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High-Sensitivity Read-Write Volume Holographic Storage in Reduced $KNbO_3$ Crystals

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Abstract. Reduced $KNbO_3$ is a photoconductive ferroelectric in which holograms can be recorded by the photorefractive effect. Read-write volume hologram storage and erase sensitivities of $S^{-1}=100\,J/cm^2$ and $S^{-1}=84\,J/cm^2$ ($S=d(\Delta n)/d(I_0 t)|_{t=0}$) have been measured at zero applied electric field, where the charge transport is shown to be due to diffusion of photoexcited electrons. By applying an electric field along the *c*-axis, the migration length of the photoexcited electrons becomes comparable to the holographic grating spacing. This leads to storage sensitivities comparable to high-resolution photographic plates. Experimental data on storage and erase sensitivity as a function of the grating spacing, applied electric field, writing light intensity and temperature are reported and interpreted on the basis of the theoretical results of Young et al. and Amodei. Changes of the intensity ratio of the writing beams by self diffraction (beam coupling), reflections from surfaces and the residual dark conductivity are assumed to cause experimental results which deviate from the theoretical models. It is shown, that in reduced $KNbO_3$ and other ferroelectric photoconductors having photocarrier transport lengths much larger than the unit cell dimension, photovoltaic currents do not contribute significantly to the build-up of space-charges leading to the photorefractive effect.

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Recording of volume phase-holograms in electro-optic crystals can be achieved by light-induced refractive index changes (photorefractive effects). Upon exposure of the crystals to light interference-patterns, electrons or holes are excited and transferred to different sites. The resulting space-charge fields modulate the refeactive indices via electrooptic effect. Uniform illumination erases the space-charge fields and brings the crystal back to its original state.

Ferroelectric materials as $LiNbO_3$ [1], $LiTaO_3$ [2], $BaTiO_3$ [3], SBN [4], and $KNbO_3$ [5–7] as well as photoconductive nonferroelectrics as KTN [8] and $Bi_{12}SiO_{20}$ and $Bi_{12}GeO_{20}$ [9] have been used as storage materials. The characteristic parameters (photosensitivity, diffraction efficiency and storage time of the holograms) of the photorefractive effect in different materials have been shown to be mainly determined by the different charge transport processes (photovoltaic drift, photoconductivity and diffusion) of the photoexcited carriers.

The characteristic transport length $\mu\tau E$ and $(D\tau)^{1/2}$ (where μ is the mobility, τ is the lifetime before retrapping, and D is the diffusivity) have been shown to be short compared with the grating spacing in most of the ferroelectric materials. Therefore many cycles of photoexcitation, charge transport and trapping are necessary until the charges are finally trapped in dark areas of the hologram. An increase in the operative transport length (up to a certain limit comparable to the fringe spacing of the hologram), which is the case in highly photoconducting materials as KTN [8], $Bi_{12}SiO_{20}$, $Bi_{12}GeO_{20}$ [9], SBN + Ce [10], and as will be shown in this paper also in reduced $KNbO_3$, lead to an increased sensitivity in hologram writing.

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In this paper we report measurements of the storage and erase sensitivity in reduced $KNbO_3$. It is shown, how these sensitivities depend on the applied electric field and fringe spacing of recorded holograms.

1. Theoretical

The sensitivity S of phase hologram formation can be defined as the refractive index change Δn per unit of incident optical energy $I_0 \cdot t$

$$S = \left.\frac{d\Delta n}{d(I_0 t)}\right|_{t=0}. \tag{1}$$

In general the storage sensitivity depends on the light absorption, the charge-transport mechanism, the electro-optic effect of the storage material, as well as on beam coupling effects caused by the interaction of writing beams in the storage medium [11–13]. It has been shown, that in several materials the sensitivity can be increased by suitable transition metal doping (e.g. Fe, Cu, ...) and chemical annealing (reduction treatment) [1, 2, 12].

In the case of $KNbO_3$ Fe doping and reduction treatment has been shown [5] to yield a storage sensitivity which is about an order of magnitude larger than in Fe doped $LiNbO_3$ [1]. The results of photo-conductivity [5, 14] and beam coupling experiments [15] in reduced $KNbO_3$ demonstrate, that photovoltaic and photo-polarization mechanisms [1, 16] are neglectable for describing the hologram formation in these crystals, and that the carrier migration length for moderate external electric field strengths are comparable with visible light grating spacings. It is clear, that the latter causes a large increase in the storage sensitivity similar as in $Bi_{12}SiO_{20}$, a nonferroelectric electrooptic photoconductor [9].

