s_e + \beta_e) (N_D - N_D^+) - \gamma_e n_e N_D^+ - (s_h + \beta_h) N_D^+ + \gamma_h n_h (N_D - N_D^+)$$

(2.71)
Fig. 2.6 a) Model 1: simultaneous electron and hole transport with a single set of recombination centers. Electrons are photoionized from filled donors and retrapped from ionized traps.

b) Model 2: simultaneous electron and hole transport with two sets of recombination centers. The rates and populations of the level with density $N_D$ are such that electrons dominate while holes dominate the level with density $N_A$. 
\[ j_e = e n_e \mu_e E + \mu_e k_B T \nabla n_e \]  
\[ j_h = e n_h \mu_h E - \mu_h k_B T \nabla n_h \]  
\[ \frac{\partial n_e}{\partial t} = \frac{1}{e} \nabla j_e + (s_e, \beta_e)(N_D - N_D^+) - \gamma_e n_e N_D^+ \]  
\[ \frac{\partial n_h}{\partial t} = \frac{1}{e} \nabla j_h + (s_h, \beta_h) N_D^+ - \gamma_h n_h (N_D - N_D^+) \]  
\[ \nabla (\epsilon \varepsilon_0 E) = -e (n_e + N_A - N_D - n_h) \]  

\( n_e \) and \( n_d \) are the electron and hole number density, \( N_D \) is the donor number density, \( N_D^+ \) is the density of ionized donors and \( N_A \) is the density of negative ions that compensate the charge of \( N_D^+ \) in the dark. \( j_e, h, \mu_e, h, \gamma_e, h, s_e, h, \) and \( \beta_e, h \) are the current density, mobility, recombination rate, photo cross section, and dark generation rate for electrons and holes respectively.

In a linear approximation similar to Eqs.(2.42)-(2.45) the solution of Eqs.(2.71)-(2.74) follows the same steps as discussed in Sect. 2.1 for the single carrier model.

The zeroth-order equations for the mean electron and hole densities can be written

\[ n_{eo} = \frac{N_D - N_A}{\gamma_e N_A} (s_e I_0 + \beta_e) \]  
\[ n_{ho} = \frac{N_A}{\gamma_h (N_D - N_A)} (s_h I_0 + \beta_h) \]  

The characteristic time constants similar to the ones in Eqs.(2.48)-(2.51) for electrons (e) and holes (h) are

\[ \tau_{die} = \frac{\epsilon \varepsilon_0}{e \mu_e n_{eo}} \quad \tau_{dih} = \frac{\epsilon \varepsilon_0}{e \mu_h n_{ho}} \]  
\[ \tau_{Ee} = \frac{1}{K_g \mu_e E_0} \quad \tau_{Eh} = \frac{1}{K_g \mu_h E_0} \]
The steady state space-charge field without applied electric field is given by

\[ E_{sc} = -i \frac{E_q E_d}{E_q + E_d} \cdot \frac{E_q (\tau_{\text{dih}} - \tau_{\text{die}}) + E_d (\tau_{\text{lh}} - \tau_{\text{le}})}{E_q (\tau_{\text{dih}} + \tau_{\text{die}}) + E_d (\tau_{\text{lh}} + \tau_{\text{le}})} \]  

(2.81)

Where \( E_q \) is the diffusion field given by Eq.(2.25) and \( E_q \) the limiting space-charge field given by (2.34). In the case where \( E_q (\tau_{\text{dih}} - \tau_{\text{die}}) \) is much larger than \( E_d (\tau_{\text{lh}} - \tau_{\text{le}}) \) Eq.(2.81) reduces to

\[ E_{sc} = -i \frac{E_q E_d}{E_q + E_d} \cdot \frac{\sigma_e - \sigma_h}{\sigma_e + \sigma_h} \]  

(2.82)

where \( \sigma_e = e \mu_e n_e \) and \( \sigma_h = e \mu_h n_h \) are the electron and hole conductivities respectively. Eq.(2.82) illustrates the fact that there is no net diffusion to create a grating, when electron and hole conductivities are equal.

In the other case where \( E_q (\tau_{\text{dih}} - \tau_{\text{die}}) \) is much smaller than \( E_d (\tau_{\text{lh}} - \tau_{\text{le}}) \) Eq.(2.79) reduces to

\[ E_{sc} = -i \frac{E_q E_d}{E_q + E_d} \cdot \frac{\Phi_e - \Phi_h}{\Phi_e + \Phi_h} \]  

(2.83)

where \( \Phi_e \) and \( \Phi_h \) are quantum efficiencies for creating a free electron and a free hole respectively. This limit can be found for very small grating spacings (\( E_q \ll E_d \)) or in the case where \( \tau_l \approx \tau_d \).

If the inverse of the carrier production rate and the dielectric relaxation time are large as compared to the diffusion and recombination time the model leads to an exponential increase of the photoinduced space-charge field.
as a function of time with a time constant \( \tau \) given by

\[
\tau = \frac{E_q E_d}{\varepsilon_d \varepsilon_{inh}} \frac{1 + \frac{\tau_{Re}}{\tau_{De}}}{1 + \frac{\tau_{Rh}}{\tau_{De}}}
\]

(2.84)

A more general case (without the last assumptions of large inverse of the production rate and dielectric relaxation time compared to the diffusion and recombination time and with an applied electric field) also leads to a simple exponential increase of the space-charge field with a single time constant [2.8]. This model is, therefore, not able to explain experiments in materials where two or more different time constants are observed [2.3].

Model 2

In this model, illustrated in Fig.2.6b, there are two independent sets of photo-active species, one with electrons and the other one with holes as the dominant charge carriers. This model was first derived by KUKHTAREV [2.5] for the steady state and later on the dynamic behaviour was discussed by VALLEY [2.7].

Corresponding to Eqs. (2.7)-(2.9) we find the starting equations in this model to be

\[
\begin{align*}
\frac{\partial N_D^+}{\partial t} &= (s_e^+ + \beta_e^-)(N_D^- - N_D^+) - \gamma_e n_e N_D^+ \\
\frac{\partial N_A^-}{\partial t} &= (s_h^+ + \beta_h^-)(N_A^- - N_A^+) - \gamma_h n_h N_A^- \\
J_e &= e n_e \mu_e E + \mu_e k_B T \nabla n_e \\
J_h &= e n_h \mu_h E - \mu_h k_B T \nabla n_h \\
\frac{\partial n_e}{\partial t} &= \frac{1}{e} \nabla J_e + (s_e^+ + \beta_e^-)(N_D^- - N_D^+) - \gamma_e n_e N_D^+ \\
\frac{\partial n_h}{\partial t} &= -\frac{1}{e} \nabla J_h + (s_h^+ + \beta_h^-)(N_A^- - N_A^+) - \gamma_h n_h N_A^- \\
\nabla (\varepsilon \varepsilon_0 E) &= -e(n_e - n_h - N_D^+ + N_A^- - \Delta N)
\end{align*}
\]

(2.85) (2.86) (2.87) (2.88)
where \( \Delta N \) is the difference \( N_D^+ - N_A^- \) in the absence of radiation. By using the same approximation as in the one charge carrier model in Sect.2.2 (Eqs.2.29-2.32) one can find closed form solutions for the steady-state and for transient cases.

The zeroth-order equations for the mean electron and hole densities can be written

\[
\begin{align*}
    n_{eo} &= \frac{N_D - N_D^+}{\gamma_e N_{Do}^+} (s_e l_o + \beta_e) \\
    n_{ho} &= \frac{N_A - N_A^-}{\gamma_h N_{Ao}^-} (s_h l_o + \beta_h)
\end{align*}
\] (2.89)

The steady state space-charge field without any applied electric field is then given by

\[
E_{sc} = -\text{Im} \frac{E^e - E^h}{E^e + E^h} \quad (2.90)
\]

where \( E^e \) and \( E^h \) are the limiting space-charge fields for the donor and acceptor systems, respectively

\[
\begin{align*}
    E^e &= \frac{e N_{Do}^+}{\epsilon \varepsilon_0 K_g} \frac{N_D - N_{Do}^+}{N_D} \\
    E^h &= \frac{e N_{Ao}^-}{\epsilon \varepsilon_0 K_g} \frac{N_A - N_{Ao}^-}{N_A}
\end{align*}
\] (2.91) (2.92)

The important result of Eq.(2.90) is the fact that the sign of the space-charge field depends on the difference of the concentrations of the two un-ionized centers. In this model no change of sign can occur by changing the grating wave number \( K_g \).

The time constants assumed for this solution are the same as in model 1 (Eqs.(2.76-2.80)) with exception of the recombination times which in this model are given by

\[
\begin{align*}
    \tau_{Re} &= \frac{1}{\gamma_e N_{Do}^+} \\
    \tau_{Rh} &= \frac{1}{\gamma_h N_{Ao}^-}
\end{align*}
\] (2.93)
For the transient behavior an exponential time dependence can be found in which the two time constants $\tau_+$ and $\tau_-$ are the solutions of the following quadratic equation

$$
\left( \frac{1}{\tau_+} + \frac{1}{\tau_-} \right) \left( \frac{1}{\tau_+} + \frac{1}{\tau_-} \right) - \frac{1}{\tau_+ \tau_-} \frac{1}{\tau_+ \tau_-} = 0
$$

(2.94)

where $\tau_+$ and $\tau_-$ are the time constants in absence of $N_A$ and $N_D$, respectively similar to Eq.(2.65).

$$
\tau_+ = \frac{\tau_+ \tau_-}{\tau_+ \tau_-} \frac{\tau_+ \tau_-}{\tau_+ \tau_-} = 0
$$

(2.95)

$$
\tau_- = \frac{\tau_+ \tau_-}{\tau_+ \tau_-} \frac{\tau_+ \tau_-}{\tau_+ \tau_-} = 0
$$

(2.96)

Summary of results of the band transport models

Steady-state and transient response have been discussed for two models of a photorefractive material in which both electron and hole transport are important. As discussed in Sect.2.1 the sign of the space-charge field of a photoinduced grating depends on the sign of the dominant charge carrier.

The results obtained show that if a single set of recombination centers is responsible for both electrons and holes, then the carriers with the larger conductivity dominate for large grating periods, while the carriers with the larger quantum efficiency dominate for small grating periods. On the other hand, if there are two sets of recombination centers, the sign of the space-charge field is given by the sign of the carrier with the larger empty trap density.

There is a simple exponential increase of the space-charge field as a function of time for the single-recombination center model and the single time constant is inversely proportional to the intensity. For the two-center model two different response times which are inversely proportional to the intensity
can exist.

Accordingly, experiments in which both a change of the sign of the space-charge field and two different time constants were observed, cannot be explained by one of these two models. It is, however, easy to imagine that a combination of these models could explain such a behaviour (both centers in model 2 can generate electrons and holes and therefore according to model 1 both centers show a compensating point where the sign is changing). Nevertheless, the number of free parameters in such a model would be too large for a meaningful interpretation of the experimental results.
2.3 Bragg diffraction by photoinduced phase gratings

2.3.1 Introduction

Most of the crystals used for photorefractive applications are optically anisotropic at room temperature. Whereas the class of ferroelectric crystals (e.g. KNbO₃, BaTiO₃, LiNbO₃ and LiTaO₃) are strongly birefringent, the class of paraelectric crystals, like Bi₁₂SiO₂₀ (BSO) and Bi₁₂GeO₂₀ (BGO), are optically active but non-birefringent in the absence of an electric field.

For efficient diffraction by thick phase gratings one has to fulfill the Bragg condition. In the case of different polarization of the diffracted and incident optical wave (e.g. in an optically anisotropic crystal), the Bragg relation is more complicated than in the case of ordinary Bragg diffraction, where the diffraction angle is equal to the incidence angle.

We assume in the following treatment the dominant charge transport leading to the photoinduced space-charge field to be diffusion. In this case the space-charge field in a low modulation approximation (m<<1) is given by Eq.(2.36)

\[ E_{sc}(r) = \frac{E_d E_q}{E_d + E_q} \sin(K_y r) \]  
(2.100)

This field gives rise to a spatial modulation of the optical indicatrix (optical impermeability tensor \( \varepsilon^{-1} = n^{-2} \)) in the form

\[ \Delta(\frac{1}{n^2})_{ij}(r) = r_{ijk}E_{sc,k}(r) \]  
(2.101)

\( r_{ijk} \) is the electro-optic tensor (\( i,j,k \epsilon [x,y,z] \) where \( x,y,z \) are the main axes of the optical indicatrix).

From Eq.(2.101) we see that in a general case a space-charge field of the form (2.100) produces not only a spatially modulated deformation but also a modulated rotation of the optical index ellipsoid.

We will investigate diffraction gratings with ellipsoid rotations present. In such situations diffracted and incident waves have in general a different polarization. If the investigated material is also birefringent, these two waves
will propagate with different velocities, since they are subject to different refractive indices.

This situation is analogous to anisotropic diffraction in acousto-optics. Light diffracted by transverse ultrasonic waves in a birefringent medium will show in general anisotropic diffraction with the above mentioned features [2.9-2.13].

In photorefractive materials such index ellipsoid rotations occur if off-diagonal elements of the optical permittivity tensor ε are modulated (i≠j in Eq.(2.101)). Optically anisotropic photorefractive materials with electro-optic coefficients r_{ijk} with i≠j are compared with those of KNbO₃ in Table 2.1.

Table 2.1 Birefringent photorefractive materials for anisotropic Bragg diffraction (room temperature data at λ=633nm)

<table>
<thead>
<tr>
<th>Material</th>
<th>Point group</th>
<th>Electro-optic coefficient r_{ijk} (i≠j)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>KNbO₃</td>
<td>mm2</td>
<td>r_{51} = 105·10^{-12} m/V</td>
<td>[1.29]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>r_{42} = 380·10^{-12} m/V</td>
<td></td>
</tr>
<tr>
<td>LiNbO₃</td>
<td>3m</td>
<td>r_{42} = r_{51} = 33·10^{-12} m/V</td>
<td>[2.14]</td>
</tr>
<tr>
<td>LiTaO₃</td>
<td>3m</td>
<td>r_{42} = r_{51} = 20·10^{-12} m/V</td>
<td>[2.14]</td>
</tr>
<tr>
<td>BaTiO₃</td>
<td>4mm</td>
<td>r_{42} = r_{51} = 1640·10^{-12} m/V</td>
<td>[2.14]</td>
</tr>
</tbody>
</table>

2.3.2 Isotropic Bragg diffraction

Optimum light diffraction by refractive index gratings occurs if the Bragg condition is satisfied. This means that the grating wave vector is exactly equal to the difference between the incident and diffracted optical propagation vectors in the medium. Figure 2.7 shows the assumed diffraction geometry.

First we shall discuss the case of ordinary isotropic diffraction. In an isotropic medium, where the light velocities are independent of the propagation directions, the Bragg condition is determined easily.
In diffraction processes the laws of conservation of energy and momentum must be fulfilled (see Fig.2.8a), which leads to

\[ \omega_d = \omega_i \pm \Omega \quad (2.102) \]

\[ k_d = k_i \pm k_g \quad (2.103) \]

\( \omega_i, \omega_d \) are the frequencies and \( k_i, k_d \) are the wave vectors of the incident and diffracted waves, respectively (\( k_{i,d} = n_{i,d} \cdot 2\pi/\lambda_r; n_{i,d} \) stands for the effective refractive indices of incident, diffracted wave; \( \lambda_r \) is the vacuum wavelength of the read-out beam). \( \Omega \) is the oscillation frequency of the phase grating (in acousto-optics \( \Omega \) is equal to the ultrasonic frequency \( \omega_s \); for static photorefractive gratings \( \Omega = 0 \), for moving gratings \( \Omega \neq 0 \)).

Conservation of momentum Eq.(2.103) leads to the condition for the diffraction angle (see Fig.2.8a)

\[ \sin \theta_i = \sin \theta_d = \frac{\lambda_r}{2\Lambda_g n} \quad (2.104) \]

\( \theta_i, \theta_d \) are the incident and diffraction angles measured inside the crystal, \( \Lambda_g \) is the grating spacing and \( n \) is the refractive index. In optically isotropic materials \( k_i = k_d \) leads to \( \theta_i = \theta_d \).
Fig. 2.8  a) Wave vector diagram for isotropic Bragg diffraction
b) Wave vector dependence of incident and diffraction angles $\theta_i$ and $\theta_d$ for isotropic Bragg diffraction (for $n=2.2$)
Figure 2.8b shows the Bragg angles as a function of the relative grating wave number $K_g/k_r$ ($k_r=2\pi/\lambda_r$) for a material with a refractive index $n=2.2$. In the case of maximal $K_g$ the two wave vectors $k_i$ and $k_d$ are antiparallel ($K_g=2k_i=2k_d$), the incident wave is back-reflected by the phase grating.

### 2.3.3 Anisotropic Bragg diffraction in uniaxial crystals

First experiments on anisotropic Bragg diffraction in optically uniaxial crystals were performed by STEPANOV et al. [2.15] in LiNbO$_3$. They showed that this type of diffraction can improve the resolution of holographically recorded images when the read out beam has a different wavelength than that one used for recording [2.16].

When the diffracted optical polarization differs from the incident one in an anisotropic medium, the optical birefringence usually requires $k_d \neq k_i$. To derive a graphical method for determining the Bragg condition, let us rewrite Eq.(2.103) in the form

$$k_r n_d d = k_r n_i l \pm K_g$$  \hspace{1cm} (2.105)

where $l$ and $d$ are the unit vectors in the direction of the propagation vectors of the incident and diffracted waves. The refractive indices of the two waves $n_d, n_i$ are obtained as a function of the direction of the wave vector ($k_r=2\pi/\lambda_r$, where $\lambda_r$ is the vacuum wavelength of the incident beam).

![Wave vector surface for an optically uniaxial crystal](image)
Fig. 2.10  a) Wave vector diagram for anisotropic Bragg diffraction in an uniaxial crystal.

b) Wave vector dependence of incidence and diffraction angles $\theta_i$ and $\theta_d$ for anisotropic Bragg diffraction (for $n_o=2.287$ and $n_e=2.192$ as in LiNbO$_3$ for $\lambda_r=633\text{nm}$).
For a given direction of the wave vector we always have two propagation modes which are subject to different refractive indices. We can construct a three-dimensional surface with two shells, where the distance from the origin in a given wave vector direction corresponds to one of the two wave numbers $k_r n$. Figure 2.9 shows such a "wave vector surface" for an optically uniaxial crystal (e.g. LiNbO$_3$ or BaTiO$_3$ at room temperature).

With the help of these surfaces we are now able to construct the Bragg condition Eq.(2.105) geometrically. Any two points lying on these shells form with the center point 0 a triangle. Thus we can easily find the necessary diffraction parameters from arbitrary diffraction geometries.

Let us suppose, that the photoinduced grating vector $K_g$ is parallel to the $y$-axis (Fig.2.10a) and the light propagation and polarization direction of the incident waves lies in the $(x,y)$-plane. In this case a non-vanishing electro-optic tensor element of the type $r_{yz}$ gives rise to a diffracted wave polarized along the $z$-direction (e.g. $r_{42}$ in LiNbO$_3$ or BaTiO$_3$).

Figure 2.10a shows a central section of the wave vector surface perpendicular to the $z$-axis. It consists of two circles, the radii of which are $n_0 k_r$ and $n_e k_r$, respectively ($n_0$ ordinary and $n_e$ extraordinary refractive index of an optically uniaxial crystal).

From Fig.2.10b we see that there exists a lower limit for the grating wave number $K_{gm}^{min}$ below which no diffraction is possible. $k_i$ and $k_d$ are in the same direction whereas $K_g$ is antiparallel (Fig.2.11); the diffracted wave travels in the same direction as the incident wave but the polarizations of both waves are different:

$$K_{gm}^{min} = |n_0 - n_e| k_r$$

(2.106)

As in the isotropic Bragg diffraction - in the case of maximal grating wave number $K_g$ - the two wave vectors $k_i$ and $k_d$ are antiparallel. But in contrast to the isotropic case the back-reflected wave has changed its state of polarization.

Another special situation of great practical interest is that of a minimal input angle $\Theta_{imin}$ where the diffracted angle is exactly zero degrees. In this
case the grating wave number $K_g^*$ is given by

$$K_g^* = \sqrt{\left| n_o^2 - n_e^2 \right|} \cdot k_r$$  \hspace{1cm} (2.107)

This configuration is also shown in Fig. 2.11. It will be demonstrated later, that it can be applied to optically controllable light deflectors (see Section 6).

Fig. 2.11
Diffraction geometry for $K_{g\text{min}}$, the minimal grating wave number and $K_g^*$, the grating wave number for the minimal incidence angle $\theta_i^\text{min}$.

Table 2.2 shows the calculated grating spacings $\Lambda^{max}$ and $\Lambda^*$ corresponding to the two grating wave numbers $K_{g\text{min}}$ and $K_g^*$ respectively for LiNbO$_3$ and BaTiO$_3$ at room temperature.

2.3.4 Anisotropic Bragg diffraction in biaxial crystals

In biaxial crystals (orthorhombic, monoclinic and triclinic systems) the optical indicatrix is a triaxial ellipsoid. The three main axes correspond to the three main indices of refraction $n_1$, $n_2$ and $n_3$. In the orthorhombic system, which is considered in the following, the main axes of the indicatrix coincide with the crystallographic axes.

In this case a central section of the wave-vector surface perpendicular to the z-axis no longer consists of two circles. One curve is a circle with a
Fig. 2.12  a) Wave vector diagram for anisotropic Bragg diffraction in KNbO$_3$

b) Wave vector dependence of incidence and diffraction angle $\theta_i$ and

$\theta_d$ for anisotropic Bragg diffraction in KNbO$_3$ for active $r_{42}$. 
radius \( k_r n_z \) and the other curve is given by

\[
k_1 = k_r(\theta_i) = k_r \cdot n(\theta_i) = k_r \left( \frac{\cos^2 \theta_i}{n_y^2} + \frac{\sin^2 \theta_i}{n_x^2} \right)^{-1/2}
\]  
(2.108)

Figure 2.12b shows the Bragg angles as a function of the relative grating wavenumber \( K_g/k_r \) for optically biaxial \( \text{KNbO}_3 \) for a wavelength \( \lambda_r = 633\text{nm} \).

The Bragg angles must fulfill the following set of equations

\[
\frac{K_g}{k_r} = n(\theta_i) \cdot \sin(\theta_i) - n_z \cdot \sin(\theta_d)
\]  
(2.109)

and

\[
n_z \cdot \cos(\theta_d) = n(\theta_j) \cdot \cos(\theta_i)
\]  
(2.110)

The angles \( \theta_j \) and \( \theta_d \) outside the crystal are then given by Snell's law

\[
\sin(\theta_j) = n(\theta_j) \cdot \sin(\theta_i)
\]  
(2.111)

\[
\sin(\theta_d) = n_z \cdot \sin(\theta_d)
\]  
(2.112)

Table 2.2 Maximal grating spacing \( \Lambda_{\text{max}} \) and \( \Lambda^* \) needed for minimal input angle for anisotropic Bragg diffraction in the photorefractive materials of Table 2.1. (Refractive index data for \( \lambda = 633\text{nm} \) interpolated from [2.14] and for \( \text{KNbO}_3 \) from [1.28] respectively)

<table>
<thead>
<tr>
<th>Material</th>
<th>Refractive indices for ( \lambda = 633\text{nm} )</th>
<th>( \Lambda_{\text{max}} )(( \mu \text{m} ))</th>
<th>( \Lambda^* )(( \mu \text{m} ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{KNbO}_3 )</td>
<td>( n_1=2.279, n_2=2.329, n_3=2.168 )</td>
<td>5.7</td>
<td>0.76\textsuperscript{a}</td>
</tr>
<tr>
<td>( \text{LiNbO}_3 )</td>
<td>( n_1=n_2=n_o=2.287, n_3=n_e=2.192 )</td>
<td>3.9</td>
<td>0.88\textsuperscript{b}</td>
</tr>
<tr>
<td>( \text{LiTaO}_3 )</td>
<td>( n_1=n_2=n_o=2.177, n_3=n_e=2.182 )</td>
<td>144.0</td>
<td>5.57</td>
</tr>
<tr>
<td>( \text{BaTiO}_3 )</td>
<td>( n_1=n_2=n_o=2.404, n_3=n_e=2.316 )</td>
<td>14.7</td>
<td>1.40</td>
</tr>
</tbody>
</table>

\( \textsuperscript{a} \) for grating wave direction parallel to \( x_1 \) (eff. el. opt. coeff. \( r_{51} \))  
\( \textsuperscript{b} \) for grating wave direction parallel to \( x_2 \) (eff. el. opt. coeff. \( r_{42} \))
In Table 2.2 we have calculated the grating spacings \( \Lambda_{\text{max}} \) and \( \Lambda^* \) corresponding to the two grating wave vectors \( K_{g}^{\text{min}} \) and \( K_{g}^{*} \), respectively for \( r_{42} \) and \( r_{51} \) (active in KNBO\(_3\)) given by the formulas

\[
K_{g}^{\text{min}} = |n_x - n_z| k_r
\]

\[
K_{g}^{*} = \frac{n_x}{n_y} \sqrt{|n_y^2 - n_z^2|} \cdot k_r
\]

(2.113) (2.114)

2.3.5 Anisotropic self-diffraction in photorefractive materials

In the isotropic diffraction case the two beams recording the photoinduced grating naturally fulfill the Bragg condition for their self-created grating. Therefore, the two beams will be partly diffracted into each other's direction. This can lead to the well-known beam-coupling effect, where energy is exchanged between the two beams. This effect is theoretically discussed in Section 2.4.3.

In the case of anisotropic self-diffraction no coupling of the writing beams occurs. They cannot interact with the phase grating they have produced because they do not fulfill the Bragg condition for their own grating (they do not "see" their own grating). Only for a well defined incidence angle \( \theta \) and for

![Phase matching condition for anisotropic self-diffraction in an optically uniaxial crystal.](Image)

Fig.2.13 Phase matching condition for anisotropic self-diffraction in an optically uniaxial crystal.
A specific writing polarization is the photoinduced grating capable of diffracting one or both of the writing beams. Figure 2.13 illustrates the phase matching condition needed for anisotropic self-diffraction in a negative uniaxial crystal (e.g. BaTiO$_3$). The two writing beams with wave vectors $k_-$ and $k_+$ have extraordinary polarization and enter the crystal at the incidence angles $\pm \theta$. They create a phase grating with a wave-vector $K_g$. If one of the vectors $(k_-, \pm K_g)$ and $(k_+, \pm K_g)$ ends on the outer shell of the wavevector surface, then the corresponding wave (with wave vector $k_-$ or $k_+$) will be self-diffracted from this grating. For a negative uniaxial crystal (e.g. BaTiO$_3$) the angles outside the crystal are given by [2.19]

$$\sin(\theta') = \sqrt{(n_o^2 - n_e^2)/8}$$

$$\sin(\psi') = 3 \sin(\theta')$$

The angles $\theta$, $\phi$ are measured inside and $\theta'$, $\psi'$ outside the crystal.

In the case of optically biaxial KNbO$_3$ the situation is more complicated (Fig.2.14). Using Eq.(2.108) we find

$$\sin(\theta') = \sqrt{(n_y^2 - n_x^2)/(9n_y^2/n_x^2 - 1)}$$

$$\sin(\psi') = 3 \sin(\theta')$$

Fig.2.14 Phase matching condition for anisotropic self-diffraction in an optically biaxial crystal.
2.4 Coupled-wave theory for Bragg diffraction in photorefractive crystals

2.4.1 Introduction

The coupled-wave theory discussed here assumes monochromatic light entering the photorefractive phase grating at or near the Bragg angle [2.17]. Only two significant light waves are assumed to be present in the grating: the incoming reference wave $R$ and the outgoing signal wave $S$ (Fig. 2.15).

Considering a non magnetic ($\mu=1$) source free medium ($\rho=0$, $j=0$), one obtains from Maxwell's equations the wave equation

$$\Delta \mathbf{E}(r,t) - \nabla (\nabla \cdot \mathbf{E}(r,t)) - \mu_0 \varepsilon_0 \varepsilon \frac{\partial^2 \mathbf{E}(r,t)}{\partial t^2} = 0$$

(2.119)

$\varepsilon_0$ is the permittivity and $\mu_0$ the permeability of the vacuum, $\varepsilon$ is the dielectric tensor at the optical frequency.

The optical electric field $\mathbf{E}$ is assumed to be a sum of monochromatic plane waves of the same frequency

$$\mathbf{E}(r,t) = \sum_j \mathbf{E}_j(r) e^{i\omega t} = \mathbf{E}(r) e^{i\omega t}$$

(2.120)
With this expression the wave equation (2.119) has the form

$$\Delta \mathbf{E}(r) - \nabla (\nabla \mathbf{E}(r)) - \epsilon k^2 \mathbf{E}(r) = 0$$

(2.121)

where $k_o$ is the vacuum wave number $k_o = \omega/c$.

We assume that in the undisturbed system (without any photoinduced phase grating) the inverse optical dielectric tensor $\varepsilon_u^{-1}$ is diagonal

$$\varepsilon_u^{-1} =\begin{pmatrix} \varepsilon_x^{-1} & 0 & 0 \\ 0 & \varepsilon_y^{-1} & 0 \\ 0 & 0 & \varepsilon_z^{-1} \end{pmatrix} =\begin{pmatrix} n_x^{-2} & 0 & 0 \\ 0 & n_y^{-2} & 0 \\ 0 & 0 & n_z^{-2} \end{pmatrix}$$

(2.122)

Due to the photorefractive effect the inverse optical dielectric tensor is spatially modulated by the photoinduced space charge field $E_{sc}$

$$\varepsilon^{-1} = \varepsilon_u^{-1} + r E_{sc}$$

(2.123)

$r$ is the third rank electro-optic tensor. With this the dielectric tensor is written as

$$\varepsilon = \varepsilon_u - \varepsilon_u r E_{sc} \varepsilon_u$$

(2.124)

The photoinduced space charge field is assumed to be sinusoidal according to Eq.(2.31) and produced by diffusion only. With Eq.(2.36) we get

$$E_{sc} = \frac{E_{sc}}{2i} (e^{iK_g r} - e^{-iK_g r})$$

(2.125)

This leads to a sinusoidal perturbed dielectric tensor Eq.(2.124) and can lead to an interaction or coupling of the optical waves in Eq.(2.120).

The optical field in the photorefractive crystal is the superposition of the incoming reference wave and the diffracted signal beam with field vectors $R$ and $S$ respectively

$$\mathbf{E}(r) = R e^{-i k_1 r} + S e^{-i k_2 r}$$

(2.126)
2.4.2 Anisotropic Bragg diffraction with nearly fulfilled Bragg condition

As discussed in Sect. 2.3 anisotropic Bragg diffraction can occur if off-diagonal elements are involved in the photorefractive grating formation. If we assume the photoinduced space-charge field in y-direction (Fig. 2.16) and the occurrence of an electro-optic tensor element of the form \( r_{yz} \) then Eq. (2.124) has the form

\[
\varepsilon = \begin{pmatrix}
    n_x^2 & 0 & 0 \\
    0 & n_y^2 & \varepsilon_{23} \\
    0 & \varepsilon_{23} & n_z^2 \\
\end{pmatrix}
\]  

(2.127)

with

\[
\varepsilon_{23} = -r_{yz} E_{scy} n_y^2 n_z^2 = -i \Delta \left( e^{iK_y y} - e^{-iK_y y} \right)
\]

(2.128)

where

\[
\Delta = \frac{1}{2} r_{yz} n_y^2 n_z^2 E_{scy}
\]

(2.129)

![Fig. 2.16](image)

Geometry for anisotropic Bragg diffraction

The sinusoidal perturbed optical dielectric tensor Eq. (2.127) gives rise to a coupling of waves having polarization components in the y- and z-direction, respectively. The field vectors \( R \) and \( S \) of the incoming and diffracted
waves can be written as

\[
R(x) = \begin{pmatrix}
R_x(x) \\
R_y(x) \\
0
\end{pmatrix}
\quad \text{and} \quad
S(x) = \begin{pmatrix}
0 \\
0 \\
S_z(x)
\end{pmatrix}
\quad (2.130)
\]

In the next calculation we assume the Bragg condition to be at least nearly fulfilled

\[
k_d = k_i + K_g + \delta K
\quad (2.131)
\]

where \(\delta K\) is the Bragg mismatch vector shown in Fig 2.17.

![Diagram](image)

**Fig. 2.17**

Geometry for nearly fulfilled Bragg condition. \(\delta K\) is the Bragg mismatch vector.

In the general case \(\delta K\) has to maximize the following expression \([2.18]\)

\[
1 = \int e^{i\delta K \cdot r} d^3r
\quad (2.132)
\]

where the integration is over the whole interaction region. In this case the diffraction direction violates as little as possible the momentum conservation.

If the hologram expansion parallel to the \(y\)-axis is much larger than the hologram thickness (\(x\)-expansion) the direction of \(\delta K\) is parallel to the \(x\)-axis.
We are now ready to derive the coupled-wave equations. We insert Eq. (2.126) into the wave equation (2.121) using Eqs. (2.127) and (2.131). If we assume the energy exchange between R and S to be slow (this is fulfilled for a small perturbation of the indicatrix, as assumed for this calculation), we can neglect second order derivatives and we arrive at

\[ (-k_f^2 + k_{r y}^2 + k_{r z}^2) R_y + k_{i y} k_{i x} R_x - 2i \cdot k_{i x} \frac{dR_y}{dx} + \]

\[ + ik_{i y} \frac{dR_x}{dx} - ik_{r}^2 S_z \Delta \cdot e^{-i\sigma_k r} = 0 \]  

(2.133)

\[ (-k_a^2 + k_{r}^2 n_z^2) S_z - 2i \cdot k_{d x} \frac{dS_z}{dx} + ik_{r}^2 R_y \Delta \cdot e^{i\sigma_k r} = 0 \]

(2.134)

Using the orthogonality condition for eR and k (eR \perp k_{r})

\[ n_{x}^2 k_{i x} R_x + n_{y}^2 k_{i y} R_y = 0 \]

(2.135)

the wave number of the diffracted beam

\[ k_a^2 = n_z^2 k_{r}^2 \]  

(2.136)

and the condition

\[ (-k_{i x}^2 + k_{i y}^2 + k_{r}^2 n_{y}^2) R_y + k_{i y} k_{i x} R_x = 0 \]

(2.137)

which is the plane wave solution of Eq. (2.121), we get a system of coupled differential equations for the amplitudes R and S

\[ (2k_{i x} + \frac{n_y^2 k_{i y}^2}{n_x^2 k_{i x}}) \frac{dR_y}{dx} + k_r^2 S_z \Delta \cdot e^{-i\sigma_k r} = 0 \]

(2.138)

\[ 2k_{d x} \frac{dS_z}{dx} - k_r^2 R_y \Delta \cdot e^{i\sigma_k r} = 0 \]

(2.139)
Now we insert Eq.(2.139) into Eq.(2.138) and obtain the differential equation for the signal wave \( S \)

\[
\frac{d^2S_Z}{dx^2} - i\delta K \frac{dS_Z}{dx} + \frac{\Delta^2 k_F^4}{2k_{dx}} \left( \frac{n_x^2 k_1}{2k_{fx} n_x^2 + k_{fy} n_y^2} \right) S_Z = 0
\]  
(2.140)

(remember that \( \delta K_x = \delta K \))

The general solution of this equation of the type \( S'' + pS' + qS = 0 \) is

\[
S = S_1 e^{\gamma_1 x} + S_2 e^{\gamma_2 x} \quad \text{with} \quad \gamma_{1,2} = -p/2 \pm \sqrt{p^2/4 - q}
\]  
(2.141)

where \( S_1 \) and \( S_2 \) are constants which depend on the boundary conditions, in our case given by

\[
S_Z(0) = 0 \quad \text{and} \quad R_y(0) = R_y^0
\]  
(2.142)

Using this we find with Eq.(2.139)

\[
\left. \frac{dS_Z}{dx} \right|_{x=0} = \frac{\Delta^2 k_F^2}{2k_{dx}} R_y^0
\]  
(2.143)

The solution for the amplitude \( S \) is

\[
S_Z(x) = \frac{\Delta^2 k_F^2}{2k_{dx}} R_y^0 e^{-i\delta K x} \left[ \frac{\sin(Wx)}{W} \right]
\]  
(2.144)

where

\[
W^2 = \left( \frac{\delta K}{2} \right)^2 + \frac{\Delta^2 k_F^4}{2k_{dx}} \left( \frac{n_x^2 k_1}{2k_{fx} n_x^2 + k_{fy} n_y^2} \right)
\]  
(2.145)

If we assume a unit amplitude \( |R(0)| = 1 \) for the incident wave at \( x=0 \) we find with Eq.(2.137)
and we can rewrite the diffraction efficiency of the grating as

$$\eta = \frac{|S(d)|^2}{|R(0)|^2} = |S(d)|^2$$  \hspace{1cm} (2.147)$$

where \( d \) is the crystal thickness.

Using Eq.(2.144) we finally obtain

$$\eta = A \cdot \frac{\sin^2 \left( \sqrt{\xi^2 + \nu^2} \right)}{1 + \xi^2/\nu^2}$$  \hspace{1cm} (2.148)$$

where

$$\nu = B \cdot \Delta \cdot d \cdot \pi / \lambda$$  \hspace{1cm} (2.149)$$

$$\xi = \delta K \cdot d / 2$$  \hspace{1cm} (2.150)$$

are the coefficients commonly used for isotropic Bragg diffraction, and

$$A = \frac{\cos(\theta_d)}{\cos(\theta_i)} \cdot \frac{1 + n_y^2/n_x^2 \cdot \tan^2(\theta_i)/2}{1 + n_y^2/n_x^2 \cdot \tan^2(\theta_i)}$$  \hspace{1cm} (2.151)$$

and

$$B = \left( \cos(\theta_d) \cdot \cos(\theta_i) \cdot [1 + n_y^2/n_x^2 \cdot \tan^2(\theta_i)/2] \right)^{-1/2}$$  \hspace{1cm} (2.152)$$

Equation (2.148) is similar to the one for isotropic diffraction [2.17], except that the coefficient \( \nu \) is defined differently (Eq.(2.129)), and the geometry factors \( A \) and \( B \) (in the isotropic case the latter two coefficients are both equal to unity).

In the anisotropic case the factor \( B \) is also equal to unity in a good approximation, whereas the factor \( A \), which limits the maximal diffraction efficiency is in general smaller than unity.

For the common diffraction geometries in KNBO\(_3\) (a- or b-plate) \( A \) is
about 0.9. This means that the maximal diffraction efficiency obtainable for anisotropic Bragg diffraction in KNbO₃ crystals is limited to about 90%.

Figure 2.18 shows the results of theoretical calculations of the maximal attainable diffraction efficiency as a function of the Bragg-off angle for different diffraction geometries in KNbO₃. From this figure it is evident that near the minimum incident angle a broad maximum for the diffraction efficiency is attainable whereas for increasing diffraction angles the behaviour is similar to that in isotropic Bragg diffraction.