A general expression for the diffraction efficiency η for large migration lengths and including beam coupling (i.e. interference of incident light beams with its own diffracted beam inside the recording material) has been derived by Kukhtarev and Vinetsky [17]:

$$\eta = \frac{2m \exp\frac{\Gamma \cdot l}{2}[ch(\Gamma l/2) - \cos(\delta A l)]}{(1+m)[1+m\exp(\Gamma \cdot l)]}, \tag{2}$$

where

$$\delta = \pi \cdot n_3^3 \cdot r_{33}/\lambda_0 \cos(\theta_0/2),$$

n_3 is the refractive index, r_{33} is the effective electrooptic coefficient, λ_0 and θ_0 are the wavelength and the angle between reading beams, m is the modulation ratio of recording beams, l is the crystal thickness, A is the amplitude component of the space-charge field forming the holographic grating, which is in phase with the interference pattern and Γ the gain characterizing the energy transfer of writing beams (see Ref. [17] for the exact definition of Γ).

It has been shown, that in reduced $KNbO_3$ the steady-state holographic grating is shifted by about a quarter of the fringe spacing with respect to the interference pattern [15]. In this case and for small gain Γ ($\Gamma l \to 0$) and small diffraction efficiency ($\eta \ll 1$), this general expression reduces to the well known formula derived by Kogelnik [18]

$$\eta = \left(\frac{\pi l \Delta n}{\lambda_0 \cos\theta_0/2}\right)^2. \tag{3}$$

It must be noted however, that the approximation (3) becomes invalid for large electric fields ($E_0 > 2\,\mathrm{kV/cm}$) and for small fringe spacings ($\Lambda < 2\,\mu\mathrm{m}$), since Γ increases from $0.2\,\mathrm{cm}^{-1}$ for $\Lambda = 10\,\mu\mathrm{m}$ to $\approx 2\,\mathrm{cm}^{-1}$ for $\Lambda = 1\,\mu\mathrm{m}$ [15] (in these experiments $l = 3.3\,\mathrm{mm}$).

From (1) and (3) we get for the sensitivity

$$S = \frac{\lambda_0 \cos\theta_0/2}{\pi \cdot l} \cdot \left.\frac{d\eta^{1/2}}{d(I_0 t)}\right|_{t=0}. \tag{4}$$

2. Experimental

In these experiments nominally pure single domain crystals of $KNbO_3$ with a Fe concentration of $(46 \pm 11)\,\mathrm{ppm}$ [7] were used. Reduction of the Fe^{3+} has been performed electrochemically by annealing the samples in silicon oil at 200 °C during 100 h. The optical absorption spectra of the reduced crystals show the characteristic maximum near 2.55 eV [5]. The absorption coefficient for $\lambda = 488\,\mathrm{nm}$ was $\alpha = 3.8\,\mathrm{cm}^{-1}$. Rectangular shaped crystals with dimensions $a \times b \times c = l = 6 \times 3 \times 3.3\,\mathrm{mm}^3$ were coated with Ag electrodes on the $\pm c$ faces. Recording of holograms was in an area of approximately $4 \times 3\,\mathrm{mm}^2$, voltages of up to 2 kV ($E = 7\,\mathrm{kV/cm}$) were applied at the electrodes.

Holograms were recorded by interfering two Ar laser beams from a Lexel Laser Model 95 ($P_0 = 1\,\mathrm{W}$ for $\lambda = 488\,\mathrm{nm}$), which were polarized in the plane of incidence. The polar c axis of the sample was perpendicular to the bisector of the incident beams. By using a specially designed beam splitter prism [15] the angle of intersection could be changed, thus allowing to vary the fringe spacing Λ between 1 μm and 10 μm. One of the recording beams (object beam) was periodically blocked by means of a mechanical chopper. The pulse duration of the object beam was 5 ms.

The diffraction efficiency η of the recorded hologram was monitored by a 1 mW He–Ne laser having the same polarization plane as the Ar laser. The intensity of the diffracted light was measured relative to the

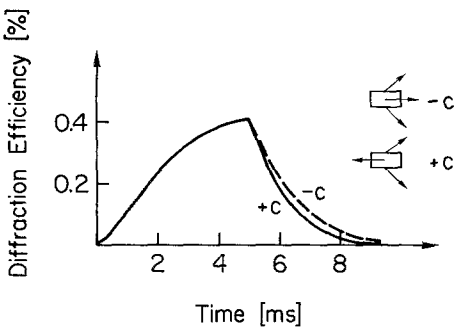

Fig. 1. Hologram writing and erasure for two directions of the crystallographic c-axis. (Polarity of the c-axis is relative to the reference beam) ($I_0 = 1\,\mathrm{W/cm^2}$, $\Lambda = 4.5\,\mu\mathrm{m}$, $E_0 = 0$)