Fig.2.18 Theoretical calculation of the angular selectivity of anisotropic Bragg diffraction in a KNbO₃ crystal (a-plate, thickness 1mm, calculation for maximal attainable diffraction efficiency given by Eq.(2.144) ). (curves are drawn for different diffraction angles: 1: θᵋₐ=0°, 2: θᵋₐ=5°, 3: θᵋₐ=20°)
2.4.3 Isotropic Bragg diffraction with beam coupling

In this section we will investigate the case where the writing beams will be partly diffracted by their own photoinduced grating. Figure 2.19 illustrates the assumed diffraction geometry. Whenever a phase shift between intensity grating and induced phase grating occurs, energy will be exchanged between the two beams - the two beams couple together. This means that after passing the crystal the ratio of the two writing beam intensities has changed \( \left( \frac{I_S(0)}{I_R(0)} \neq \frac{I_S(d)}{I_R(d)} \right) \).

![Fig.2.19 Assumed diffraction geometry for beam coupling](image)

The observation of beam coupling can be easily explained by the fact that the writing beams evidently fulfill the Bragg condition with their self-induced grating. If the above mentioned phase shift is non-zero then the diffracted wave of one beam and the undiffracted wave of the other beam interfere constructively whereas in the other direction the interference is destructive.

If the two beams have very different intensities then it is possible to amplify very effectively the weaker beam. Amplification factors of up to 4000 have been observed [2.20].

Besides applications in image amplification, the beam coupling effect is very important for the investigation of the photorefractive centers (see Sect.3.3.4).
In this Section 1 will discuss the application of the coupled wave theory for beam coupling in the two cases where the polarizations of the writing beams are vertical and horizontal, respectively. We assume that no external field is applied and that one type of carrier (electrons or holes) is dominant. In this case the photoinduced space-charge field $E_{sc}$ is given by Eq.(2.36). For KNbO$_3$ at room temperature (orthorhombic point group) the disturbed dielectric tensor Eq.(2.124) is

$$
\epsilon = \begin{pmatrix}
    n_1^2 - n_1^4 r_{13} E_{sc} & 0 & 0 \\
    0 & n_2^2 - n_2^4 r_{23} E_{sc} & 0 \\
    0 & 0 & n_3^2 - n_3^4 r_{33} E_{sc}
\end{pmatrix}
$$

(2.153)

with

$$
E_{sc} = \frac{m}{2} \frac{E_q E_d}{E_q + E_d} [i e^{i K_g Z} - i e^{-i K_g Z}]
$$

(2.154)

Notice that the sign of $E_q E_d/(E_q + E_d)$ depends on the sign of the dominant charge carriers

- $E_q E_d/(E_q + E_d) > 0$ for electrons
- $E_q E_d/(E_q + E_d) < 0$ for holes

a) **Vertical polarization of the writing beams**

In this case the amplitudes of the signal beam $S$ and the reference beam $R$ are given by

$$
R = R(y) \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} e^{-i K_R r}
$$

(2.155)

$$
S = S(y) \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} e^{-i K_S r}
$$

(2.156)

The intensity is

$$
I = |R + S|^2 = I_0 \left[ 1 + \frac{m}{2} (e^{i K_g Z} + e^{-i K_g Z}) \right]
$$

(2.157)

with $I_0^2 = R^2 + S^2$ and the modulation ratio $m$ is

$$
m(y) = \frac{2 R S}{R^2 + S^2}
$$

(2.158)
The wave equation (2.121) leads to the differential equation

\[
\left( \frac{\partial^2 R}{\partial y^2} - 2 \text{i} k_R \frac{\partial R}{\partial y} + \text{i} k_R^2 n_1^2 \frac{m}{2} \frac{E_{qE_d}}{E_q + E_d} S \right)e^{\text{i}k_Ry} + \\
\left( \frac{\partial^2 S}{\partial y^2} - 2 \text{i} k_S \frac{\partial S}{\partial y} - \text{i} k_R^2 n_1^2 \frac{m}{2} \frac{E_{qE_d}}{E_q + E_d} R \right)e^{\text{i}k_Sy} = 0
\] (2.159)

If we assume the energy exchange between R and S to be slow (as we did in Sect.2.4.2), we may neglect second order derivatives and we get with the help of Eq.(2.158)

\[
\frac{\partial R}{\partial y} = \frac{\pi n_1^3}{\lambda \cos \Theta} r_{13} \frac{E_{qE_d}}{E_q + E_d} \frac{RS^2}{R^2 + S^2} \\
\frac{\partial S}{\partial y} = -\frac{\pi n_1^3}{\lambda \cos \Theta} r_{13} \frac{E_{qE_d}}{E_q + E_d} \frac{R^2S}{R^2 + S^2}
\] (2.160)

(2.161)

Finally, if we assume the amplitude S of the signal beam to be much smaller than that of the reference beam (S \ll R, \, m \ll 1), then Eq.(2.161) reduces to

\[
\frac{\partial S}{\partial y} = -\frac{\pi n_1^3}{\lambda \cos \Theta} r_{13} \frac{E_{qE_d}}{E_q + E_d} S
\] (2.162)

With S(y=0)=S_0 we find the solution of Eq.(2.162)

\[
S(x) = S_0 e^{-\Gamma y/2}
\] (2.163)

where the exponential gain factor \( \Gamma \) is defined as

\[
\Gamma = \frac{2\pi n_1^3}{\lambda \cos \Theta} r_{13} \frac{E_{qE_d}}{E_q + E_d}
\] (2.164)

The intensity of the signal wave is given by

\[
I_S(y) = S_0^2 e^{-\Gamma y} = I_S^0 e^{-\Gamma y}
\] (2.165)

and can be determined experimentally at y=d, where d is the crystal thickness. Together with a linear absorption \( \alpha \) the same analysis leads to

\[
I_S(d) = I_S^0 e^{-(\Gamma + \alpha)d}
\] (2.166)
b) **Horizontal polarization of the writing beams**

In the case of horizontal polarization of the writing beams, the signal beam $S$ and the reference beam $R$ are given by

$$R = R(y) \begin{pmatrix} 0 \\ -c \end{pmatrix} e^{-ik_R r} \quad (2.167)$$

$$S = S(y) \begin{pmatrix} 0 \\ c \end{pmatrix} e^{-ik_S r} \quad (2.168)$$

where $c=\cos\theta$ and $s=\sin\theta$. The intensity is given by Eq.(2.157) but the modulation ratio in contrast to Eq.(2.158) is given by

$$m(y) = \frac{2RS}{R^2+S^2} \cos(2\theta) \quad (2.169)$$

Multiplying the wave equation (2.121) either by $\begin{pmatrix} 0 \\ -c \end{pmatrix} e^{ik_R r}$ or by $\begin{pmatrix} 0 \\ c \end{pmatrix} e^{ik_S r}$ we get the two differential equations for $R$ and $S$

$$\frac{\partial R}{\partial y} = \frac{\pi \cos\theta}{\lambda} \frac{n_3 r_{33} - n_2 r_{23} \tan^2 \theta}{n} \cdot \frac{E_q E_d}{E_q + E_d} \cdot \frac{RS^2}{R^2+S^2} \cos(2\theta) \quad (2.170)$$

$$\frac{\partial S}{\partial y} = -\frac{\pi \cos\theta}{\lambda} \frac{n_3 r_{33} - n_2 r_{23} \tan^2 \theta}{n} \cdot \frac{E_q E_d}{E_q + E_d} \cdot \frac{R^2S^2}{R^2+S^2} \cos(2\theta) \quad (2.171)$$

$n$ is the effective refractive index given by $n^{-2}=(\cos^2\theta \cdot n_3^{-2} + \sin^2\theta \cdot n_2^{-2})$

With the same "small signal beam" approximation as in the vertical polarization case, Eq.(2.171) reduces to

$$\frac{\partial S}{\partial y} = -\frac{\pi \cos\theta}{\lambda} \frac{n_3 r_{33} - n_2 r_{23} \tan^2 \theta}{n} \cdot \frac{E_q E_d}{E_q + E_d} \cdot S \cos(2\theta) \quad (2.172)$$

The intensity of the signal beam (including a linear absorption) after passing a crystal with thickness $d$, is given by Eq.(2.166). The exponential gain factor $\Gamma$ for horizontal polarization is defined as
\[ \Gamma = \frac{2\pi \cos \theta (n_3^2 r_{33} - n_2^2 r_{23} \tan^2 \theta)}{\lambda n} \frac{E_q E_d}{E_q + E_d} \cos(2\theta) \] (2.173)

In the case where \( r_{33} > r_{23} \) (as in KNbO\(_3\)) the direction of the energy exchange between the two beams depends only on the sign of \( \frac{E_q E_d}{E_q + E_d} \).

**Remark**

The assumed crystal geometry Fig.2.19 was for a KNbO\(_3\) b-plate. For KNbO\(_3\) a-plates in the vertical polarization case \( n_1 \) and \( r_{13} \) have to be changed to \( n_2 \) and \( r_{23} \) in all equations. For horizontal polarization \( n_2 \) and \( r_{23} \) have to be changed to \( n_1 \) and \( r_{13} \).

For deriving Eq.(2.164) and Eq.(2.173) we have assumed the case of pure diffusion in the "one carrier"-model. In a general case, however, in Eqs.(2.164) and (2.173) the term \( \frac{E_q E_d}{E_q + E_d} \) has to be changed into \( \text{Im}(E_\infty) / m \) [2.21].
3. Investigation of the Photorefractive Centers in KNbO₃

3.1 Introduction

The optimization of the material properties for photorefractive applications requires the detailed knowledge of the microscopic processes involved. Of particular interest are informations on the centers supplying charge carriers, on the trapping centers and on optical excitation and transport processes.

Of great interest are transition metal dopants and impurities which occur in different valence states. The generation of space-charge fields, photovoltaic effects and photoconductivity are correlated with such defect centers.

For these reasons photorefractive experiments supply a large number of information on impurity centers involved in photorefractive effects. In this section a number of such investigations are discussed and experimental results obtained with KNbO₃ crystals are presented.

A variety of spectroscopic methods for the investigation of defect and impurity centers in electro-optic crystals can be applied to investigate or even to identify specific photorefractive active centers.

Since doping a crystal with iron improves its photorefractive efficiency, it is widely believed that iron impurities are the main centers for photorefraction. Therefore many spectroscopic investigations of such centers have been performed (e.g. EPR-experiments in LiNbO₃ [3.1] and BaTiO₃ [3.2] and Mössbauer spectroscopy in LiNbO₃ [3.3]).

In iron doped KNbO₃ crystals EPR experiments have been performed by SIEGEL et al. [3.4] with the object of identifying the local position of the iron impurities in the host lattice. They found two centers occurring. One center is Fe³⁺ at the Nb site without local charge compensation. The other center is most probably a center consisting of the Fe³⁺ ion and a local charge compensation by an oxygen vacancy. In the last case two possibilities have been proposed: i) Fe³⁺-V₀ center, if Fe³⁺ is at the Nb⁵⁺ site and an oxygen vacancy in the nearest neighborhood; ii) Vₖ center, if Fe³⁺ is at the K⁺ site with a K⁺ vacancy in the first configuration shell of the K⁺ ions, as partial charge
compensation. Nevertheless looking at Tab. 3.1 the ionic radius of Fe$^{3+}$ is much closer to the value of Nb$^{5+}$ than to the one of K$^+$. On the possible sites of Fe$^{2+}$ impurities still no information is available.

Tab. 3.1 Ionic radii of some transition metals [3.5]

<table>
<thead>
<tr>
<th>Ion</th>
<th>K$^+$</th>
<th>Nb$^{4+}$</th>
<th>Nb$^{5+}$</th>
<th>Fe$^{2+}$</th>
<th>Fe$^{3+}$</th>
<th>Mn$^{2+}$</th>
<th>Mn$^{3+}$</th>
<th>Mn$^{4+}$</th>
<th>Mg$^+$</th>
<th>Mg$^{2+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ionic radius (Å)</td>
<td>1.33</td>
<td>0.74</td>
<td>0.69</td>
<td>0.74</td>
<td>0.64</td>
<td>0.80</td>
<td>0.66</td>
<td>0.60</td>
<td>0.82</td>
<td>0.66</td>
</tr>
</tbody>
</table>

Besides cation impurities or dopants also vacancies can lead to additional deep levels in the band gap and thus could influence photorefractive effects. Even in undoped crystals, cation impurities are always present in a concentration range up to 100 ppm. Vacancies, even besides those correlated with the charge compensation of heterovalent ions, are always present in the 100 to 1000 ppm range and their concentration should change during the pulling process [1.25].

The K$_2$O stoichiometry in KNbO$_3$ crystals can be, moreover, influenced by evaporation phenomena at elevated temperatures as pointed out by FLÜCKIGER [1.26].

Tab. 3.2 Iron analysis of doped samples

<table>
<thead>
<tr>
<th>Crystal No.</th>
<th>210</th>
<th>226</th>
</tr>
</thead>
<tbody>
<tr>
<td>Iron concentration in the melt (At %)</td>
<td>3.1</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Iron concentration in the sample:

- mass spectroscopy (ALUSUISSE) (At ppm) 306±9 129±25
- atom absorption and ICP atom emission (EMPA) (At ppm) 276±15 154±12

Mean value: (At ppm) 300 150 (cm$^{-3}$) $4.6 \cdot 10^{18}$ $2.3 \cdot 10^{18}$

Average distribution coefficient k $\sim 0.01$ $\sim 0.03$
A mass spectroscopic analysis of the starting material (ceramic KNbO₃ powder) showed that about 15 ppm Fe and 15-40 ppm Ta are present. In addition a large number of other atoms have been traced. The concentration of Fe in nominally pure KNbO₃ crystals is reported in Ref.[1.26] to be typically 50 ppm, whereas new crystals prepared under more careful condition exhibit less than 5 ppm of iron. In Tab.3.2 results of Fe concentration analysis of two differently doped samples is shown.

Besides dopants, impurities and oxide non-stoichiometry 3 other types of elementary non-stoichiometries are possible as point defects in a ABO₃ compound. The most probable elementary non-stoichiometry in KNbO₃ is an oxygen deficiency. During crystal growth or processing at high temperatures, these species are the most likely to evaporate, thus creating e.g. oxygen vacancies, denoted as Vₒ. Since the oxygen ion in the KNbO₃ lattice has a charge O²⁻, an oxygen vacancy has a charge Vₒ²⁺. Thus an oxygen vacancy can trap one or two electrons, leading to donor levels in the band gap. In BaTiO₃ these levels are found to be shallow [3.6]. The estimation of the energy depth lead to 0.2 eV for an O²⁻ vacancy with one trapped electron and to 0.025 eV for an O²⁻ vacancy with two trapped electrons [3.7]. At room temperature these levels are expected to be thermally ionized, and are thus not significantly populated.

After reduction of undoped LiNbO₃ in vacuum at high temperature and subsequent illumination with light of short wavelength (λ<600nm) strong Nb⁴⁺ EPR signals were observed by SWEENY et al. [3.19]. It has been found by VON DER LINDE et al. [3.20] that these self trapped electrons are also involved in the photorefractive effect of LiNbO₃.
3.2 Reduction treatment of $\text{KNbO}_3$

As soon as the importance of impurities and defects for the photorefractive effect was realized, it was clear that a controlled treatment of the materials should be studied in order to improve their photorefractive properties [1.21, 1.22].

Besides doping the crystals, thermal treatment is a common way to change the defect structure of a perovskite material. The crucial influence of transition metal impurities on the photorefractive properties has already been mentioned in Sect. 3.1. Changing the valence state of transition metal dopants may induce the generation of space-charge fields. Usually such dopants are added to the melt, e.g. as oxides like $\text{Fe}_2\text{O}_3$. The valence state of these transition metal dopants can be greatly influenced by suitable thermal annealing treatments in different atmospheres. Heating crystals of $\text{LiNbO}_3$, $\text{LiTaO}_3$ and $\text{BaTiO}_3$ in an oxygen atmosphere to temperatures of about 1000°C for several hours tends to oxidize the impurities, e.g. $\text{Fe}^{2+}$ to $\text{Fe}^{3+}$. This process is reversible, and heating in an atmosphere with a low oxygen partial pressure (e.g. hydrogen or argon) or vacuum tends to reduce such impurities [3.5, 3.8, 3.9].

At high temperatures hydrogen reduction would be the simplest reduction procedure also for $\text{KNbO}_3$ crystals. However, this method is complicated by the fact that the crystal has to go repeatedly through two structural phase transitions and can crack and might - depending on the temperature of such a treatment - loose $\text{K}_2\text{O}$ too.

In order to avoid these difficulties the following techniques were investigated:

1. Reduction by reducing atmosphere during pulling [1.24]
2. Pulling in a neutral atmosphere from a melt containing additions of $\text{Fe}^{2+}$ [3.16]
3. Reducing by means of a dc electric field at temperatures below the phase transition at 220°C [1.4, 1.24]
The electro-chemical reduction method.

The electric dc field reduction method (3) gave the best results up to now. With this method electrons are injected into the crystal by passing a current of up to 1000\(\mu\)A/cm\(^2\) through the crystal at a temperature below 220°C. During the reduction, the crystal is placed in an environment which protects the crystal from the oxygen of the atmosphere (e.g. Silicon oil or vacuum) or even in air (Fig.3.1).

The reduction starts at the anode and after a period of up to 100h spreads all over the sample. The degree and the homogeneity of reduction were investigated by measuring the spatial variation of the diffraction efficiency of anisotropic self-diffraction (see Sect.2.3.5) over the whole sample. This is an extremely sensitive method for detecting spatial variations of the reduction state.

Figures 3.2 and 3.3 show typical pictures of the anisotropically self-diffracted beam after different reduction times. Although the linear optical homogeneity in the as grown state is usually almost perfect (interferometrically determined refractive index change less than \(10^{-5}\)), one can easily see that at the initial stage of the reduction treatment the photorefractive index change of the crystal was very inhomogeneous. Depending on the crystal quality, after a certain time (10-100hrs), however, a homogeneous state may be reached (Fig.3.2c and 3.3d).

During reduction the current flowing through the crystal is recorded. If one is assuming that the creation of donors is proportional to the current, an exponential increase during the reduction is expected. In reality at the beginning of the reduction treatment such a behaviour is usually observed but then the current tends to go to a saturation value or even the conductivity of the crystal is oscillating with a long time period (several hours). Figure 3.4 shows the current measured during the reduction of the crystal 179 (nominally pure crystal).

In addition to the increase of the conductivity (and therefore of the complex part of the dielectric constant) also the refractive indices (and therefore the real part of the dielectric constant) are affected by the reduction treatment. The investigation of the Bragg angles of anisotropic self-diffraction in an
inhomogeneously reduced crystal showed that a change of the refractive indices in the order of $\Delta n=10^{-3}$ was induced by reduction (differences between reduced and unreduced parts).

![Fig. 3.1](image1.png)

**Fig. 3.1** Schematic representation of the reduction apparatus. The KNbO$_3$ crystal is placed in the reduction oven and a dc voltage is applied. The oven is placed in a vacuum chamber to avoid oxygen diffusion into the crystal during reduction.

![Fig. 3.2](image2.png)

**Fig. 3.2** Anisotropic self-diffraction pattern of a KNbO$_3$ b-plate at different reduction states (a: after 16h, b: after 20h, c: after 50h).
Fig. 3.3  Anisotropic self-diffraction pattern of a KNbO₃ α-plate at different reduction states (a: after 21h, b: after 40h, c: after 102h, d: after 118h, e: after 135h and f: after 153h).
Fig. 3.4  Current density during reduction of crystal 179 (nominally pure) with an applied voltage of 500V (applied electric field $E=833\,\text{V/cm}$) at a temperature $T=190^\circ\text{C}$. 
3.3 Experimental techniques for the investigation of photorefractive centers

3.3.1 Dark- and photoconductivity

The transport of charges upon illumination is of basic importance for the photorefractive process. Neglecting transient terms (e.g. pyroelectric currents) this migration of free charges is due to diffusion and to drift in an electric field. In the case of ferroelectric photoconductors a third mechanism, the photovoltaic effect, occurs.

The transport equation for monochromatic excitation and one type of charge carriers is given by [3.10]

\[ j = \chi \alpha I + \left[ \sigma_d + \sigma_{ph}(I) \right] E - qD \nabla n \]  \hspace{1cm} (3.1)

where

\[ \chi = \frac{ef_j \Phi}{h\nu} \]  \hspace{1cm} (3.2)

\[ \sigma_d = e n_d \mu \]  \hspace{1cm} (3.3)

\[ \sigma_{ph} = e n_{ph} \mu \]  \hspace{1cm} (3.4)

\( l_o \) is the photovoltaic drift length, \( \Phi \) the quantum efficiency for exciting a free charge carrier, \( \nu \) the light frequency, \( \mu \) the mobility and \( n_d \) is the concentration of free charge carriers in the dark and \( n_{ph} \) under illumination.

\[ n_{ph} = g \tau_R = \frac{\alpha \Phi}{h \nu \tau_R} \]  \hspace{1cm} (3.5)

In cases where no single type of charge carrier is dominant, the conductivity is given by the sum of electron and hole conductivity

\[ \sigma = \sigma_h + \sigma_e = \mu_h n_h + \mu_e n_e \]  \hspace{1cm} (3.6)

In general, the transport equation (3.1) is a tensor equation because of the tensor properties of the conductivity (second rank tensor) and of the photovoltaic effect. The latter one can be described by the third rank tensor \( \beta_{ijkl} \) [3.11]
where $\mathbf{E}_k = E_k e^{i(\omega t - kr)}$ are the components of the electric field of light.

In the following we will discuss the conductivity to be along the polar c-axis in KNbO$_3$.

Detailed experimental studies of the volume photovoltaic effect were first carried out in LiNbO$_3$ doped with iron (GLASS et al. [3.12]). They have attributed this effect to an asymmetric charge transport of photoionized defects. Directional intervalence transfer from a substitutional cation in a pyroelectric host will occur if the overlap of defect orbitals with the host cations in the ± polar directions differ due to the local asymmetry [3.13].

With Eq.(3.1) it is obvious that in the equilibrium ($j=0$) a homogeneous illumination of the crystal creates a photovoltaic field $E_{ph}$

$$E_{ph} = \frac{-\chi \alpha l}{\sigma_d + \sigma_{ph}} = \frac{t_0 \Phi}{\sigma_d + \sigma_{ph}} \frac{e/h\nu}{\alpha l}$$

With increasing intensity the photovoltaic field $E_{ph}(I)$ is approximating the photovoltaic saturation field $E_{sat}^{\text{ph}}$

$$E_{sat}^{\text{ph}} = \frac{-I_0}{\mu t}$$

We consider the case that extrinsic impurity centers act as filled and empty traps and that the number of these traps is not much changed by the excitation and trapping processes. In this case the generation rate is proportional to the light intensity and the concentration of the filled traps ($N_D - N_A$) as assumed in Eq.(3.5). The lifetime $t_R$ on the other hand is inversely proportional to the concentration $N_A$ of empty traps capturing the charge carriers. The mobility $\mu$ is mainly determined by the band structure and independent on the impurity centers in this approximation.

This simple model is approximately confirmed by the photoconductivity measurements of nominally pure and Fe-doped KNbO$_3$ crystals discussed in
Sect. 3.4.1. A relation close to $\sigma_{ph} \sim l$ is found, in contrary to a relation $\sigma_{ph} \sim l^{1/2}$ which is expected for intrinsic excitation and recombination processes.

From the measurement of the current density as a function of the applied field strength and the intensity of the illuminating beam, the characteristic figures for the conductivity can be determined: the dark conductivity $\sigma_d$, the product $\Phi \mu \tau_R$ of the quantum efficiency $\Phi$, the mobility $\mu$, and the recombination time $\tau_R$, and the product $\tau \Phi$ of the photovoltaic drift length $\tau$ and the quantum efficiency $\Phi$.

The experimental set-up for the measurement of photoconductivity data is schematically shown in Fig. 3.5. An expanded Ar$^+$-laser beam is illuminating the crystal with monochromatic light (488 or 515nm). The voltage applied to the crystal in its polar direction is produced by a computer controlled high voltage power supply. The current flowing through the crystal is measured by a "Keithley 197" digital multimeter. For low conductivity crystals the current was measured in the high input resistivity DC-VOLT mode ($R > 1 \, G\Omega$) with the help of a resistor $R$ (Fig. 3.5a). For highly conductive crystals (reduced samples) the current was directly measured in the current mode with 10 nA resolution (Fig. 3.5b).

An extrinsic semiconductor shows a temperature dependence of the dark conductivity given by

$$j = j_0 e^{-\Delta E/2kT} + j_{0(\text{ex})} e^{-\Delta E_{\text{ex}}/2kT}$$  \hspace{1cm} (3.10)

where $\Delta E$ is the band gap and $j_0$ the corresponding current. $\Delta E_{\text{ex}}$ is the gap between impurity level and conduction band and $j_{0(\text{ex})}$ the current resulting from excitations from these impurity levels.

Experimentally the dark conductivity in KNbO$_3$ can be fitted by $j \sim A_\omega e^{-W/kT}$ (see Sect. 3.4.1), where $W$ is the activation energy for the thermal excitation of charge carriers and $A_\omega$ is a constant.
Fig. 3.5 Diagram of the experimental set-up for dark- and photoconductivity measurement.

a) Low current mode         b) high current mode
3.3.2 Optical Spectroscopy

Optical absorption measurements in photorefractive crystals can provide various information about the identity of impurities and defects, about their charge, the energy levels and the nature of transitions. Fig. 3.6 shows the different transitions contributing to the optical absorption.

The specific energy levels affect optical absorption because of optically induced carrier transitions between defect energy levels and the conduction or valence band. Photoconductivity measurement as a more sensitive technique is usually employed to investigate electronic transitions. Certain processes can produce absorption bands without any photoconductivity. (e.g. intra-band transitions and polarons indicated in Fig. 3.6). In some cases these other processes can be used to obtain microscopic information about the nature of individual defects.

For photon energies larger than the band gap, absorption is caused by electron-hole pair generation. In wavelength regions corresponding to a significant fraction of band gap energy, electronic transitions between localized energy levels and the band edge tend to dominate. In the longer wavelength portion of the spectrum, however, nonelectric absorption bands are observed. Even electrically inactive defects produce infrared absorption bands (e.g. exciton formation).

![Diagram of electronic transitions in photoconductors](image)

Fig. 3.6 Electronic transitions in photoconductors

(1) Interband absorption (2), (3) absorption at localized imperfections in the crystal (4) intra-band transition, (5) polaron transition.
An idealized absorption spectrum illustrating these transitions is given in Fig.3.7. There is a cutoff of absorption at the minimum energy required for a certain transition; this minimum corresponds to the band gap for transition 1 of Fig.3.6. The transitions (2,3) of Fig.3.6 on the other hand lead to the absorption (b,c) in Fig.3.7.

![Absorption spectrum diagram](image)

**Fig.3.7** Representative absorption spectrum.

(a) Absorption due to a transition from the valence band to the conduction band; (b,b') absorption due to a transition from an imperfection level to the conduction band; (c,c') absorption due to a transition from the valence band to an imperfection level.

(after Ref.[3.14])

The optical absorption measurements described in this work were performed by using a Perkin-Elmer 330 spectrometer having a wavelength range from 200nm to 2500nm. The absorption spectrum is measured with polarized light.
3.3.3 Electron Paramagnetic Resonance (EPR) Experiments

The electron paramagnetic resonance (EPR) technique is a particularly useful tool for the study of defects in insulators and semiconductors \[3.15\]. Electrical, optical and other properties of semiconductors are extremely sensitive to small concentrations of impurities. Paramagnetic resonance is one of the most sensitive tools for investigating the detailed electronic nature of impurity sites and it also provides informations on their concentration.

In photorefractive materials EPR has been used to identify paramagnetic transition metal ions, trapped hole centers and electrons close to the conduction band \[3.1, 3.2, 3.15\]

In EPR experiments transitions within Zeeman levels of the electronic ground state manifold are induced by a microwave magnetic field. The energy splitting of the electronic ground state levels is due to one or more of the following influences:

- External magnetic field
- Electron-nuclear interaction (hyperfine splitting)
- Crystal field effects

The corresponding Spin-Hamiltonian is \[3.15\]

\[
H = \beta S g H + S A I + S D S + \ldots
\]  \(3.11\)

where only three representative terms are given for illustration:

\[\beta S g H : \text{ Zeeman splitting}\]
\[\beta: \text{ Bohr's magneton}\]
\[S: \text{ Spin operator}\]
\[g: \text{ Zeeman interaction tensor}\]
\[H: \text{ External magnetic field}\]

\[S A I : \text{ Magnetic hyperfine interaction}\]
\[I: \text{ Nuclear spin operator}\]
\[A: \text{ Tensor of the magnetic hyperfine interaction}\]

\[S D S : \text{ Zero field splitting by the crystal field}\]
\[D: \text{ Zero field splitting tensor}\]
The geometrical arrangement of a defect implies the symmetry of the coupling tensors which in many cases can be derived by observing the change of the spectra on varying the angle between crystal axes and magnetic field. The identity of the nuclei in the center can be derived from their hyperfine splittings, generally into $2I+1$ lines for interaction with one kind of nuclei having spin $I$. The concentration of defects is obtained by comparing the intensity of its EPR spectrum with that of a standard of known number of spins.

Fig.3.8 shows room temperature EPR spectra of KNbO$_3$:Fe. The point group symmetry of the impurity center is mm2 for a Fe$^{3+}$ ion ($^6S_{5/2}$) substituting a Nb atom in orthorhombic KNbO$_3$ [3.17].

Fig.3.8 Room temperature EPR spectra of KNbO$_3$:Fe at 9.1GHz [3.17].

$\theta$ is the angle between a-axis and magnetic field in the (010) plane.

(---- Experimental lines, ------ Calculated transitions)

For this work EPR experiments were performed using a X-band spectrometer (Varian E109; Anorganisches Chemisches Institut der Universität Zürich, Prof. Dr. H.R. Oswald). The microwave frequency is fixed to 9.51GHz and a static magnetic field can be varied from 0 to 10kG.
3.3.4 Photorefractive beam coupling

A useful technique which provides information on the magnitude and the sign of the steady state space-charge field (and thus on a number of important photorefractive parameters) is the measurement of the beam coupling gain $\Gamma$.

In Sect.2.4.3 the coupled wave analysis for beam coupling was discussed in the one carrier model. The main assumption to get the final equations was the small modulation approximation ($m \ll 1$). Experimentally this is very easy to achieve by attenuating one of the two writing beams. The expression for the intensity of the signal beam after passing the crystal with a thickness $d$ is given by Eq.(2.166)

$$I_S(d) = I_S(0) e^{-(\Gamma + \alpha)d}$$

Beam coupling experiments in KNbO$_3$ crystals are usually performed in the geometry shown in Fig.3.9. The active electro-optic coefficients are $r_{13}, r_{23}, r_{33}$ or mixtures of these coefficients depending on the polarization of the writing beams and on the crystal cut.

In the following we will discuss two cases of charge carrier transport:

a) pure diffusion in the one carrier model
b) pure diffusion in the two carrier model with one single recombination center (Model 1 in Sect.2.2)
Whereas case a) is a good approximation for doped as grown and for reduced crystals (doped and nominally pure), case b) can explain the observations in pure as grown samples

a) Pure diffusion in the one carrier model

This is the case already assumed in the coupled wave analysis of Sect.2.4.3. The results obtained there - Eqs. (2.164) and Eqs. (2.173) - for the two main orientations in KNbO₃ crystals are:

1) Vertical polarization in KNbO₃ (a-plate):

\[ \Gamma = \frac{2\pi n_2^3}{\lambda \cos \theta} \frac{E_q E_d}{E_q + E_d} \]  \( (3.12) \)

2) Vertical polarization in KNbO₃ (b-plate):

\[ \Gamma = \frac{2\pi n_1^3}{\lambda \cos \theta} \frac{E_q E_d}{E_q + E_d} \]  \( (3.13) \)

3) Horizontal polarization in KNbO₃ (a-plate):

\[ \Gamma = \frac{2\pi n_2^3}{\lambda \cos \theta} \frac{E_q E_d}{E_q + E_d} \cos(2\theta) \]  \( (3.14) \)

with \( n^{-2} = (\cos^2 \theta n_3^{-2} + \sin^2 \theta n_1^{-2}) \)

4) Horizontal polarization in KNbO₃ (b-plate):

\[ \Gamma = \frac{2\pi n_2^3}{\lambda \cos \theta} \frac{E_q E_d}{E_q + E_d} \cos(2\theta) \]  \( (3.15) \)

with \( n^{-2} = (\cos^2 \theta n_3^{-2} + \sin^2 \theta n_2^{-2}) \)

The diffusion field \( E_d \) is given by Eq.(2.25)

\[ E_d = \frac{q D K_g}{\mu} = \frac{q}{e} \left( \frac{k_B T}{e} \right) K_g \]
and the limiting field $E_q$ by Eq.(2.34)

$$E_q = \frac{q}{\varepsilon_0 K_g} \frac{1}{N_D} \frac{N_A}{N_D} (N_D - N_A)$$

We define $E_{qo} = (e/q)(E_q/A)$ and $E_{do} = (e/q)E_d\Lambda$ by

$$E_{do} = 2\pi \left( \frac{k_B T}{e} \right)$$

and

$$E_{qo} = \frac{q}{2\pi \varepsilon_0} \frac{N_A}{N_D} (N_D - N_A)$$

With these expressions Eqs.(3.12)-(3.15) lead to

$$\Gamma = \frac{q}{e} \frac{2\pi n^3}{\lambda \cos \theta} \frac{r_{\text{eff}}}{E_{do} E_{qo}} \frac{E_{do} E_{qo}}{E_{do}/A + E_{qo} \Lambda} \cos(2\theta \cdot p)$$

where $r_{\text{eff}}$ is the effective electro-optic coefficient and $n$ the active refractive index (both dependent on the angle $\theta$ and the polarization of the writing beams) and $p$ is the polarizing factor ($p=0$ for vertical and $p=1$ for horizontal polarization). For angles $\theta$ (inside the crystal) which are not too large, $r_{\text{eff}}$ and $n$ can be assumed to be independent of the incidence angle $\theta$.

Fig.3.10 Theoretical calculation of the grating spacing dependence of the exponential gain $\Gamma$ for typical photorefractive parameters in KNbO$_3$. 

**Fig.3.10** Theoretical calculation of the grating spacing dependence of the exponential gain $\Gamma$ for typical photorefractive parameters in KNbO$_3$. 

$N_A = 10^{16}$ cm$^{-1}$ 
$r_{\text{eff}} = 28$ pm/V
A measurement of the exponential gain as a function of the grating spacing leads to an easy determination of the sign of the charge carriers and of the number density of the ionized donors $N_A$. The theoretical basis for the determination of this value is given by Eq.(3.18). An illustration of the qualitative dependence $\Gamma(\Lambda)$ is shown in Fig.3.10.

The photorefractive beam coupling experiments reported here are performed with $a$- or $b$-plates. The polarizations are parallel to the plane of incidence (Fig.3.9). In this configuration the effective electro-optic coefficient $r_{eff}$ is dominated by $r_{33}$.

From Eq.(3.18) one sees that a plot of $1/\Lambda \Gamma$ as a function of $1/\Lambda^2$ gives a straight line for small angles $\theta$ ($\cos \theta \approx 1$)

$$\frac{1}{\Lambda \Gamma} = \frac{1}{\Lambda \Gamma} \approx \frac{e \lambda \cos \theta}{q 2\pi n^3 r_{eff} \cos \theta} \frac{E_{do}/\Lambda^2 + E_{qo}}{E_{do} E_{qo}}$$

(3.19)

The error in setting $\cos \theta \approx 1$ is more than 10% for $\Lambda < 0.5 \text{ \mu m}$. It is necessary, therefore, to introduce the reduced gain $\Gamma_r$ defined by

$$\frac{1}{\Lambda \Gamma_r} = \frac{\cos \theta}{\Lambda \Gamma \cos \theta} = \frac{e \lambda}{q 2\pi n^3 r_{eff}} \frac{E_{do}/\Lambda^2 + E_{qo}}{E_{do} E_{qo}}$$

(3.20)

and fit experimental data with a least-squares fit.

The sign of $\Gamma$ depends on the sign of the free carriers $q$. By investigating the direction of the energy flow in a beam coupling experiment, it is easy to decide whether electrons or holes are the dominant carriers. Figure 3.11 illustrates these two possibilities for KNbO$_3$ crystals.

Besides the determination of the sign of the free carriers the main feature of the beam coupling experiment is the possibility to determine the concentration $N_A$. It is easy to see in Eq.(3.18) that for given diffraction geometry (angle and polarization), electro-optic coefficients and refractive indices, the only free parameter is $(N_D-N_A)N_A/N_D$. This expression reduces to $N_A$ if $N_A \ll N_D$

It turns out, in reality, that experimental values of $r_{eff}$ obtained by beam coupling experiments are in general smaller than the theoretical ones. This can
be due to inhomogeneities of the photorefractive properties or to a reduction of the fringe contrast due to Fresnel reflections at the uncoated end faces of the crystal. This reduction could also be described by a reduced crystal thickness $d_{\text{red}}$ in Eq. (2.166). In BaTiO$_3$ also 180° ferroelectric domains should contribute to this reduction [2.4].

b) Pure diffusion in the two carrier model

In this case the term $E_q E_d/(E_q + E_d)$ in Eq. (3.18) has to be changed to an expression - Eq. (2.81) - already developed in Sect. 2.2. With this the general expression for the exponential gain Eq. (3.18) has to be generalized to

$$
\Gamma = q \frac{2\pi n^3}{\lambda \cos \theta} r_{\text{eff}} \frac{E_q E_d}{E_q + E_d} \left[ \frac{E_q (\tau_{\text{dilh}} - \tau_{\text{die}})}{E_q (\tau_{\text{dilh}} + \tau_{\text{die}})} + \frac{E_d (\tau_{\text{ilh}} - \tau_{\text{ile}})}{E_d (\tau_{\text{ilh}} + \tau_{\text{ile}})} \right] \cos(2\theta p)
$$

(3.21)

After certain transformations, using Eqs. (2.25), (2.34), (2.75), (2.76) and (2.80) we get the form [2.5]

$$
\Gamma = q \frac{2\pi n^3}{\lambda \cos \theta} r_{\text{eff}} \frac{E_q E_d}{E_q + E_d} \frac{(1-C)}{(1+C)} \cos(2\theta p)
$$

(3.22)
where \( C \) is given by

\[
C = \frac{s_h}{s_e} \frac{N_A}{(N_D-N_A)} \frac{(K_e^2 + K_o^2)}{(K_e^2 + K_h^2)} \quad (3.23)
\]

\( K_e^{-1} = L_e \) and \( K_h^{-1} = L_h \) are average distances traveled by the two kinds of charge carrier between excitation and recombination:

\[
K_e^{-2} = \frac{k_B T \mu_e}{e \gamma_e N_D} \quad K_h^{-2} = \frac{k_B T \mu_h}{e \gamma_h (N_D-N_A)} \quad (3.24)
\]

In the single charge carrier case \((1-C)/(1+C)=1\) for electrons and \((1-C)/(1+C)=-1\) for holes. Note that if the two carriers occur simultaneously, \((1-C)/(1+C)\) exhibits a \( K \) dependence, which even allows the inversion of its sign.