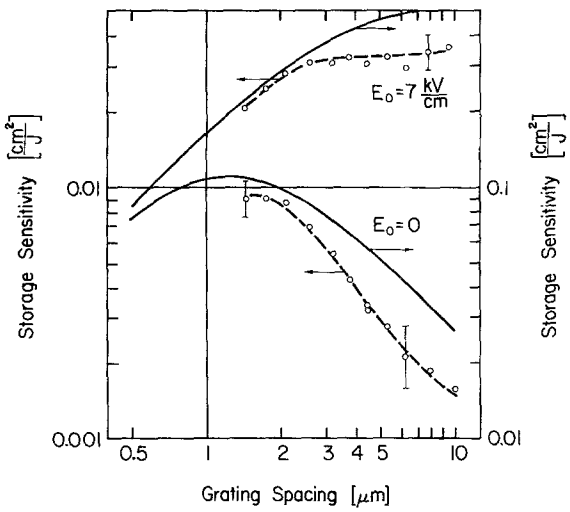

Fig. 2. Storage sensitivity vs. grating spacing for different electric fields ($I_0 = 1\,\mathrm{W/cm^2}$). Solid lines: theory; dashed lines: experiment

transmitted intensity before recording with a photomultiplier and an oscilliscope.

3. Results

The time dependence of the diffraction efficiency with periodically blocked object beam ($\tau \approx 5$ ms) is shown in Fig. 1. Recording sensitivity $S_R = 0.0033\,\mathrm{cm^2/J}$ and erasure sensitivity $S_E = 0.0042\,\mathrm{cm^2/J}$ are larger than in strongly reduced low Fe concentration $LiNbO_3$:Fe, where $S \simeq 0.00028\,\mathrm{cm^2/J}$ [1, 19].

Write and erase sensitivities are slightly different for different polarities of the c-axis with respect to the reference beam. However, this effect is smaller than in $LiNbO_3$ [13, 19].

In the following, we shall report the dependences of the hologram recording and erasure sensitivity as a function of several experimental parameters. These results should be useful for finding the experimental

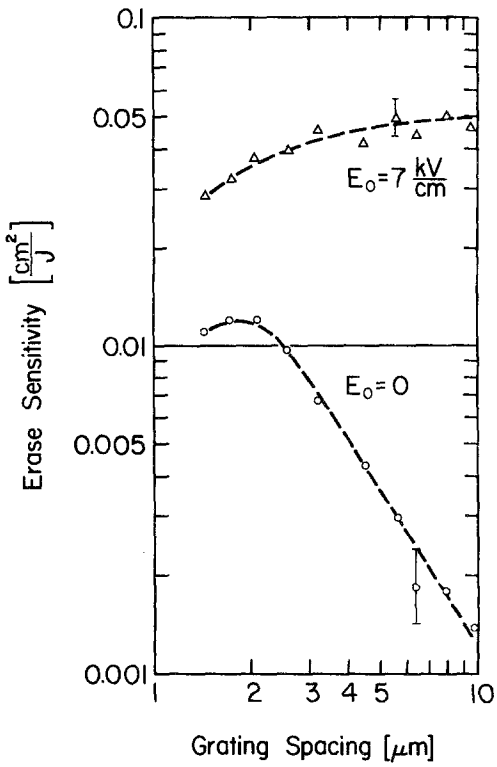

Fig. 3. Erasure sensitivity vs. grating spacing for different electric fields (experimental data) ($I_0 = 1\,\mathrm{W/cm^2}$)

conditions for high sensitivity read-write-erase applications.

3.1. Fringe Spacing Dependence

In Fig. 2 it is shown, that the storage sensitivity at zero applied electric field decreases approximately inversely proportional to the grating spacing Λ. For $\Lambda = 1.8\,\mu\mathrm{m}$ a maximum storage and erase sensitivity of $S_R^{-1} = 100\,\mathrm{J/cm^2}$ and $S_E^{-1} = 84\,\mathrm{J/cm^2}$ have been measured. By applying an electric field, the migration length will be increased, resulting in an increase of the sensitivity. The sensitivity for $E_0 \neq 0$ is seen to increase also with increasing fringe spacing Λ. This is in contrast to $LiNbO_3$:Fe where the storage sensitivity does not depend on the grating spacing [13], and only the erase sensitivity becomes a maximum for $\Lambda \approx 1\,\mu\mathrm{m}$ [20]. Both erase and storage sensitivities show analogous fringe spacing dependences (Figs. 2, 3).

3.2. Electric Field Dependence

The storage sensitivity in reduced $KNbO_3$ can be increased by applying an electric field E_0 to this photoconductive ferroelectric material. Since the photoconductivity is substantially larger than in other materials [1, 19, 21], this increase is much more pro-

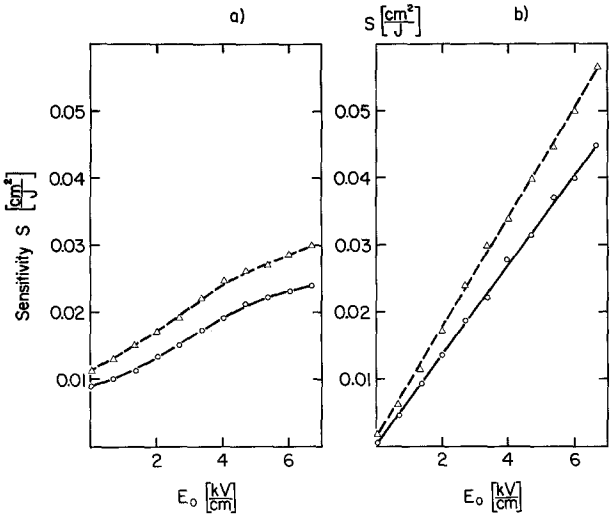

Fig. 4a and b. Storage (solid lines) and erasure (dashed lines) sensitivity vs. external electric field for $\Lambda = 1.5\,\mu m$ (a) and $\Lambda = 10\,\mu m$ (b) ($I_0 = 1\,W/cm^2$)

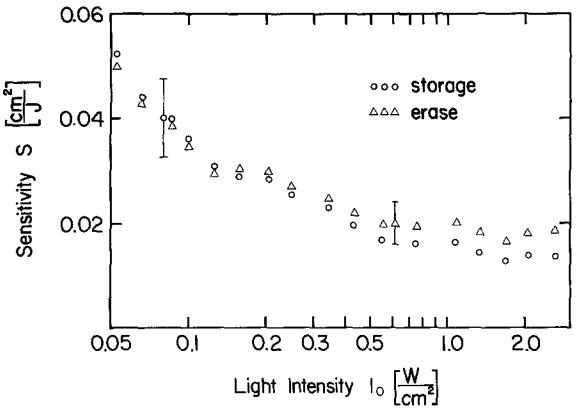

Fig. 5. Storage and erasure sensitivity vs. light intensity for $\Lambda = 2\,\mu m$ and $E_0 = 0$

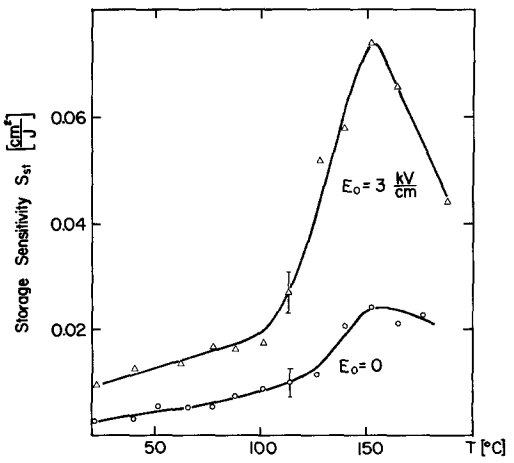

Fig. 6. Temperature dependence of storage sensitivity for different external fields ($I_0 = 1\,W/cm^2$, $\Lambda = 4\,\mu m$)

nounced than e.g. in $LiNbO_3$, and similar to the increase in nonferroelectric $Bi_{12}SiO_{20}$ and $Bi_{12}GeO_{20}$.

The increase of S is more pronounced for large grating spacings Λ, where $S_R \sim E_0$. For large Λ the storage sensitivity S_R depends almost linearly on E_0. The erase sensitivity is somewhat larger and shows an analogous electric field dependence as the storage sensitivity (Fig. 4).

3.3. Light Intensity Dependence

Increasing the writing light intensity I leads to a decrease of both the storage and erase sensitivity, and to a larger difference between them (Fig. 5). This has been observed for $E_0 = 0$ and $E_0 \neq 0$. Reciprocity between I and t observed in $LiNbO_3$:Fe leads to an intensity independent storage sensitivity. Reciprocity failure in materials where photoconductivity is the main charge transport process has already been suggested in [22].

3.4. Temperature Dependence

In the orthorhombic phase of $KNbO_3$, existing between $T_I = -52\,°C$ (transformation to the rhombohedral phase) and $T_{II} = 219\,°C$ (transformation to the tetragonal phase) [23], the storage sensitivity is shown to increase with increasing temperature (Fig. 6). At $T = 160\,°C$ the storage sensitivity exhibits a peak. This dependence, observed also for the diffraction efficiency [15], can be explained by the temperature dependences of the transport parameters (photo- and dark conductivity).

4. Discussion

Hologram formation by the photorefractive effect in reduced $KNbO_3$ is characterized by (i) carrier migration lengths comparable with the grating spacing, (ii) a dark conductivity which is large enough to be taken into account, (iii) the presence of beam coupling effects [7, 14, 15]. All these factors have to be considered in a theoretical description of our experimental results. Unfortunately no general theory describing the storage sensitivity for conditions (i), (ii), and (iii) exists. The theory of Young et al. [11] shows that an increased operative transport length leads to increasing hologram writing sensitivities up to a certain limit [12]. However, beam coupling effects and non-vanishing dark conductivities are not included in this theory. The dynamic theory of Ninomiya [24], Vahey [25], Magnusson et al. [26], and Moharam et al. [27] are valid only for "short" transport lengths. The dynamic theory of Kukhtarev and Vinetskii [17] applies for

large transport lengths, however, it describes only the steady-state diffraction efficiency. The influence of dark conductivity on the hologram formation and storage sensitivity has been discussed in [7, 5, 27, 28].