Interpretation of experimental results is performed by adjusting the inverse transport lengths \( K_e \) and \( K_h \), the limiting space charge field \( E_q \) (and therefore \((N_D-N_A)N_D/N_A\)) and the ratio of excitation rates for holes and electrons \( S=s_h N_D/s_e (N_D-N_A) \) by a least-squares fit.

Figure 3.12 shows the experimental set-up for the investigation of beam coupling in KNbO\(_3\) crystals. With the computer controlled shutters one is able to measure the effective gain \( \gamma_0 \)

\[
\gamma_0 = \frac{l_0(d) \text{ with reference beam}}{l_0(d) \text{ without reference beam}} \quad (3.25)
\]

When depletion of the reference wave can be neglected (justified for small modulation \( m \)) \( \gamma_0 \) can be written as

\[
\gamma_0 = e^{-\Gamma d} \quad (3.26)
\]

The fringe spacing dependence of the exponential gain can be measured by changing the crystal position and re-adjusting the directions of the signal and reference beam.
Fig. 3.12 Experimental set-up for beam coupling experiments in photorefractive KNbO$_3$ a- or b-plates.
3.4. Experimental Results

3.4.1 Dark- and photoconductivity

Figs. 3.13-3.14 show typical experimental data of photoconductivity measurements in different KNbO₃ crystals. The full lines are least-squares fits of the experimental points. In Tab. 3.3 the results are summarized.

![Diagram showing photoconductivity measurement results for nominally pure, as grown KNbO₃ and nominally pure, reduced KNbO₃ crystals.](image)

*Fig. 3.13 Experimental data of photoconductivity measurement in a nominally pure KNbO₃ crystal.*

a) as grown  b) reduced sample
Fig. 3.14 Experimental data of photoconductivity measurement in a iron doped KNbO₃ crystal.

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<table>
<thead>
<tr>
<th>Crystal No.</th>
<th>Dopant</th>
<th>Treatment</th>
<th>$\lambda$ nm</th>
<th>$\alpha$ cm$^{-1}$</th>
<th>$\sigma_d$ ((\Omega \text{cm}))$^{-1}$</th>
<th>$\Phi \mu \tau_R$ cm$^2$ V$^{-1}$</th>
<th>$i_0 \Phi$ pm</th>
<th>$i_o/\mu \tau_R$ V cm$^{-1}$</th>
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<tr>
<td>179</td>
<td>pure as grown</td>
<td>488</td>
<td>0.81</td>
<td>$7.8 \cdot 10^{-13}$</td>
<td>$5.2 \cdot 10^{-10}$</td>
<td>340</td>
<td>65</td>
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<tr>
<td></td>
<td></td>
<td>515</td>
<td>0.75</td>
<td>$2.3 \cdot 10^{-10}$</td>
<td>100</td>
<td>43</td>
<td></td>
<td></td>
</tr>
<tr>
<td>179</td>
<td>pure reduced</td>
<td>515</td>
<td>1.42</td>
<td>$3.3 \cdot 10^{-8}$</td>
<td>$7.6 \cdot 10^{-8}$</td>
<td>36000</td>
<td>47</td>
<td></td>
</tr>
<tr>
<td>226</td>
<td>Fe:150 as grown</td>
<td>488</td>
<td>1.00</td>
<td>$2.3 \cdot 10^{-14}$</td>
<td>$2.9 \cdot 10^{-11}$</td>
<td>137</td>
<td>380</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>515</td>
<td>0.72</td>
<td>$2.9 \cdot 10^{-11}$</td>
<td>164</td>
<td>380</td>
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<td></td>
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<td>Fe:150 reduced</td>
<td>488</td>
<td>7.00</td>
<td>$8.7 \cdot 10^{-11}$</td>
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<td>603</td>
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<td>$2.1 \cdot 10^{-11}$</td>
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<td>650</td>
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Table 3.3 Photoconductivity parameters obtained from conductivity measurements in KNbO$_3$ crystals.
Figure 3.15 shows the temperature dependence of the dark conductivity in two reduced \( \text{KNbO}_3 \) samples (nominally pure and iron doped). As predicted in Sect. 3.3.1 an exponential temperature dependence is observed. The activation energy was found to be \( W = 0.7 \text{ eV} \) in the pure and \( W = 1 \text{ eV} \) in the iron doped sample.

\[ \begin{align*}
\text{Conductivity } \sigma \ (1/\Omega \text{cm}) & \\
10^{-5} & \\
10^{-6} & \\
10^{-7} & \\
10^{-8} & \\
10^{-9} & \\
10^{-10} & \\
\end{align*} \]

\[ \begin{align*}
\text{Inverse Temperature } T^{-1} \ (K^{-1}) & \\
0.002 & \\
0.003 & \\
\end{align*} \]

**Fig. 3.15** Temperature dependence of the dark conductivity in two different \( \text{KNbO}_3 \) samples (reduced, nominally pure and iron doped).

### 3.4.2 Optical Spectroscopy

Figure 3.16 shows the optical absorption of a pure as grown and a pure reduced \( \text{KNbO}_3 \) sample. In Fig. 3.17 the absorption spectra of an iron doped \( \text{KNbO}_3 \) crystal for the as grown as well as the reduced state is shown.

The dotted lines in Figs. 3.16 and 3.17 is the difference between the absorption spectra of the reduced and the as grown state. It indicates that in the reduced samples a strong absorption due to a transition from an impurity level to the conduction band near 2.75 eV is present.
Fig. 3.16 Absorption spectra of a pure as grown and a pure reduced KNbO$_3$ sample. The dotted line is the difference between the two curves.

Fig. 3.17 Absorption spectra of an iron doped as grown and reduced KNbO$_3$ sample. The dotted line is the difference between the two curves.
3.4.3. Electron paramagnetic resonance (EPR) experiments

Figure 3.18 shows EPR spectra of an iron doped, as grown KNbO₃-crystal (150ppm Fe) for the two directions of the static magnetic field directions $H_o$ parallel to the spontaneous polarization $P_s$ and parallel to the crystallographic a-axis, respectively. The strong line at high magnetic field was found to be insensitive to a small angle detuning whereas the other signals showed a stronger angular dependence. The strong line was, therefore, taken as reference line for determining the concentration change of the Fe³⁺-ions at different reduction states. In Fig.3.19 this line is shown for an iron doped...

Fig.3.18 EPR spectra of an iron doped, as grown KNbO₃-crystal (150ppm Fe) for the static magnetic field directions $H_o$ parallel to the spontaneous polarization $P_s$ (c-axis) and parallel to the a-axis.
KNbO$_3$-crystal (150ppm Fe) at different reduction states. Fig.3.19a corresponds to the spectrum of Fig.3.18 measured in an as grown crystal. Fig.3.19b and 3.19c are the lines of the crystal at different states of the electrochemical reduction (b:30min at 180°C and $E=4kV/cm$; c:additional 100h at 190°C and $E=1kV/cm$) and Fig.3.19d shows the lines of a thermally reduced crystal (48h at 800°C in a N$_2$ atmosphere containing 1% H$_2$). By comparing these amplitudes one finds that the Fe$^{3+}$ concentration is lowered by 15±3% after the first electrochemical treatment and by 65±10% after the second reduction. In the case of the thermally reduced crystal no Fe$^{3+}$ signal can be observed and hence the Fe$^{3+}$ reduction is 99±1%. In Fig.3.20 the EPR spectrum of a nominally pure, as grown crystal is shown. The characteristic Fe$^{3+}$-lines are not visible and therefore this concentration must be below 1ppm.

Fig.3.19 Characteristic EPR-line ($H_0||P_e$) for an Iron doped KNbO$_3$-crystal (150ppm Fe) at different reduction states.

a) as grown ; b) 30min at 180°C and $E=4kV/cm$; c) additional 100h at 190°C and $E=1kV/cm$; d) 48h at 800°C in a N$_2$ atmosphere with 1% H$_2$

Fig.3.20 EPR spectrum of a nominally pure, as grown KNbO$_3$-crystal.
3.4.4 Beam Coupling Experiments

Fig.3.21a shows an example of a plot of $1/\Lambda^r_{\chi}$ as a function of $1/\Lambda^2$ as discussed in Sect.3.3.4, Eq.(3.20) for KNbO$_3$:Fe(300ppm). The full line is a least-squares fit. Figs.3.21b and 3.22 show experimental data and the corresponding least-squares fit for the gain $\Gamma$ as a function of the fringe spacing $\Lambda$ for KNbO$_3$:Fe (300ppm) and KNbO$_3$:Fe (130ppm) in the as grown and reduced state.

![Graph showing inverse gain coefficient $\Gamma$ times grating spacing $\Lambda$ as a function of $1/\Lambda^2$ for KNbO$_3$:Fe(300ppm). The full line is a least-squares fit.](image1)

![Graph showing gain $\Gamma$ as a function of the grating spacing $\Lambda$ of KNbO$_3$:Fe(300ppm).](image2)

Fig.3.21  a) Inverse gain coefficient $\Gamma$ times grating spacing $\Lambda$ as a function of $1/\Lambda^2$ for KNbO$_3$:Fe(300ppm). The full line is a least-squares fit.

b) Gain $\Gamma$ as a function of the grating spacing $\Lambda$ of KNbO$_3$:Fe(300ppm)
Fig. 3.22 Gain $\Gamma$ as a function of the grating spacing $\Lambda$ of KNbO$_3$:Fe(150 ppm) in the as grown and reduced state. The full lines are least-squares fits using Eq. (3.18).
In Tab. 3.4 the results of these beam coupling experiments are summarized assuming the one carrier model discussed in Sect. 3.3.4.

Table 3.4 Summary of the data derived from beam coupling experiments

<table>
<thead>
<tr>
<th>Crystal No.</th>
<th>Dopand</th>
<th>Treatment</th>
<th>( \lambda ) nm</th>
<th>Sign of carriers</th>
<th>( r_{\text{eff}} ) pm/V</th>
<th>( N_A \cdot (N_D - N_A)/N_D ) (10^{18}\text{cm}^{-3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>179</td>
<td>pure</td>
<td>reduced</td>
<td>515</td>
<td>-</td>
<td>26</td>
<td>0.31</td>
</tr>
<tr>
<td>226 Fe:150</td>
<td>as grown</td>
<td></td>
<td>515</td>
<td>+</td>
<td>14</td>
<td>1.1</td>
</tr>
<tr>
<td>226 Fe:150</td>
<td>reduced</td>
<td></td>
<td>515</td>
<td>-</td>
<td>17</td>
<td>87</td>
</tr>
<tr>
<td>210 Fe:300</td>
<td>as grown</td>
<td></td>
<td>515</td>
<td>+</td>
<td>43</td>
<td>2.0</td>
</tr>
</tbody>
</table>

Fig. 3.23 Fringe spacing dependence of the gain \( \Gamma \) in a pure, as grown crystal. The full lines are theoretical fit curves using the electron-hole model Eq. (3.21).
In the case of pure undoped KNbO$_3$ crystals the single carrier model is not compatible with the experimental results. Figure 3.23 shows the fringe spacing dependence of the gain $\Gamma$ measured in a nominally pure, as grown KNbO$_3$ crystal. The full line is a theoretical fit using the two carrier model as discussed in Sect. 3.3.4. From Fig. 3.23 one sees that a zero crossing of the gain occurs at a certain grating spacing. In this point the contributions from electrons and holes compensate each other. Table 3.5 summarizes the results obtained by data fitting.

Table 3.5 Parameters for fitting the gain-curves of Fig. 3.23

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>488</th>
<th>515</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma_{\text{eff}}$ (pm/V)</td>
<td>45</td>
<td></td>
</tr>
<tr>
<td>$L_h=2\pi/K_h$ (\mu m)</td>
<td>0.18</td>
<td></td>
</tr>
<tr>
<td>$L_e=2\pi/K_e$ (\mu m)</td>
<td>0.35</td>
<td></td>
</tr>
<tr>
<td>$N_A/NA$</td>
<td>1.3</td>
<td>2.0</td>
</tr>
<tr>
<td>$N_A/ND$</td>
<td>$7.8 \times 10^{15}$</td>
<td></td>
</tr>
</tbody>
</table>
3.5 Summary and Discussion

The electrochemical procedure is well suited for a controlled reduction of KNbO₃ crystals as discussed in Sect. 3.2. With EPR investigations (Sect. 3.4.3) we have experimentally shown that this method actually acts on the Fe³⁺. From the absorption measurement (Sect. 3.4.2) one sees that by electrochemical reduction of KNbO₃ crystals a new band is created. This new band at about 2.7 eV was observed already in earlier work [1.4] and ascribed to Fe²⁺-centers.

In Tab. 3.3 the data of conductivity measurements are summarized. The iron concentration, given in the second column, has been determined by different spectroscopic methods (see Sect. 3.1). Reduced KNbO₃ samples show a more pronounced photovoltaic effect and larger photoconductivities and, therefore, much larger photovoltaic drift lengths Φφ and Φμτ products than as grown crystals. By reduction also an increase of the dark conductivity can be observed. The photovoltaic saturation field (φφ/μτR) in nominally pure crystals is about one order of magnitude smaller than in iron doped samples. In Table 3.6 these data are summarized and compared to data of other photorefractive crystals.

Tab.3.6 Absorption constant α, dark conductivities σ₃ and transport parameters, derived from photoconductivity measurements, for a series of electro-optic materials (room temperature values)

<table>
<thead>
<tr>
<th>Material</th>
<th>α (cm⁻¹)</th>
<th>σ₃ (Ωcm⁻¹)</th>
<th>ΦμτR (cm² V⁻¹)</th>
<th>φφ</th>
<th>μτR (V cm⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KNbO₃</td>
<td>0.5-10</td>
<td>10⁻¹⁴-10⁻⁷</td>
<td>10⁻¹¹-10⁻⁷</td>
<td>100-36 000</td>
<td>40-800</td>
</tr>
<tr>
<td>LiNbO₃</td>
<td>10⁻²-10²</td>
<td>10⁻⁸-10⁻¹⁹</td>
<td>(0.03-0.6)·10⁻¹²</td>
<td>2.4-7.2</td>
<td>400-15000</td>
</tr>
<tr>
<td>BaTiO₃</td>
<td>2.4</td>
<td>1.3·10⁻¹²</td>
<td>10⁻¹¹-10⁻⁹</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Bi₁₂SiO₂₀</td>
<td>1.3-3</td>
<td>10⁻¹⁵</td>
<td>1·10⁻⁷</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Bi₁₂GeO₂₀</td>
<td>1.3-3</td>
<td>10⁻¹⁴</td>
<td>0.8·10⁻⁷</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
From the photorefractive experiments, i.e. beam coupling, it was possible to determine the sign of the dominant carriers. Holes were found to be dominant in iron doped as grown crystals whereas in undoped crystals a competition between holes and electrons was observed. In reduced crystals (undoped as well as iron doped samples) electrons are the dominant charge carriers. In addition the number density of acceptors \( N_A \) which corresponds to the number of ionized donors in the dark was determined. In pure as grown as well as in reduced crystals \( N_A \) was found to be in the order of \( 0.5 \times 10^{16} \text{cm}^{-3} \). In iron doped as grown crystals \( N_A \) was \( 1-2 \times 10^{16} \text{cm}^{-3} \). In the reduced iron doped sample \( N_A \) was increased by a factor of about \( 10^2 \) \( (N_A=87 \times 10^{16} \text{cm}^{-3}) \). Table 3.7 gives a summary of these data.

<table>
<thead>
<tr>
<th>Fe-conc.</th>
<th>Treatment</th>
<th>sign of dominant carriers</th>
<th>( N_D=N_{Fe} ) ( (\text{cm}^{-3}) )</th>
<th>( N_A \times (N_D-N_A)/N_D ) ( (\text{cm}^{-3}) )</th>
<th>( N_A )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(&lt;1\text{ppm})</td>
<td>as grown</td>
<td>(+/-)</td>
<td>(&lt;1.5 \times 10^{16})</td>
<td>0.6 \times 10^{16}</td>
<td></td>
</tr>
<tr>
<td>(&lt;1\text{ppm})</td>
<td>reduced</td>
<td>(-)</td>
<td>(&lt;1.5 \times 10^{16})</td>
<td>0.3 \times 10^{16}</td>
<td></td>
</tr>
<tr>
<td>150ppm</td>
<td>as grown</td>
<td>(\dagger)</td>
<td>(230 \times 10^{16})</td>
<td>1 \times 10^{16}</td>
<td>(1 \times 10^{16})</td>
</tr>
<tr>
<td>150ppm</td>
<td>reduced</td>
<td>(-)</td>
<td>(230 \times 10^{16})</td>
<td>(87 \times 10^{16})</td>
<td>(\approx N_D/2)</td>
</tr>
<tr>
<td>300ppm</td>
<td>as grown</td>
<td>(\dagger)</td>
<td>(460 \times 10^{16})</td>
<td>(2 \times 10^{16})</td>
<td>(2 \times 10^{16})</td>
</tr>
</tbody>
</table>

The models used for the calculation of the data of Tab.3.7 were all "one center models". The data obtained from fitting with these models are in agreement with the assumption that iron substitutes are the relevant defects responsible for the photorefractive effect. In Tab.3.7 we have identified, therefore, the donor concentration \( N_D \) with the total iron concentration \( N_{Fe} \). From Tab.3.7 we see, in addition, that in iron doped as grown crystals the ratio \( Fe^{3+}/Fe^{2+}=230 \), whereas in reduced samples this ratio can be lowered to \( Fe^{3+}/Fe^{2+}=1 \). This decrease of the \( Fe^{3+} \) concentration is confirmed by the EPR experiments of Sect.3.4.3.
The conductivity data together with the results obtained from beam coupling experiments ($N_A$ and $\rho_{\text{eff}}$) of Tab. 3.4 and 3.5 characterize the photorefractive behaviour of these crystals as discussed in Sect. 2. These methods represent, therefore, a fast and non-destructive way of a photorefractive characterization.

These data, however, cannot give a full insight into the microscopic structure of the relevant defect centers. Several defect types, present in $\text{ABO}_3$ compounds [3.18], can have an additional influence on the photorefractive response. Possible defects are:

0-dimensional defects:
1. Schottky defects (vacancies)
2. Elementary non-stoichiometry $\text{ABO}_3\pm x$
3. Binary non-stoichiometry $\text{AO(BO}_2)_1\pm x$
4. Substitutions: iso- or heterovalent (higher or lower valency).

1-dimensional defects:
1. Dislocations

2-dimensional defects:
1. Ferroelectric domains
2. Stacking faults

3-dimensional defects:
1. Inhomogeneity of the distribution of 0 dimensional defects, e.g. dopants.
2. Strain

In the present work only the substitution of niobium by iron has been investigated, which can be considered as the most important defect center influencing the photorefractive effect in nominally pure and doped crystals.
4. Isotropic Diffraction and Beam Coupling in Reduced KNbO₃ Crystals

4.1 Grating build-up in reduced KNbO₃

Real-time optical information processing, optical phase conjugation and dynamic holography require strongly nonlinear optical materials among which photorefractive materials belong to the most promising candidates. However, in most of the photorefractive materials investigated up to now the recording time required to build up the refractive index changes or space-charge fields is too long to be useful for processing time varying images in parallel. The most photosensitive photorefractive materials in the visible spectral range are Bi₁₂SiO₂₀ [4.1] and reduced KNbO₃ [4.2, 4.3]. The latter one has the advantage of showing much larger refractive index changes due to its electro-optic coefficients which are up to two orders of magnitude larger than those in the sillenite.

The speed of the photoinduced grating build-up depends on the reduction state of photoactive defects. We are primarily interested in a rapid redistribution of photoinduced charge carriers. This is the case in materials having a small concentration of trapping centers \( N_A \) where the drift length \( L_d = \Phi \mu E \) is large (comparable to the grating fringe spacing; \( \mu \): carrier mobility, \( \tau = (\gamma_R N_A)^{-1} \): carrier life time, \( \gamma_R \): carrier recombination constant, \( E \): electric field). The relative concentration of donor (\( N_D \)) and acceptor centers (\( N_A \)) can be influenced in oxygen octahedra ferroelectrics by reduction as discussed in Sect. 3.2. For that reason investigation of the physical properties of photoactive centers and their influence on photorefractive nonlinearities are essential for optimizing the relevant properties of photorefractive crystals for different applications.

In Fe-doped as well as in nominally pure KNbO₃ crystals the photorefractive effect is ascribed to Fe-impurities creating donor- and trapping centers with energy levels between the valence and conduction band (Sect. 3). In KNbO₃ a comparison of ionic radii implies that the Fe-ions are at the Nb⁵⁺ sites combined with oxygen vacancies for charge compensation. In the simplest model iron appears in the two valence states Fe²⁺ and Fe³⁺. If the free charge carriers are electrons then Fe²⁺ acts as donor center and Fe³⁺ provide the corresponding trapping centers. In contrast in the case of hole transport Fe³⁺...
are the donor centers and Fe$^{2+}$ are the corresponding trapping sites. In the as grown state the concentration of Fe$^{3+}$ usually is higher than that of Fe$^{2+}$ (Fe$^{3+}$/Fe$^{2+}$ $\approx$ 1 - 10$^{+2}$) and we, therefore, find holes to be the dominant free charge carriers.

In the case of grating periods which are large compared to the diffusion length and the Debye screening length, the time constant for the build-up of photorefractive gratings can be approximated by the dielectric relaxation time (see Sect.2.1.2)

$$\tau_{dl} = \frac{\varepsilon_0 \varepsilon}{\sigma_d \sigma_{ph}}$$  \hspace{1cm} (4.1)

where $\varepsilon_0$ is the permittivity of the vacuum, $\varepsilon$ the dielectric constant, $\sigma_d$ the dark conductivity and $\sigma_{ph}$ the photoconductivity given by Eq.(3.6)

$$\sigma_{ph} = \sigma_h + \sigma_e = \frac{eI}{h \nu} (\alpha_h \Phi_h \mu_h \tau_{Rh} + \alpha_e \Phi_e \mu_e \tau_{Re})$$  \hspace{1cm} (4.2)

For grating periods in the order of 1$\mu$m this dielectric time constant is a good approximation for the build-up time of the grating in doped and pure as grown KNbO$_3$ crystals.

For KNbO$_3$ the dark conductivity $\sigma_d$ is in the range of 10$^{-9}$ to 10$^{-14}$ (Vcm)$^{-1}$ and $\Phi_{\mu}t$ of 1 $\cdot$ 10$^{-7}$ to 5 $\cdot$ 10$^{-12}$ cm$^2$/V. Whereas the smallest values of $\sigma_d$ as well as of $\Phi_{\mu}t$ are found in as grown pure and doped crystals, the largest conductivities are measured in highly reduced samples. This is due to the larger mobility of the electrons and to the increase of the absorption constant by reduction.

For that reason it is obvious that for an efficient decrease of the time constant the crystals have to be reduced, which in KNbO$_3$ crystals means the transfer of Fe$^{3+}$ sites into the valence state Fe$^{2+}$.

For strongly reduced KNbO$_3$ crystals the dielectric time constant $\tau_{dl}$ is no longer a good approximation for the build-up time of the grating. More general expressions have been derived in Sect.2.2. For highly photoconductive materials the time for photogenerating the necessary number of free charge carriers for a given space-charge (see Eq.(2.70)) is the limiting
To compare the experimental results of the measured time constants with this theoretical limit, we rewrite Eq. (4.3)

\[ \tau_{\text{lim}} = \frac{\hbar}{l\alpha} \frac{N}{N_D} (N_D - N_A) \]  

(4.3)

where \( N_p = N/\Phi \) is the number of photoexcitations per unit volume and \( N \) the number density of separated charge particle pairs.

In our reduction experiments we were able to prepare reduced KNbO\(_3\) crystals with a photoconductivity up to \( \sigma_{\text{ph}} = 10^{-4} \cdot l \) (\( \Omega \text{cm} \))\(^{-1} \), where \( l \) is given in W/cm\(^2\). Such crystals were used for the investigation of the diffraction efficiency and the time response of photoinduced gratings. The experimental

![Diagram of experimental setup](image)

**Fig. 4.1** Experimental set-up for the investigation of the diffraction efficiency and time constants of photoinduced gratings in photorefractive KNbO\(_3\).
set-up is shown in Fig. 4.1. An acousto-optic modulator was used as a fast shutter for the Ar$^+$ laser beam recording the photoinduced grating. The time behaviour of the diffraction efficiency was investigated by detecting the diffracted light of a HeNe laser beam with a photomultiplier and an oscilloscope. The photoinduced grating spacing used in our experiments was $\Lambda = 2\mu m$ for a writing wavelength of $\lambda = 488\text{nm}$.

The time evolution of the diffraction efficiency $\eta$ after switching on the writing beams can - in the simplest case - be described by an exponential law (square of Eq.(2.61) because of $\eta \sim E^2_{sc}$)

$$\eta(t) = \eta_0 (1 - e^{-t/\tau})^2$$

where $\eta_0$ is the saturation value and $\tau$ the exponential time constant.

In our experiments $\tau$ was determined for different writing intensities from the signal recorded on the oscilloscope. Figure 4.2 shows the experimental results of the diffraction efficiency and build up time as a function of the writing intensity. The solid curve which follows the experimental points of the diffraction efficiency is a smoothed curve. From that curve we calculated the corresponding curve for the limiting time constant $\tau_{lim}$ with Eq.(4.4) in the following way:

The refractive index modulation $\Delta n$ was determined by using Kogelnik's formula for the diffraction efficiency of thick phase gratings [2.17]

$$\Delta n = \frac{\lambda_r}{d\pi} \text{asin}(e^{\alpha d} \sqrt{\eta})$$

where $d$ is the crystal thickness and $\alpha$ the absorption constant at the read-out wavelength $\lambda_r$.

The amplitude of the photoinduced space-charge field can be expressed in terms of the electro-optic parameters by

$$E_{sc}^{o} = \frac{2\Delta n}{n_3^2 r_{33}}$$
Fig. 4.2 Diffraction efficiency (o) and grating build-up time (•) in a highly reduced KNbO₃ crystal. The full lines, calculated from the smoothed curve following the experimental data of the diffraction efficiency, show the theoretical limit for different assumed quantum efficiencies. (Grating spacing $\Lambda=2\mu m$, crystal thickness $d=7.3 mm$)
This space-charge field is caused by a redistribution of the ionized donor sites. The amplitude of this sinusoidal space-charge distribution is given by the Poisson law

\[ \varphi_0 = \frac{2\pi \varepsilon_0 \varepsilon}{\Lambda} E_0 \]  \hspace{1cm} (4.8)

The number density of necessary photoexcitations \( N_p \) can be calculated by integrating Eq.(4.8) over one grating period

\[ N_p = \frac{\rho_0}{\pi \varepsilon} \cdot \frac{1}{\Phi} \]  \hspace{1cm} (4.9)

In Fig.4.2 we have plotted the limiting time constants for different quantum efficiencies according to Eq.(4.3). The agreement with the experimental points is excellent for \( \Phi=36\% \). This leads to the conclusion that in highly reduced KNbO\(_3\) crystals, indeed, the theoretical time limit can be nearly reached.

4.2. Diffraction and beam amplification in reduced KNbO\(_3\) crystals with an applied electric field

In this section we will discuss the influence of externally applied electric fields on the photorefractive behaviour, e.g. on the diffraction efficiency \( \eta \) and the beam coupling gain \( \Gamma \) in reduced KNbO\(_3\) crystals.

For the case of an externally applied electric field \( E_o \), the electric space-charge field was derived for the "one species model" in Sect.2.1.1

\[ E_{sc}^0 = \frac{m}{E_q + E_d + iE_o} \]  \hspace{1cm} (2.35)

By increasing \( E_o \) the amplitude of the space-charge field approaches \((imE_q)\) which is proportional to the fringe spacing \( \Lambda \). The limiting element in Eq.(2.35) for large grating spacings without applied field is the diffusion field \( E_d \). An applied electric field can, therefore, strongly increase the amplitude of the steady state space-charge field. In reduced KNbO\(_3\) - characterized by small \( N_A \), large photoconductivities and fast response times - the amplitude
of the induced space-charge field in the pure diffusion case is rather small. Applying an electric field to such crystals can lead to the desired combination of high diffraction efficiency and fast response times. In Fig. 4.3 the electric field dependence of the diffraction efficiency in a reduced KNbO₃ crystal (r eff=24pm/V and Na=0.4·10¹⁸cm⁻³) for a fringe spacing of Λ=8.4μm (E_d = 0.19 kV/cm and E_q = 18 kV/cm) is shown. The full line is a theoretical fit using the Kogelnik formula [2.17]

\[ \eta = e^{-\alpha d} \sin^2 \left( \frac{\Delta n \cdot d \cdot \pi}{\lambda_r} \right) \] (4.8)

where \( d \) is the crystal thickness, \( \alpha \) the absorption constant for the read out wavelength \( \lambda_r \), and \( \Delta n \) the amplitude of the photoinduced phase grating given by

\[ \Delta n = \frac{1}{2} n_3^3 r_{33} |E_{dc}| \] (4.9)

where \( E_{dc} \) the amplitude of the space-charge field is given by Eq.(2.35).

Fig. 4.3 Electric field dependence of the diffraction efficiency in a reduced KNbO₃ crystal at a fringe spacing Λ=8.4μm. (Writing wavelength \( \lambda_o = 488\text{nm} \).)
As stated at the end of Sect. 2.4.3, the exponential gain $\Gamma$ Eq. (3.18) in the case of an externally applied field $E_o$ is

$$\Gamma = \frac{2\pi n^3}{\lambda \cos \theta} r_{\text{eff}} \frac{1}{m} \text{Im}(E_{sc}^o) \cos(2\theta p)$$

(4.10)

where $\text{Im}(E_{sc}^o)$ is the imaginary part of the space-charge field Eq. (2.35). For increasing electric field strength this imaginary part approaches $(m_{E_q})$

$$\text{Im}(E_{sc}^o) = m \frac{E_q [E_d (E_q + E_d) + E_{sc}^2]}{(E_q + E_d)^2 + E_{sc}^2}$$

(4.11)

In reduced crystals the beam coupling efficiency for large grating spacings can be increased by applying an electric field for the same reasons as in the case of diffraction discussed above. Figure 4.4 shows the field dependence of the gain $\Gamma$ in a reduced KNbO$_3$ crystal for $\Lambda = 8.9 \mu m$ ($E_d = 0.18$ kV/cm and $E_q = 19$ kV/cm). The full line is a theoretical fit using Eq. (4.10) with the parameters $r_{\text{eff}} = 24 \text{pm/V}$ and $N_A = 0.4 \times 10^{16} \text{cm}^{-3}$.

![Electric Field Dependence of Gain in a Reduced KNbO$_3$ Crystal](image)

**Fig. 4.4** Electric field dependence of the gain $\Gamma$ in a reduced KNbO$_3$ crystal for fringe spacing $\Lambda = 8.9 \mu m$ (writing wavelength $\lambda_o = 515$nm). The full line is a theoretical fit using Eq. (4.10).
One sees from Fig. 4.4 that for a grating spacing $\Lambda = 8.9\mu m$ and an applied field $E_0 = 3kV/cm$ the gain is $\Gamma = 2cm^{-1}$. The gain can be further increased by increasing the diffusion field, e.g. by decreasing the fringe spacing. For a grating spacing $\Lambda = 1\mu m$ a value of $\Gamma = 3cm^{-1}$ has been found experimentally for $E=0$. For further increasing the gain larger electric field strength are required. In Ref.[1.32] a gain of up to $15cm^{-1}$ has been observed at a field strength of $E=20kV/cm$. Another technique already used in Bi$_{12}$SiO$_{20}$ and Bi$_{12}$GeO$_{20}$ [4.8-4.10] is the "moving grating" method already mentioned in Sect.2.1.2. It could also lead to an increased gain in reduced KNbO$_3$ crystals.
5. Anisotropic Diffraction and Light Scattering in KNbO₃

5.1 Anisotropic Diffraction in KNbO₃

The anisotropic Bragg angles occurring in biaxial crystals have been discussed in Sect. 2.3.4. Experimental measurements of these angles as a function of the photoinduced grating spacing were performed in a- and b-plates of KNbO₃ (Fig. 5.1). The gratings were recorded using the electro-optic coefficients \( r_{42} \) and \( r_{51} \) respectively. Figure 5.2 shows the experimental setup used for an investigation of the anisotropic diffraction.

![Diagram of anisotropic Bragg diffraction](image)

Fig.5.1 The two main geometries for anisotropic Bragg diffraction by a photoinduced grating in KNbO₃-crystals using the large electro-optic coefficients \( r_{42} \) and \( r_{51} \) respectively.

![Experimental setup diagram](image)

Fig.5.2 Experimental set-up for the investigation of the anisotropic Bragg diffraction in photorefractive KNbO₃ crystals.
Fig. 5.3 Measurement of the incident and diffraction angles $\theta_i$ and $\theta_d$ (outside the crystal) for anisotropic Bragg diffraction in an $a$-plate of KNbO$_3$ (electro-optic coefficient $r_{42}$). The full lines are theoretical curves calculated by using (2.109-2.112).
Fig. 5.4 Measurement of the incident and diffraction angles $\theta_i$ and $\theta_d$ (outside the crystal) for anisotropic Bragg diffraction in a b-plate of KNbO$_3$ (electro-optic coefficient $r_{51}$). The full lines are theoretical curves calculated by using (2.109-2.112).
In Figs. 5.3 and 5.4 the experimental values are shown. The full lines are the theoretical curves calculated using Eqs. (2.109)-(2.112) derived in Sect. 2.3.4. The excellent agreement between experimental results and theoretical calculation is due to the very accurate knowledge of the refractive indices of KNbO₃ [1.28].

Fig. 5.5 shows the intensity dependence of the efficiency of anisotropic diffraction in an iron doped KNbO₃ b-plate (KNbO₃:Fe 300ppm). The full line is a theoretical curve using the relation Eq.(2.152) which for fullfilled Bragg condition can be written as

\[ \eta = A \cdot \sin^2 (c \cdot M \lambda_0) \]  

(5.1)

where \( c \) is a constant, \( M \) the modulation ratio and \( \lambda_0 \) the mean intensity of the fringes. \( M \), given by Eq.(2.19), is dependent on the ratio of dark- and photoconductivity \( M = m/(1+\sigma_d/\sigma_{ph}) \) and therefore the diffraction efficiency is intensity dependent up to high intensities were it reaches a saturation value \( \sigma_d \ll \sigma_{ph} \) and hence \( M = m \). A peak value of \( \eta = 45\% \) has been reached at a writing intensity of 2.4 W/cm².
5.2 Anisotropic light scattering in photorefractive materials

Anisotropic scattering of a linearly polarized laser beam into a conical ring of different polarization has been observed in LiTaO$_3$:Cu [5.1] and BaTiO$_3$ [5.2, 5.3]. The intensity within the ring is maximal in the direction perpendicular to the polar c-axis and zero along this axis, as shown in the photographs of Fig. 5.6.

Fig. 5.6 Anisotropic scattering of a beam polarized along the polar c-axis entering the crystal at normal incidence. The photographs illustrate the scattering into a ring of light polarized perpendicular to the c-axis in BaTiO$_3$, LiNbO$_3$ and KNbO$_3$.

When a single laser beam is propagating in a crystal, a part of its intensity will be scattered at inhomogeneities, impurities, etc. These scattered waves, which mostly have the same polarization as the strong incident beam, create "noisy" photorefractive gratings of small amplitude by interference with the incident beam. Some of these gratings are capable of diffracting anisotropically the strong incident beam. Figure 5.7 illustrates the phase matching condition for this type of scattering (for simplicity drawn in the (x,y)-plane of a negative uniaxial crystal such as BaTiO$_3$). The incident beam $k_i$ scatters in many directions, but only two of the scattered beams $k_{isc}$ lie at an angle $\Theta_{sd}$ so that the grating vector $K_g$ allows an anisotropic self-diffraction of the incident beam.
Fig. 5.7 Phase matching condition for anisotropic light scattering at photo-induced noisy gratings in an uniaxial crystal.

The scattering angle for negative uniaxial crystals is given by [5.4]

\[
\sin(\phi_{sc}') = \sqrt{n_0^2 - \left[\frac{3n_e^2 - n_0^2}{4n_e}\right]^2} \tag{5.1}
\]

\(\phi_{sc}'\) is the scattering angle measured outside the crystal.

The disappearance of scattered light along the c-axis is easy to explain by the anisotropic self-diffraction. A noisy intensity grating along the c-direction cannot induce an anisotropic phase grating as described in Sect. 2.3.3. In the crystals discussed here (see Table 5.1) no non-vanishing electro-optic coefficient of the form \(r_{ij3}\) (with \(i \neq j\)) can exist for symmetry reasons and therefore no diffraction or scattering may take place in this direction.

Table 5.1 shows our experimental results and theoretical calculations of the scattering angle \(\phi_{sc}'\) for \(\text{KNO}_3\) and some other crystals for comparison.
Table 5.1 Anisotropic scattering angle for different photorefractive crystals (Incident wavelength 514.5nm, room temperature)

<table>
<thead>
<tr>
<th>Material</th>
<th>Experimental scattering angle $\psi_{sc}$</th>
<th>Theoretical scattering angle $\psi_{sc}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaTiO$_3$</td>
<td>23.6°</td>
<td>21.0°</td>
</tr>
<tr>
<td>LiNbO$_3$</td>
<td>27.3°</td>
<td>26.6°</td>
</tr>
<tr>
<td>KNbO$_3$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>i) 38.8°</td>
<td>39.0°</td>
</tr>
<tr>
<td></td>
<td>ii) 40.7°</td>
<td>40.2°</td>
</tr>
</tbody>
</table>

1) measured in the (001)-plane
2) measured in the (01 1)-plane
6. Optically Induced Variable Light Deflection by Anisotropic Bragg

Diffraction in Photorefractive K\textsubscript{NbO\textsubscript{3}}

Laser-induced dynamic gratings can be used for deflecting other light waves [6.1-6.4]. An opto-optical deflector operates in a similar way as acousto-optic devices. While their speed is limited by the sound velocity much faster response times can be obtained by using laser-induced gratings.

In this section we will discuss the use of anisotropic Bragg diffraction for opto-optical light deflection. Diffraction geometries near the minimal incidence angle \( \theta_{i}^{\text{min}} \) are applied for such an application (see Sect.2.3.4). In this configuration, as shown in Fig.6.1, the diffraction angle \( \theta_{d} \) is close to zero degrees.