In the following we assume that in a first approximation the influence of beam coupling effects on the storage sensitivity can be neglected. This approximation is reasonable for most of our experimental parameters. We have shown in [15] that beam coupling effects in reduced $KNbO_3$ lead to only about ten percent changes of the intensities of writing beams. These changes lead to a small decrease of the modulation ratio m of the interference pattern and to a small decrease of the sensitivity ($S \sim m$). This effect is particularly small for two beams having a modulation ratio $m \simeq 1$ to be recorded.

In our experiment most of the measurements were performed with light intensities $I_0 \approx 1\,\mathrm{W/cm^2}$, for which $\sigma_p \gg \sigma_d$, where σ_p and σ_d are the photo and dark conductivities respectively[1]. Therefore, in most of our experiments, the influence of the residual dark conductivity has been neglected. However for recording with small light intensities and for crystal temperatures above room temperature we had also to consider the dark conductivity.

4.1. Storage Sensitivity

The storage sensitivity for neglectable dark conductivity and beam coupling effects can be derived from the results of Young et al. [11] and Moharam et al. [36]. For photorefraction caused by diffusion currents one gets

$$S_R^D = b \frac{\frac{2\pi}{\Lambda} \cdot L'^2}{\left[1 + \left(\frac{2\pi}{\Lambda} L\right)^2\right]}, \tag{5}$$

and for the drift case

$$S_R^d = b \frac{L}{\left[1 + \left(\frac{2\pi L}{\Lambda}\right)^2\right]^{1/2}}, \tag{6}$$

where

$$L = \left(\frac{kT}{e} \mu\tau\right)^{1/2} \tag{7}$$

is the diffusion length,

$$L = \mu\tau E_0 \tag{8}$$

[1] $\sigma_p/\sigma_d = 1150$ for $I_0 = 0.05\,\mathrm{W/cm^2}$ at room temperature. Under our laboratory conditions residual light lead to a somewhat smaller ratio of σ_p/σ_d.

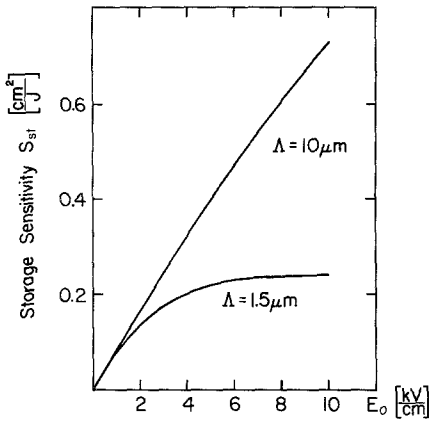

Fig. 7. Electric field dependence of the storage sensitivity for two grating spacings (theory)

the drift length,

$$b = \frac{n_3^3 r_{33} e \Phi \alpha m}{2\varepsilon_0 \varepsilon_{33} h\nu}, \tag{9}$$

μ and τ the mobility and lifetime of excited charge carriers, ε_{33} and ε_0 are the dielectric constants of the sample and the free space, n_3 the refractive index, r_{33} a electro-optic coefficient, Φ the quantum efficiency, e the electronic charge and $h\nu$ the photon energy.

The electric field and grating spacing dependence of the storage sensitivity has been calculated by using (5) and (6) and with the parameters for reduced $KNbO_3$: $\mu\tau = 10^{-8}\,\mathrm{cm^2/V}$ [14], $n_3^3 = 10.65$ [29], $r_{33} = 64\,\mathrm{pm/V}$ [30], $\varepsilon_{33} = 50$ [31], $\alpha = 3.8\,\mathrm{cm^{-1}}$ [5], $h\nu = 2.53\,\mathrm{eV}$; $\Phi = 1$. The diffusion length is $L = 0.2\,\mu\mathrm{m}$ [Eq. (7)], the drift length for an applied field $E_0 = 7\,\mathrm{kV/cm}$ is $L = 0.7\,\mu\mathrm{m}$ and the parameter $b = 1.2 \cdot 10^4\,\mathrm{cm/J}$. It is shown in Figs. 2, 4, and 7, that the theoretical curves in Figs. 2 and 7 qualitatively well describe the experimental data in Figs. 2 and 4.

For zero applied electric fields the photorefraction and the storage sensitivity are caused by diffusion of photoexcited carriers. For grating spacings Λ much larger than the diffusion length L the storage sensitivity decreases inversely proportional to the grating spacing (Fig. 2). For grating spacings comparable or smaller than the diffusion length the storage sensitivity according to (5) should be proportional to Λ. However, no experimental results were obtained in this range of small fringe spacings. A maximum storage sensitivity $S_R^D = b \cdot L/2 = 0.12\,\mathrm{cm^2/J}$ can be determined from (5) (Table 1).