The broad minimum in the angle of incidence around \( K_{g}^{*} \) (see Fig.6.1a) is very useful in light deflectors, because the light incident at a fixed angle can be effectively deflected within a broad range of grating wavenumbers \( K_{g} \) (or recording wavelengths \( \lambda_{g} \)), resulting in a large range of deflection angles. For a small variation of the grating wave number \( K_{g} \) the incidence Bragg angle does not have to be changed whereas the deflection angle can vary considerably (Fig.6.1).
We see from Tab. 2.2 that the grating spacings $\Lambda^*$ needed for the minimal incidence angles $\Theta_{\text{min}}^*$ are all in the order of one micrometer, i.e. they can be easily produced by letting visible laser beams interfere in the volume of photorefractive materials.

To find an expression for the change of the diffraction angle $\Theta_d$ as a function of the recording wavelength change at fixed incidence angle $\Theta_i = \Theta_{\text{min}}^*$ we rewrite Eq.(2.114)

$$K_g^* = \frac{n_x}{n_y} \sqrt{n_y^2 - n_z^2} k_r = \frac{4\pi}{\lambda^*} \sin(\Theta) \quad (6.1)$$

where $\lambda^*$ is the recording wavelength and $\Theta$ the angle between the beams inducing the grating with a fringe spacing $\Lambda^* = 2\pi / K_g^*$. With Eq.(6.1) a change of the grating wavenumber $\Delta K_g$ around $K_g^*$ can be written as

$$K_g = K_g^* + \Delta K_g = \frac{4\pi}{\lambda_o^*} \sin(\Theta) \frac{\lambda_o^*}{\lambda_o} \quad (6.2)$$

where $\lambda_o = \lambda_o^* + \Delta \lambda_o$ is the recording wavelength.

The change of the diffraction angle is then given by

$$\Delta \Theta_d = \sin^{-1} \left( \frac{\Delta K_g}{k_r} \right) \quad (6.3)$$

Finally, if we assume that the wavelength change is small in comparison to the recording wavelength ( $\Delta \lambda_o \ll \lambda_o$) we get

$$\Delta \Theta_d' = \frac{\Delta K_g^*}{k_r} = \frac{K_g^*}{k_r} \frac{\Delta \lambda_o}{\lambda_o^*} = \frac{\lambda_r}{\Lambda^*} \frac{\Delta \lambda_o}{\lambda_o^*} \quad (6.4)$$

Figure 6.2 shows experimental data of the change of the deflection angle $\Delta \Theta_d'$ for a wavelength change around the center wavelength $\lambda_o = 488\text{nm}$ for different Ar$^+$ laser wavelengths. The read-out was performed by a HeNe-laser beam ($\lambda_r = 632.8\text{nm}$). The full line is a theoretical calculation of the deflection angle using the refractive index data of KNbO$_3$ from [1.28].
Fig. 6.2 Experimental data of the deflection angle $\Delta \theta_d$ for a wavelength variation around the center wavelength $\lambda_0 = 488\text{nm}$ for the different Ar$^+$ laser wavelengths in a KNbO$_3$ a-plate. The read out was performed by a HeNe-laser beam ($\lambda_r = 632.8\text{nm}$). The full line is a theoretical calculation of the deflection angle using the refractive index data of KNbO$_3$ from [1.28].

Experimental data for the relative diffraction efficiency as a function of the deflection angle $\theta_d$ induced by changing the interaction angle between the writing beams at fixed wavelength $\lambda_0 = 488\text{nm}$ are shown in Fig. 6.3. The measurements were performed in a nominally pure KNbO$_3$ a-plate with a thickness of 2.55mm.

In another experiment a HeNe laser beam incident under the Bragg angle was deflected in several directions by changing the wavelength of the writing beams between 457.9nm and 514.5nm (Ar$^+$-laser lines). Figure 6.3d is a photograph of the diffracted spots for the different wavelengths. The variation of the deflection angle was 5.7$^\circ$ for $\Delta \lambda_0 = 57.7\text{nm}$. Whatever writing wavelength used, no adjustment of the incident angles was required.
The full and dotted lines in Fig.6.3 are theoretical curves fitted by means of Eq.(2.148). The effective thickness of the crystal used for the calculation was 1mm instead of a real thickness of 2.55mm. An explanation for this discrepancy could be the finite divergence of the read out beam (HeNe laser beam with divergence angle $\delta \theta = 1$ mrad) but, most probably, it is predominantly influenced by the inhomogeneous reduction state of the crystal used.

The number of resolvable spots for a divergence angle $\delta \theta$ of the read out beam is

$$ N = \frac{\Delta \theta_d}{\delta \theta} \quad (6.3) $$

For a typical beam divergence $\delta \theta = 1$ mrad of a HeNe laser this leads to a number $N=100$ of resolvable points. This number will be further increased by using optical beams having smaller divergence angles, which finally is given by the diffraction limit of the crystal aperture.

To stay within a 50% range of the maximum diffraction efficiency for all wavelengths used, a slight increase of the Bragg angle (about 0.07° in our case) is needed to enlarge the width of the curve in Fig.6.3a. This configuration was employed for the measurement of the dotted curve and is explained by a wave vector diagram in Fig.6.3c. The dip at $\theta_d=0°$ occurs because of the slight off-Bragg adjustment for $\Delta K_g=0$.

Because of the fact that the incidence angle $\theta_i$ and the diffraction angle $\theta_d$ for anisotropic Bragg diffraction depend on the refractive indices (see e.g. Sect.2.3) it is clear that these angles show a temperature dependence determined by the corresponding dependence of the refractive indices. Figure 6.4 shows a measurement of the temperature dependence of the diffraction angle $\theta_d$ in a KNbO$_3$ a-plate. The grating spacing was adjusted to give a zero diffraction angle $\theta_d=0$ at room temperature. The full line in Fig.6.4 is a theoretical calculation using the refractive index data of Ref.[1.28]. The discrepancy between experimental points and the theoretical curve seems to be due to the accuracy of the refractive index data above room temperature.
CONTROL WAVELENGTH $\lambda_r$ (nm)

514.5 589.7 496.5 488.0 476.5 472.7 465.8 467.9

WAVELENGTH CHANGE $\Delta \lambda_r$ (nm)

DEFLECTION ANGLE $\theta_d$ (DEG)

DIFFRACTION EFFICIENCY (%)
Fig. 6.4  Experimental data of the temperature dependence of the diffraction angle $\theta_d$ near the non-critical configuration. The grating spacing was adjusted to have a zero diffraction angle $\theta_d = 0$ at room temperature. The full line is a theoretical calculation using the refractive index data of Ref. [1.28].

Fig. 6.3  a) Diffraction efficiency and deflection angle for anisotropic Bragg-diffraction by photorefractive gratings in KNbO$_3$.
- $\bigcirc$, $\bullet$ : Recording at fixed wavelength $\lambda_o = 488$ nm and fixed incidence angle $\theta_i = 56.23^\circ$ (o) and $\theta_i = 56.30^\circ$ (•); tuning of the deflection angle by changing the angle $2\theta$ between the recording beams.
- •: Recording at fixed angle $2\theta = 36.9^\circ$ between the recording beams and fixed incidence angle $\theta_i = 56.23^\circ$; tuning of the deflection angle by changing the recording wavelength.

b) Wave vector diagram for the experimental configuration of the (o, •)-points.
c) Wave vector diagram for the experimental configuration of the (•)-points.
d) Photograph of the diffracted spots for six different recording wavelengths (Ar$^+$-laser lines).
From the experimental data of Fig.6.4 a temperature dependence

\[ \Delta \theta'_d = 3.6 \cdot 10^{-4} \cdot \Delta T \]  

(6.5)
can be calculated.

Grating spacings of photoinduced gratings for anisotropic Bragg diffraction in a KNbO₃ crystal (a-plate, \( r_{42} \) active) and the resulting incidence and diffraction angles for read-out at different wavelength (\( \lambda = 633 \text{ nm}, 850 \text{ nm}, 1300 \text{ nm} \) and 1550 nm) are shown in Fig.6.5. These wavelengths correspond to the He-Ne laser line (633 nm) and to important semiconductor laser lines (850 nm, 1.3 \( \mu \text{m} \) and 1.55 \( \mu \text{m} \)). In Table 6.1 the grating spacings \( \Lambda \) and the incidence angles \( \theta'_i \) for zero diffraction angle and the relative diffraction angle changes \( \Delta \theta'_d / (\Delta \lambda_o / \lambda_o^* \) are listed for a- and b-plates of KNbO₃.

<table>
<thead>
<tr>
<th>Wavelength (( \mu \text{m} ))</th>
<th>Crystal Orientation (active el.opt.coeff.)</th>
<th>( \Lambda^* ) (( \mu \text{m} ))</th>
<th>( \theta'_i ) (deg)</th>
<th>( \Delta \theta'_d / (\Delta \lambda_o / \lambda_o^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.633</td>
<td>a-plate (( r_{42} ))</td>
<td>0.76</td>
<td>56.2</td>
<td>0.83</td>
</tr>
<tr>
<td></td>
<td>b-plate (( r_{51} ))</td>
<td>0.88</td>
<td>45.9</td>
<td>0.71</td>
</tr>
<tr>
<td>0.85</td>
<td>a-plate (( r_{42} ))</td>
<td>1.09</td>
<td>51.6</td>
<td>0.78</td>
</tr>
<tr>
<td></td>
<td>b-plate (( r_{51} ))</td>
<td>1.25</td>
<td>43.2</td>
<td>0.68</td>
</tr>
<tr>
<td>1.3</td>
<td>a-plate (( r_{42} ))</td>
<td>1.73</td>
<td>48.7</td>
<td>0.75</td>
</tr>
<tr>
<td></td>
<td>b-plate (( r_{51} ))</td>
<td>1.96</td>
<td>41.5</td>
<td>0.66</td>
</tr>
<tr>
<td>1.55</td>
<td>a-plate (( r_{42} ))</td>
<td>2.08</td>
<td>48.2</td>
<td>0.75</td>
</tr>
<tr>
<td></td>
<td>b-plate (( r_{51} ))</td>
<td>2.35</td>
<td>41.2</td>
<td>0.66</td>
</tr>
</tbody>
</table>
Fig. 6.5 Fringe spacing for photoinduced gratings and incidence and diffraction angle for anisotropic Bragg diffraction in a KNbO$_3$ crystal (a-plate, $r_{42}$ active) for different read out wavelengths.
7. Optically Addressable Fiber Interconnection by Anisotropic Bragg Diffraction in Photorefractive KNbO₃

7.1. Introduction

Within the last decade there has been a rapid transition from electromechanical switching of communication channels to solid state computer-controlled switching systems for public and private telephone exchanges. These systems offer high reliability and flexible service facilities, but they are limited to baseband voice channels or relatively slow digital data links. During the same period data communication rates have increased significantly and the use of wideband video channels has become widespread. These speed-related factors have helped to enhance the acceptance of fiber optic data links.

Fiber-optic communication systems that fully utilize the possibilities of optical fibers use components such as connectors, couplers, attenuators, switches, isolators, and wavelength division (de-) multiplexers.

In today's communication technology switching is done electronically. Although the development of optical fiber transmission now permits signals in the form of light to be transmitted cheaply and reliably, the light is converted to electrical signals at each end of the fiber. But, as the trend towards still higher bit rates (and shorter times per bit) continues, it will become more and more attractive to perform some optical signal processing operations, i.e. to use photonics in place of electronics.

In a first step it is necessary to have optical fiber switches. Not long ago only mechanical switches for routing signals from one optical fiber to another were available commercially. With such devices low-loss operation can be obtained but their speed is limited in the millisecond regime.

A number of electro-optic switching and modulation elements have been proposed and demonstrated in recent years, and some are now available commercially. Most work has been done on integrated optical LiNbO₃ devices [7.1-7.3]. Such integrated electro-optic switch arrays on LiNbO₃ were developed up to a size of 8x8 ports, they present the most advanced electro-optic device today.
Light deflection by photoinduced gratings can be applied for the build-up of an optically addressable interconnection between fiber matrices. Such devices using isotropic Bragg diffraction have been proposed by PAULIAT et al. [7.4].

In this Section I will discuss the concept and present experimental results of an optically addressable fiber switching device using anisotropic Bragg diffraction in reduced $\text{KNbO}_3$ crystals.

### 7.2. Device description and discussion of the limiting factors

The switching device in a $1\times N$ configuration, schematically represented in Fig.7.1, consists of an input fiber and a linear array of $N$ output fibers. The light beam leaving the input fiber is collimated by the lens $L_1$ and partially diffracted by the photoinduced grating in the $\text{KNbO}_3$ crystal. The diffraction angle $\theta_d$ is predetermined by the wavelength of the writing beams while the incidence angles are kept constant (see Sect.6). The diffracted beam is focused onto the tip of one of the output fibers by a second lens $L_2$. By changing the wavelength of the writing beams any one of the $N$ fibers can be addressed.

![Fig.7.1 Set-up of an optically controllable fiber interconnection system.](image)

By changing the control wavelength (dye laser) the signal beam can be coupled into any one of the linearly arranged fibers.
The maximum possible linear dimension \( w_{\text{max}} \) of the fiber array for a given focal length \( f_2 \) of the lens L2 (see Fig. 7.2) is given by

\[
w_{\text{max}} = N \cdot d_{\text{cl}} = f_2 \cdot \Delta \theta'
\]  

(7.1)

where \( N \) is the number of linearly arranged fibers with a cladding diameter \( d_{\text{cl}} \) and \( \Delta \theta' \) is the deflection range of an anisotropically Bragg diffracted beam in the non-critical configuration Eq.(5.4).

Fig. 7.2 Coupling geometry for optimal fiber switching with anisotropically Bragg diffraction in a non-critical configuration.

With Eq.(7.1) it seems to be possible to reach more and more fibers of a given diameter by increasing the focal length \( f_2 \). The focal length is limited, however, by the demand of a high efficiency for coupling into the output fiber. Large coupling losses are to be expected, whenever the diffraction limited focal spot size \( s \) is larger than the core diameter of the fiber. The spot size \( s \) is given by

\[
s \approx \frac{\lambda f_2}{a_{\text{lim}}} \leq d_{\text{core}}
\]  

(7.2)

where \( \lambda \) is the wavelength of the information beam, \( a_{\text{lim}} \) is the limiting aperture of the system (crystal- or lens aperture or beam diameter), and \( d_{\text{core}} \) the core diameter of the fiber.
With Eqs. (7.1) and (7.2) we find the final condition for the focal length $f_2$

$$\frac{N \cdot d_{cl}}{\Delta \theta^*} \leq f_2 \leq \frac{d_{core} \alpha_{lim}}{\lambda} \tag{7.3}$$

Fig. 7.2 shows pictures of a fiber array fabricated as a prototype by VOLPL AG in Schlieren. This array consists of 128 linearly arranged multimode fibers having a cladding diameter of 70\,\mu m and a core diameter of 50\,\mu m. With a deflection range of $\Delta \theta^* \approx 3.5^\circ$ a minimal focal length $f_2 = 150\,\text{mm}$ was chosen. Figure 7.2a shows the array mounted on an optical positioning device. Fig. 7.2b is a microscopic image of a part of the array front, illustrating the linear arrangement of the fibers. The mounting accuracy is about $\pm 1.5\,\mu m$.

Another limiting element for this device is the stability and bandwidth of the control wavelength. Under the condition that the deflection width $\Delta \theta_0^*$ has to be smaller than the core diameter of the fiber, we find with Eq. (6.4)

$$\frac{\lambda \cdot \Delta \lambda_0}{\lambda \cdot \lambda_0} \leq \frac{d_{core}}{f} \tag{7.4}$$

where $\Delta \lambda_0$ is the bandwidth of the control beam. Using a dye laser with a simple birefringent filter, a spectral bandwidth of typically 35\,GHz or $\Delta \lambda_0 = 0.42 \cdot 10^{-10}\,\text{m}$ can be obtained. With this the limit of the bandwidth is less critical than the diffraction limit Eq. (7.3).

Finally, the temperature dependence of the diffraction angle leads to the need of a good thermal stability of the diffracting crystal. With Eq. (6.5) we find

$$\Delta T < 2800 \cdot \frac{d_{core}}{f} \quad (^\circ \text{C}) \tag{7.5}$$

which, for a core diameter of 50\,\mu m and a focal length of 150\,mm, leads to a required temperature stability of $\Delta T < 1^\circ$.

Fig. 7.3 shows a sequence of the fully installed switching set up. The photo-induced grating is written by a Spectra Physics dye laser using Rhodamin 6G. By changing the laser wavelength, the diffracted HeNe-laser beam is focussed onto the tip of different fibers. The selected fiber then can be identified easily at the linearly arranged fiber ends.
Fig 7.2  a) Fiber array mounted on an optical positioning device.  
b) is a microscopic image of a part of the array front, illustrating 
the linear arrangement of the fibers.
Fig. 7.3 Sequence of the fully installed switching set-up. The photoinduced grating is written by a Spectra Physics Rhodamine 6G dye laser. By changing the laser wavelength the diffracted HeNe-laser beam is focussed onto the tip of different fibers. The selected fiber then can be identified easily at the linearly arranged fiber ends.
7.3 Device Performance

Using the switching device described above one can switch one input channel into any of the 128 output channels by simply adjusting the control or writing wavelength to its appropriate value. The occurring crosstalk to an adjacent fiber is found to be less than -21 dB. This crosstalk is mainly caused by the fact that the diffracted beam has a strong deviation from the plane wave limit.

The overall switching efficiency is dominated by the diffraction efficiency of the photorefractive crystal. The need for fast switching requires reduced KNbO$_3$ samples. Experimentally an overall switching efficiency of

$$\eta_{\text{switch}} \approx 0.35 \cdot \eta_{\text{diffraction}}$$  \hspace{1cm} (7.6)

was found.

Using pure, reduced crystals, switching efficiencies in the order of 1% and switching times down to the range of 10$\mu$s can be found. By applying reduced iron doped KNbO$_3$-crystals switching efficiencies of 10% and switching times of a few milliseconds can be obtained (with reduced KNbO$_3$:Fe(150ppm) a diffraction efficiency $\eta$=4.7% and a build-up time of $\tau$=6ms at $l=1$W/cm$^2$ has been found for a crystal thickness $d=1.3$mm). Fig.7.4 shows experimental results using a reduced KNbO$_3$ crystal with a thickness $d=2$mm and a Rhodamine 6G dye laser as the source of the control wavelength. The diffraction geometry in this experiment is similar to the one shown in Fig.6.3c.
Fig. 7.4 Switching efficiency and control wavelength for the 128 linearly arranged output fiber channels. In this experiment a pure, reduced KNbO\textsubscript{3} crystal having a thickness d=2mm was used.
8. Spatial Light Modulation with Photorefractive Materials

8.1 Introduction

Parallel optical processing of information is one of the main advantages of future optical processors and optical computers. To fully utilize such a parallelism it is necessary to have powerful two-dimensional light modulators. Such modulators have to fulfill a number of requirements concerning speed, resolution and sensitivity. Over the past two decades a large number of electronically and optically addressable devices for impressing informations onto one- and two-dimensional optical wavefronts have been proposed. These spatial light modulators (SLM's) were initially developed for such applications as: projection and flat-screen displays, optical memory, incoherent-to-coherent conversion, optical wavefront conversion and time-to-space conversion [8.1].

However, for a pure optical processor it is absolutely necessary to work with optically addressable spatial light modulators. They have to respond to an incident intensity distribution that represents the data field to be processed. This detected intensity is utilized to modify a given material property which finally modulates the amplitude and/or the phase of a read out beam. Table 8.1 gives a list of the most promising optically addressed spatial light modulators proposed up to now.

The photorefractive incoherent-to-coherent converter (PICOC) is a two dimensional spatial light modulator that utilizes modified holographic recording techniques for information storage and reconstruction. It was first proposed by KAMSHILIN et al. [8.10] and later on independently developed by SHI et al. [8.11].