It is seen from Fig. 4, that the photovoltaic effect does not contribute to the storage sensitivity, since $S_R(E_0)$ is symmetric with respect to $E_0 = 0$. Estimating the storage sensitivity caused by the photovoltaic effect [5]

Table 1. Storage properties for reduced $KNbO_3$ and other electro-optic materials

Material	α [cm^{-1}]	$\Phi\mu\tau$ [cm^2/V]	$S^D_{R\max}$ [cm^2/J]	$S^d_{R\max}$ [cm^2/J]	S^{pv}_R [cm^2/J]	S^d_E [cm^2/J]	Comment
$KNbO_3$-reduced calculated	3.8	$1\cdot10^{-8}$	0.10	1.85	$8\cdot10^{-5}$	0.09	$S^D_{R\max}$ is determined at $\Lambda=2\pi L$ and $E_0=0$
$KNbO_3$-reduced experiment	3.8	$1\cdot10^{-8}$	0.01	>0.05	–	0.06	$S^d_{R\max}$ and S^d_E are determined at $\Lambda=10\mu m$ and $E_0=7$ kV/cm
$KNbO_3$ + 300 ppm Fe calculated	3.1	$6\cdot10^{-12}$			$4.4\cdot10^{-5}$		[7]
$KNbO_3$ + 650 ppm Fe calculated	7	$5\cdot10^{-12}$			$12.6\cdot10^{-5}$		[7]
$LiNbO_3$ + 0.2% Fe, red.	38	10^{-13}			$53\cdot10^{-5}$		[16]
$Sr_{0.6}Ba_{0.4}Nb_2O_6$ + 0.1% Ce	10	$1\cdot10^{-8}$	0.095			0.12	[10]; $\Lambda=0.76\mu m$
$Bi_{12}SiO_{20}$	2.3	$1\cdot10^{-7}$		0.17		0.17	[9][a]
$Bi_{12}GeO_{20}$	2.1	$8.4\cdot10^{-8}$		0.04		0.04	[9][a]

[a] $\Lambda=5\mu m$, $E=6$ kV/cm

shows, that

$$S^{pv}_R=\kappa_0\frac{\alpha n_3^3 r_{33} m}{2\varepsilon_{33}\varepsilon_0} \tag{10}$$

is more than an order of magnitude smaller than our experimental values ($\kappa_0=3\cdot10^{-9}$ A cm/W [14] is the photovoltaic constant). The small influence of photovoltaic currents in comparison to photocurrents can be explained by the rather small effective photovoltaic migration length of about 0.76 Å compared to a diffusion length of $L=0.2\,\mu m$ and the drift length $L=0.7\,\mu m$ for $E_0=7$ kV/cm.

The increase of storage sensitivity with an applied electric field is shown in Figs. 2 and 4. It is seen, that the sensitivity reaches saturation for $\Lambda>2\,\mu m$ and $E_0=7$ kV/cm, since for these parameters $2\pi L/\Lambda\to0$ in (6) (drift length smaller than grating spacing). For small grating spacings and large electric fields (6) simplifies to

$$S^d_R\simeq b\frac{\Lambda}{2\pi}. \tag{11}$$

A maximum storage sensitivity for the drift case $S^d_R=1.8\,cm^2/J$ for $\Lambda=10\,\mu m$ has been calculated from (11). This maximum can be reached for large electric fields, where a complete separation of positive and negative photoexcited charges by the grating spacing Λ occurs.

For drift lengths L much smaller than the grating spacing ($L\ll\Lambda$, i.e., E_0 small and Λ large),

$$S^d_R\simeq KE_0, \tag{12}$$

where $K=b\mu\tau$. The storage sensitivity depends linearly on the applied field and not on the grating spacing, a situation which is analogous in $LiNbO_3$:Fe [12].

Table 2. Parameter $b\mu\tau$ for photoconductive recording in electro-optic crystals

Material	$b\mu\tau\left[\frac{cm^2}{J\cdot V}\right]$		Comment
	Calculated	Experimental	
$KNbO_3$ reduced	$11.6\cdot10^{-5}$	$0.6\cdot10^{-5}$	$\lambda=0.488\mu m$ this work
$LiNbO_3$:0.014 Fe			$\lambda=0.3507\mu m$
oxidized		$0.4\cdot10^{-8}$	
reduced		$0.2\cdot10^{-8}$	[21]
$Bi_{12}SiO_{20}$	$7.6\cdot10^{-5}$		[9]
$Bi_{12}GeO_{20}$	$4.6\cdot10^{-5}$		[9]

Calculated and experimental values of K are in good agreement and by three orders of magnitude larger than in $LiNbO_3$ (Table 2), since $\mu\tau$ is larger in reduced $KNbO_3$.

The fact, that the experimental and theoretical values for the storage sensitivity differ by an order of magnitude can be explained by a decrease of the modulation ratio by reflected and scattered light from illuminated electrode faces of the crystal or by beam coupling effects which were neglected in the theoretical relations used above.

4.2. *Erasure Sensitivity*

The erasure sensitivity depends on the following effects:

(i) Diffusion of photoexcited electrons and/or holes.

(ii) Drift of photoexcited electrons or holes in a space-charge field [32],

(iii) beam coupling effects due to the interaction of the readout beam and the diffracted beam [13, 19].

For optical erasure caused by diffusion processes, the decay time is given by [20]

$$\tau_0 = \frac{e}{4\pi^2} kT\mu\Lambda^2 . \tag{13}$$

For $\mu = 0.5\,\mathrm{cm^2/V \cdot s}$ [34] and $\Lambda = 1\,\mu\mathrm{m}$ we get $\tau_D \approx 0.7\,\mu\mathrm{s}$, three orders of magnitude less than the time constants observed in our experiment (Fig. 1). Therefore this effect on the erasure sensitivity will be neglected.

Beam coupling effects, i.e. interference between the readout beam and the diffracted beam show a small influence on the erasure time constants (Fig. 1). A somewhat different hologram will be recorded by this interference pattern. Reversing the direction of the c-axis with respect to the readout beam in- or decreases the net efficiency during readout in $LiNbO_3$:Fe [35]. In reduced $KNbO_3$ only a small variation in the erasure time upon reversal of the crystal orientation has been observed (Fig. 1). No changes in the diffraction efficiency can be observed in reduced $KNbO_3$, since the short erasure time prevents the formation of the new hologram. Therefore beam coupling effects will be neglected in the following.

The most important mechanism for hologram erasure is due to the drift of electrons driven by the space-charge field [32]. The field pattern is erased by the uniform read-out light, which photoexcites electrons out of the traps. The decay time of the space-charge field E_{sc} can be derived from the continuity equation [32] leading to

$$E_{sc} = E'_{sc} \exp\left(-\frac{t}{\tau}\right) \tag{14}$$

with

$$\tau = \frac{\varepsilon_{33}\varepsilon_0}{\sigma_p} . \tag{15}$$

For the initial stage of hologram formation (14) can be approximated by

$$E_{sc} \simeq E'_{sc}(1 - t/\tau) . \tag{16}$$

From (4) with (16) and the expressions for σ_p and Δn [7] *we get*

$$S_E^d = b\frac{\mu\tau}{m} E'_{sc} . \tag{17}$$

For constant light intensities and temperature the erasure sensitivity primarily depends on the space-charge field or the steady-state diffraction efficiency ($\eta \approx E'^2_{sc}$ [15]). The experimental curves $S_E(\Lambda)$ (Fig. 3) and $S_E(E_0)$ (Fig. 4) are in qualitative agreement with the theoretical ones shown in [Ref. 15, Fig. 3] and [Ref. 15, Fig. 5]. Therefore (17) gives a good qualitative description of the erasure sensitivity in reduced $KNbO_3$.

A simple expression for the ratio of storage and erasure sensitivity can be derived from (13) and (17) for $L \ll \Lambda$:

$$\frac{S_R^d}{S_E^d} = m\frac{E_0}{E'_{sc}} . \tag{18}$$

Figures 2–5 show, that the storage sensitivity S_R is always larger than the erasure sensitivity S_E. This is due to the fact, that $E'_{sc} \approx 0.1\,E_0$ [15] in (18). However, light reflection in the crystal can also lead to a reduction of the modulation ratio m, which can increase S_E^d so that $S_E^d \approx S_R$.

Another reason why the storage sensitivity S_R^d is larger than the one for erasure, is the presence of beam coupling effects, which in general leads to a decrease of the storage sensitivity. The beam coupling, characterized by the gain of the amplified beam, is more pronounced for large light intensities [15], thus this effect can lead to a decrease of S_R^d/S_E^d with light intensity (Fig. 5).

4.3. Light Intensity Dependence

For the understanding of the light intensity dependence of the photorefractive sensitivities in reduced $KNbO_3$ mainly two effects have to be considered:

(i) the decrease of the lifetime of the charge carriers with light intensity I_0 [14] and

(ii) the increase of the photocurrents σ_p and of the ratio σ_p/σ_d with I_0 [14].

A decrease of the carrier life time τ with intensity I_0 and its influence on the sensitivity can be deduced from (5), (6), (17) (see also [14]). At room temperature an increase of the light intensity from 0.1 to 1 $\mathrm{W/cm^2}$ leads to a 2.4 times decrease of the lifetime τ [14]. We therefore get a 2.2 times decrease of the sensitivity for $L \ll \Lambda$ (Fig. 5). The increase of σ_p/σ_d [14] with light intensity can increase the sensitivity.