By letting two coherent plane waves interfere in the bulk of a photorefractive crystal one produces an intensity grating and consequently a phase grating. In the photorefractive incoherent-to-coherent conversion (PICOC) or spatial light modulation, in addition to the uniformly induced holographic grating, an incoherent image is focused into the volume of the photorefractive material, creating a spatial modulation of the fringe contrast. It can be transferred onto a coherent readout beam by reconstructing the holographic grating. The spatial modulation of the diffracted coherent beam will be a negative replica of the incoherent input image [8.10-8.12]. In Figs.8.1 and 8.2 the whole process is summarized.
<table>
<thead>
<tr>
<th>NAME/TYP</th>
<th>MODULATING MATERIAL</th>
<th>OPTICAL SENSOR</th>
<th>DEVELOPED AT</th>
<th>RESOL</th>
<th>SENSIT.</th>
<th>TIME RESPONSE</th>
<th>SIZE</th>
<th>OPER. TEMP.</th>
<th>REF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photo-Titus</td>
<td>K*DP</td>
<td>Si Photodiode</td>
<td>Lockheed</td>
<td>10</td>
<td>2</td>
<td>&lt;0.5</td>
<td>5sec</td>
<td>4cm</td>
<td>-50 C</td>
</tr>
<tr>
<td>LCLV</td>
<td>Twisted nematic liquid crystal</td>
<td>CdS</td>
<td>Hughes</td>
<td>30</td>
<td>6</td>
<td>10</td>
<td>15</td>
<td>15msec</td>
<td>2.5x2.5cm</td>
</tr>
<tr>
<td>RUTICON</td>
<td>Deformable Elastomer</td>
<td>Amorph.Se</td>
<td>Xerox</td>
<td>40</td>
<td>30</td>
<td>5</td>
<td>4</td>
<td>15min</td>
<td>Room</td>
</tr>
<tr>
<td>PROM</td>
<td>BSO or BGO</td>
<td>BSO or BGO</td>
<td>USC</td>
<td>6</td>
<td>5</td>
<td>&lt;0.1</td>
<td>2hrs</td>
<td>4cm</td>
<td>Room</td>
</tr>
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Table 8.1 List of optically addressable spatial light modulators
Fig. 8.1 Principles of operation of the photorefractive incoherent-to-coherent optical conversion.
Fig. 8.2 Photorefractive incoherent-to-coherent optical conversion by decreasing the modulation of the writing beams and therefore decreasing the amplitude of the photoinduced space-charge field.
8.2 Theoretical model of Incoherent-to-coherent conversion

The photorefractive incoherent-to-coherent conversion in the case of no externally applied electric field (pure diffusion) can be theoretically described by the following model.

The interference pattern produced by the two writing beams is given by (see Eq.(2.13))

$$I_{coh}(x) = I_{coh}^0 \left( 1 + m \cos(K_g x) \right)$$ (8.1)

where $m$ is the modulation ratio of the two coherent writing beams ($I_0 = I_{+1} + I_{-1}$ and $m = 2 \sqrt{I_{-1} I_{+1}} / I_0$).

An additional incoherent signal beam

$$I_{inc}(x,z) = I_{inc}^0 f(x,z)$$ (8.2)

where $0 \leq f(x,z) \leq 1$ is a two dimensional signal function, leads to a concentration of the photoexcited free carriers similar to Eq.(2.15)

$$n(x,z) = n_d + n_{coh} \left( 1 + m \cos(K_g x) \right) + n_{inc} f(x,z)$$ (8.3)

$n_d$ is the dark concentration, $n_{coh}$ and $n_{inc}$ respectively are given by (see Eq.(2.16))

$$n_{coh} = I_{coh}^0 \frac{s_{coh} (N_D - N_A)}{\gamma R N_A} ; \quad n_{inc} = I_{inc}^0 \frac{s_{inc} (N_D - N_A)}{\gamma R N_A}$$ (8.4)

where $s_{coh}$, $s_{inc}$ are the cross section for photoionization at the writing and signal wavelength respectively.

The stationary photoinduced space-charge field in the non-saturated limit can be calculated from Eq.(2.20)

$$E_{sc} = q \frac{k_B T}{e} \frac{\delta n/\delta x}{n}$$ (8.5)
If the spatial frequencies of the incoherent image are much smaller than the frequency of the grating

$$|\partial f/\partial x| \ll K_g$$  \hspace{1cm} (8.6)

and if the concentration of the thermally induced carriers $n_d$ is neglectable ($n_d \ll n_{coh}$, $n_{inc}$), we get with Eq.(8.5)

$$E_{sc} = q e \frac{k_B T}{e} \frac{K_g m \sin(K_g x)}{1 + \frac{l_{inc}}{l_{coh}} + m \cos(K_g x)}$$  \hspace{1cm} (8.7)

From Eq.(8.7) one can easily see that an incoherent signal leads to a modulation of the amplitude of the photoinduced space-charge field. For high incoherent intensities $l_{inc}$ the space-charge field approaches zero.

Assuming only first order diffraction we find the first order Fourier coefficient of Eq.(8.7) (see Eq.(2.27))

$$E_{sc}^1(x,z) = q e \frac{2k_B T}{e} \frac{K_g (1 - \sqrt{1 - M_{eff}^2}) \sin(K_g x)}{M_{eff}}$$  \hspace{1cm} (8.8)

where

$$M_{eff} = \frac{m}{1 + \frac{l_{inc}}{l_{coh}} f(x,z)}$$  \hspace{1cm} (8.9)

is the effective modulation.

The efficiency of the diffraction from a photoinduced grating depends on the amplitude of the space-charge field, e.g. in a linear approximation $\eta \approx E_{sc}^2$. From Eq.(8.8) and Eq.(8.9) it is obvious that the amplitude of the space-charge field and therefore the diffraction efficiency depends on the ratio $l_{inc}^0/l_{coh}^0$ of the intensities of the incoherent signal beam and the coherent writing beams. The intensity transfer function $\eta(l_{inc})$ is in a nonlinear function of the incoherent intensity. From the fact that $\eta$ approaches zero for high incoherent intensities it is obvious that in this incoherent-to-coherent conversion process a contrast inversion takes place.
8.3 Spatial resolution of the converter

The resolution of the PICOC-modulator is influenced by different factors: the quality of the incoherent projection, the grating induced by the coherent writing beams and the material properties.

i) Incoherent projection

The image projected into the crystal is only well focused in one plane, but efficient diffraction takes place if the grating, and therefore the crystal, has a certain thickness. This leads to a limitation in the resolution which can be determined by the illustration shown in Fig. 8.3. The optimal focal plane occurs in the center of the crystal. An object point projected into the crystal has on its surface a spot diameter \(W\). A resolution criteria can be written as [8.13]

\[
R = \frac{2}{W} = \frac{4nf}{dD}
\] (8.10)

where \(n\) is the refractive index and \(d\) the thickness of the crystal, \(f\) the focal length and \(D\) is the pupil diameter. For example, for \(n=2.2116, f=150\text{mm}, D=40\text{mm}\) and \(d=1\text{mm}\) the limiting resolution is approximately 33 line pairs per mm.

![Fig. 8.3 Geometric constraints imposed on PICOC resolution by the projection optics for the incoherent beam.](image)

ii) Grating produced by the coherent writing beams

The spatial frequency of the photoinduced grating must be significantly larger than the signal bandwidth. Moreover, a beam matching condition,
such as the one illustrated in Fig. 8.4, has to be at least nearly fulfilled. In simple words, the projection direction has to be close to the propagation direction of the diffracted beam in order to have a sufficient resolution.

Fig. 8.4 Illustration of the resolution constraints due to the non-collinearity of the directions of the incoherent illumination and of the diffracted beam.
The resolution limit due to the non-colinearity of the directions of the incoherent projection and the coherent diffracted beam can be estimated by

\[ R \approx \frac{1}{d \cdot \tan(\delta)} \] (8.11)

where \( d \) is the crystal thickness and \( \delta \) the angle between the incoherent projection and the diffracted beam (outside the crystal). For a thickness \( d=1 \text{mm} \) and a detuning \( \delta=5^\circ \) a resolution limit \( R=11 \text{lp/mm} \) is found. Therefore, it is obvious that in an experimental set-up great care should be taken to align the direction of the incoherent beam with that of the diffracted read-out beam.

iii) Material properties

Additional resolution limitations arise from material-dependent parameters influencing the physics of grating formation. The diffraction efficiency is not linearly dependent on the incoherent intensity and, therefore, the resolution depends on both, the incoherent and the coherent intensities. Finally all inhomogeneities of the photorefractive properties in the crystal reduce the achievable resolution. All these constraints are difficult to handle and much more work has to be done to minimize them.

8.4 Anisotropic self-diffraction for spatial light modulation

Anisotropic self-diffraction, already discussed in Sect.2.3.5, is ideally suited for spatial light modulation for two reasons:

a) The photoinduced grating is read-out by one of the writing beams, thus no additional read-out beam is needed, which also could erase the grating.

b) Due to the fact that one of the strong writing beams is diffracted by the self-induced grating, the diffracted beam may have a high intensity even for low diffraction efficiencies.

Figure 8.5 shows the schematic configuration for anisotropic self-diffraction in a \( \text{KNbO}_3 \) (b-plate) as it was used in the experiments to be discussed.
Fig. 8.5 a) Schematic configuration for anisotropic self-diffraction in a KNbO₃ crystal (b-plate). I₊₁ and I₋₁ are the intensities of the two writing beams creating the photoinduced grating. Iₛd is the intensity of the self-diffracted beam.

b) Wave-vector diagram for anisotropic self-diffraction in a KNbO₃ b-plate. θ and φ are the incident and diffraction angles, respectively, inside and θ’ and φ’ those outside the crystal (Fig. a).

Figure 8.6 shows the experimental configuration for incoherent-to-coherent conversion by anisotropic self-diffraction in KNbO₃. An incoherent input image projected into the crystal modulates the diffraction efficiency of the photoinduced grating and, therefore, transfers its information onto the coherent self-diffracted beam.

In a first experiment a reduced KNbO₃ crystal (b-plate) having the dimensions (a,b,c)=(5.2,2.25,6.0)mm was illuminated homogeneously by a commercially available slide projector with a 200-W halogen lamp. The incoherent intensity in the crystal was varied between 0 and 200mW/cm². Figure 8.7a shows the dependence of the intensity of the self-diffracted beam as a
Fig. 8.6 Experimental set-up for incoherent-to-coherent conversion by anisotropic self-diffraction in KNbO₃. An incoherent input image projected into the crystal modulates the diffraction efficiency of the photoinduced grating and therefore transfers its information onto the coherent self-diffracted beam.
function of the incoherent intensity $I_{\text{inc}}$ for different coherent intensities $I_0$. Figure 8.7b shows the diffraction efficiencies calculated from the results of Fig.8.7a as a function of the ratio $I_{\text{inc}}/I_0$. The solid line plotted in Fig.8.7b is a theoretical fit using Eq.(2.148) the diffraction efficiency for anisotropic Bragg-diffraction together with the expression for the first Fourier component of the modulated space-charge field Eq.(8.8).

In another experiment we projected several images into the crystal. Figure 8.8 shows examples of original and converted images. As mentioned in the introduction an inversion of the contrast takes place by this conversion. The original size of the projected images in the crystal were about 3mm in diameter and therefore the resolution obtained in these experiments was 7 line pairs/mm. The coherent image build-up was of the order of 2ms for a constant intensity $I_0$ and a switched-on intensity $I_{\text{inc}}$ of 200mW/cm².

The resolution could be increased further by optimizing the diffraction geometry and the incoherent projection. The incoherent projection can be optimized by choosing a projection lens with a long focal length and on the other hand by working with crystals with a minimal thickness. In this way resolutions of up to 50 line pairs/mm should be obtainable.

Such fast spatial light modulation leads to the possibility to incorporate such devices into optical signal processors, e.g. in matched filters, correlators and convolutors. Finally the non-linear intensity transfer function shown in Fig.8.7 can be used for special filter design.
Fig. 8.7 a) Intensity of the self-diffracted beam for several coherent writing intensities $I_0$ as a function of the intensity of a homogeneous illumination $I_{inc}$ of the crystal with an incoherent source (halogen lamp).

b) Efficiency of self-diffraction as a function of the relative incoherent intensity $I_{inc}/I_0$ calculated from the data of Fig. 8.7a.
Fig. 8.8 Examples of (a) original and (b) converted images. The original size of the projected patterns was 3mm in diameter.
9. Conclusion

The investigations described in this thesis can be divided in three parts. The first one deals with the characterization of the photorefractive properties of doped and pure KNbO$_3$ crystals. A second one is the discussion of the physical processes which lead to polarization transformation upon diffraction in birefringent crystals, so called anisotropic Bragg diffraction. In the last part the design and characterization of optical devices applying this special type of diffraction are presented.

KNbO$_3$ single crystals exhibit excellent properties for real-time holography. The possibility to "tailor" crystals for photorefractive applications makes them very attractive. Doping the crystals with transition metal ions is only one way to improve the photorefractive sensitivity. Another way is the chemical reduction of doped and pure KNbO$_3$ samples. The electrochemical method gives best results up to now. Grating build-up times near the theoretical limit were observed in strongly reduced crystals. The problem of preparing crystals in a homogeneous reduction state is not completely solved up to now. This type of reduction requires very homogeneous starting material and very stable conditions.

Such material treatment, however, requires a fast and non-destructive characterization of the photorefractive properties. Such methods, applied in this work, are found to be photoconductivity and absorption measurement and beam coupling experiments. From the results obtained with these methods the temporal response of a KNbO$_3$ crystal on coherent illumination can be predicted.

For the investigation of the microscopic mechanism of the photorefractive effect and of the relevant defect centers preliminary EPR experiments in as grown and reduced iron doped crystals have been performed. It was found that the electrochemical reduction method leads to a controlled reduction of Fe$^{3+}$. For a full understanding of the microscopic mechanisms of the photorefractive effect in KNbO$_3$, however, much more work has to be done.

Another main topic of this thesis is the analysis of light diffraction in anisotropic materials. It is shown that the Bragg condition requires a careful match between grating, incident and diffracted wave vectors. The angles of
incidence and diffraction are generally different because of the different refractive indices for the incident and diffracted waves with polarization directions normal to each other.

By using a noncritical configuration for anisotropic Bragg diffraction by photoinduced gratings in KNbO₃ crystals we were able to build a light deflector, the deflection angle of which can be altered within a range of 5.67° by changing the wavelength of the laser beams used for the recording of the grating by 57.7nm. It is shown that the configuration of this deflector is much simpler as compared to deflectors using isotropic Bragg diffraction in photorefractive materials, in which additional elements such as beam-correction gratings or other beam-tilting techniques are required to assure the critical Bragg condition. Because of the fact that the incidence and diffraction angle depend on the refractive indices these angles show a certain temperature dependence. In applications of this effect, therefore, a temperature stabilization is necessary.

In a second device application this type of light deflection is used for the design of an optically addressable fiber switch. In the configuration described above, a variably deflected laser beam can be coupled by a focussing lens behind the crystal into any selected fiber of a linear array. A prototype of a linear array containing 128 multimode fibers was used for a 1x128 fiber switch. In this device the information beam entering through a fiber can be deflected to any position of the 128 exit channels (fibers) by a variation of the control wavelength (produced by a Rhodamin 6G dye laser). Using reduced KNbO₃ single crystals as host of the variable grating, switching times of typically 1 ms and efficiencies of 1% have been reached (ratio of the intensities in the output and input fiber). The crosstalk to an adjacent fiber was better than -21dB.

In a further application of anisotropic Bragg diffraction a special writing geometry is used. In these experiments the writing beams are diffracted by the self-induced grating. In contrast to the case of isotropic diffraction, the deflected beam differs both in direction and polarization with respect to the writing beams. This so called anisotropic self-diffraction is used for designing an optically addressable spatial light modulator or incoherent-to-coherent converter. In this application, in addition to the two writing beams a two dimensional image (information) is projected into the crystal. Thereby the photoinduced grating amplitude is changed in such a manner that in the bright
zones of the projection the diffraction efficiency is decreased because of a local increase of the photoconductivity. In such a way the image can be transferred with inverted contrast from the incoherent beam onto the self-diffracted beam. A simple theoretical model describing the intensity transfer characteristics is proposed for this process, and the factors limiting the resolution are examined. Experiments with reduced KNbO₃ single crystals led to resolutions of up to 10 line pairs per millimeter and conversion times of the order of 1ms. Besides this fast response time two advantages make this type of photorefractive incoherent-to-coherent converter attractive:

a) The photoinduced grating is read-out by one of the writing beams, thus no additional read-out beam is needed, which also could erase the grating.

b) Due to the fact that one of the strong writing beams is diffracted by the self-induced grating, the diffracted beam may have high intensity even for low diffraction efficiencies.
### List of Symbols

#### Constants
- **c**: Velocity of light in vacuum
  - Value: \(2.99792458 \times 10^8 \) m/s
- **e**: Elementary charge
  - Value: \(1.6021892 \times 10^{-19} \) C
- **h**: Planck's constant
  - Value: \(6.6021892 \times 10^{-34} \) Js
- **k_B**: Boltzmann constant
  - Value: \(1.380662 \times 10^{-23} \) JK\(^{-1}\)
- **\(\varepsilon_0\)**: Permittivity of the vacuum
  - Value: \(8.85418 \times 10^{-12} \) As/V\(^{-1}\)m\(^{-1}\)
- **\(\mu_0\)**: Permeability of the vacuum
  - Value: \(4\ \pi \times 10^{-7} \) Vs/Am\(^{-1}\)

#### Mathematical Symbols
- **i**: Imaginary unit
  - Value: \(\sqrt{-1}\)
- **Im( )**: Imaginary part
- **Re( )**: Real part
- **\(\Delta\)**: Laplace operator
  - Value: m\(^{-2}\)
- **\(\pi\)**: Pi
  - Value: 3.1415926

#### Variables with Roman Letters
- **d_{ijk}**: Nonlinear optic tensor element
  - Units: mV\(^{-1}\)
- **d**: Crystal thickness
  - Units: m
- **D**: Diffusion constant (\(D=\mu k_B T/e\))
  - Units: m\(^2\)s\(^{-1}\)
- **E**: Electric field
  - Units: Vm\(^{-1}\)
- **E_L**: Electric field of light
  - Units: Vm\(^{-1}\)
- **E_d**: Diffusion field
  - Units: Vm\(^{-1}\)
- **E_{ph}**: Photovoltaic field
  - Units: Vm\(^{-1}\)
- **E_{q}**: Limiting space-charge field
  - Units: Vm\(^{-1}\)
- **E_{sc}**: Space-charge field
  - Units: Vm\(^{-1}\)
- **g**: Generation rate of free carriers
  - Units: m\(^{-3}\)s\(^{-1}\)
- **g_{av}**: Mean generation rate of free carriers
  - Units: m\(^{-3}\)s\(^{-1}\)
- **l**: Light intensity
  - Units: Wm\(^{-2}\)
- **j**: Electric current density
  - Units: Am\(^{-2}\)
- **k**: Wave number (\(k=2\pi/\lambda\))
  - Units: m\(^{-1}\)
- **k_r**: Wave number of the read-out beam
  - Units: m\(^{-1}\)
- **K_g**: Wave number of the photoinduced grating
  - Units: m\(^{-1}\)
- **t_o**: Photovoltaic drift length
  - Units: m
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**Greek Letters**

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References

Chapter 1


Chapter 2

Chapter 3

[3.16] J. Hulliger, private communication
[3.18] H. Arend, private communication

Chapter 4


Chapter 5


Chapter 6


Chapter 7

Chapter 8


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