This situation is quite different than in $LiNbO_3$:Fe, where reciprocity between I_0 and t has been shown to hold ($S \neq S(I_0)$; [22, 33]).

The two effects discussed above are not significant in that material, because the lifetime in $LiNbO_3$:Fe does not depend on the light intensity [16], the conductivities in this material are much smaller than in reduced $KNbO_3$ and the (small) photoconductivity depends linearily on the light intensity.

4.4. Temperature Dependence

The temperature dependence of the storage sensitivity is determined by an increase of the photoconductivity and $\Phi\mu\tau$ [14] with temperature. It has been shown

earlier, that b does not change significantly with temperature, since the polarization-optic coefficient $f_{33}=r_{33}/\varepsilon_{33}\varepsilon_0$ [30], n_3[29], and α [14] are temperature independent in first approximation. The product $\Phi\mu\tau$, which is proportional to the photoconductivity, however increases by two orders of magnitude for a temperature increase from 20 to 160 °C [14]. For the same temperature rise the storage sensitivity increases only by a factor of seven. This large difference is not due to beam coupling effects, since this effect becomes even smaller at higher temperatures [15]. Therefore we think, that the decrease of σ_p/σ_d with temperature [14], which has been shown to have the same influence on the diffraction efficiency as a decrease of the modulation ratio m [7], is the reason for this discrepancy.

The maximum in the storage sensitivity $S_R(T)$ near 160 °C has not been completely understood, but probably has to do with the appearance of photodomains near the orthorhombic-tetragonal phase transition temperature or with the appearance of electron phasons near the phase transition temperature as suggested in [37].

5. Conclusions

It has been verified experimentally, that the theories of Young et al. [11] for the storage sensitivity and the theory of Amodei [32] for the erase sensitivity qualitatively well describe the grating spacing, electric field, light intensity and temperature dependence in reduced $KNbO_3$. The limitations of the above theories are due to beam coupling effects, nonvanishing dark conductivity not considered in these treatments.

The experimental values for the storage and erase sensitivity in reduced $KNbO_3$ are up to three orders of magnitude larger than in $LiNbO_3$:Fe where the photorefractive effect is due to the photovoltaic effect. The electric field dependence and the sensitivity are similar as in the nonferroelectric photoconductors $Bi_{12}SiO_{20}$ and $B_{12}GeO_{20}$ [9], the latter quantity is as large as in silver halide plates and thus represents the highest photorefractive sensitivity obtained in electro-optic crystals.

It has been shown, that in highly photoconductive materials with large carrier transport lengths the photovoltaic currents have neglectable influence on the photorefraction. Therefore, the large storage sensitivity in other photoconducting ferroelectrics (e.g. in Ce doped $Sr_{0.6}Ba_{0.4}Nb_2O_6$ [10]) is also due to diffusion currents and not due to the photovoltaic effect. The carrier transport length in reduced $KNbO_3$ has been shown to be comparable to the grating spacing of visible light holograms, because $\Phi\mu\tau=10^{-8}\,cm^2/V$ [7].

Another reason for the large photorefractive sensitivity in reduced $KNbO_3$ is the large quantum efficiency $\Phi\sim 1$. The value of $\Phi\mu\tau$, determined from photoconductivity measurements [14], assuming $\Phi=1$ gives a good qualitative agreement between theory and experimental curves $S_R(\Lambda)$ (Fig. 2).

The large carrier transport length in reduced $KNbO_3$ is probably due to a low density of empty traps. According to [19] this leads to an increase of the carrier lifetime and photoconductivity as well as to an increase of the erasure sensitivity and a decrease of the maximum diffraction efficiency. Reduced $KNbO_3$ is indeed characterized by a larger $\Phi\mu\tau$ product and a larger sensitivity than in oxidized Fe doped $KNbO_3$ [7]. The low density of empty traps also leads to an early saturation of $S(E_0)$ and $\eta(E_0)$. However saturation has not been observed for $E_0 \leqq 7\,kV/cm$. The nature of the photorefractive centers in $KNbO_3$ has not been understood completely. The maximum in the absorption constant near 2.55 eV in reduced $KNbO_3$ could be due to photoionization of Fe^{2+} centers [7]. The trapping centers could be shallow traps, since the decay time for the holograms is only a few milliseconds.

The large values of storage and erasure sensitivities, the possibility of effectively enhance the sensitivity by an applied electric field and the symmetrical write-erase behaviour make reduced $KNbO_3$ a promising materials for dynamic holography, amplification of time variing coherent light beams and images and for write-read-erase storage in real-time devices.

The active centers involved in the storage mechanism should be identified definitely for optimizing the properties relevant for the above mentioned applications. These results should stimulate further research concerning the defect structure, transport properties and its influence on the photorefractive effect in $KNbO_3$.

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