Photorefractive charge compensation at elevated temperatures and application to KNbO$_3$

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We treat the photorefractive effect in oxide crystals at elevated temperatures where charge compensation occurs in the absence of photoexcitation of the compensating species. These species can be either mobile ions or holes in the valence band. Two models are presented that take into account particularities of ion and hole transport. In the small-modulation approximation, solutions for the steady state and the dynamic evolution of the photorefractive effect are given. The maximum space-charge field $E_s$ that can be reached depends on the effective number of electron traps in the crystal. However, in the steady state, while the component of the space-charge field that is due to electrons and the one that is due to the compensating carriers both approach the value $E_s$, an almost complete compensation of these two components occurs. The speed of compensation is slower for larger grating spacings than for smaller grating spacings and can be increased by applying an electric field. Applying an external electric field also produces a phase shift between the two gratings, therefore increasing the total space-charge field. Experiments performed in KNbO$_3$ confirm the theoretical predictions and indicate that the ionic model is more appropriate for this crystal. Implications of these compensation effects for quasi-permanent hologram storage are discussed.

1. INTRODUCTION

Volume-phase holograms in photorefractive materials are produced by the redistribution of photoinduced charge carriers under inhomogeneous illumination. These carriers induce an internal space-charge field, which leads to the formation of a refractive index grating by means of the linear electro-optic effect. The stored holograms are normally erased if the readout laser beam is of the same wavelength as the one used for recording. To avoid erasure, several techniques for fixing holographic gratings have been developed in the past, such as thermal fixing, electrical fixing, and two-photon recording. From the point of view of the theoretical understanding, most of the attention has been paid to the thermal fixing procedure in LiNbO$_3$ crystals. In this kind of crystal the thermal fixing can be explained by the motion of secondary charge carriers, which have been recognized as protons, in the space-charge field created by the primary charge carriers. Charge-compensation effects, which can be used for quasi-permanent hologram storage, have also been observed in highly photoconductive electro-optic materials such as KNbO$_3$, Bi$_{12}$SiO$_{20}$, Bi$_{12}$TiO$_{20}$, and BaTiO$_3$.

In these materials the dynamic behavior during the recording of the quasi-permanent holograms is qualitatively different from the one observed in LiNbO$_3$, where the buildup of the primary space-charge field is driven by photovoltaic currents.

Until now, the unusual dynamics observed during charge compensation in these materials had not been fully understood. The aim of this paper is to provide a theoretical explanation for such anomalous time evolution of the space-charge field.

Before starting with the description of our theoretical models we want to describe the terms of the problem and give some useful definitions with the help of Fig. 1. It shows schematically the dynamic behavior of the space-charge field observed in KNbO$_3$ crystals during charge compensation at a temperature of approximately 100°C. This dynamics was reported in a previous paper and is similar to the one that has been observed in other crystals by various authors.

In the figure four distinct regions, A, B, C, and D, are indicated. In the first two regions, A and B, the sample is illuminated with two recording laser beams. We can thus consider these two parts together as the hologram recording process. The figure shows that the space-charge field $E_1$ reaches a maximum at point 1 and then decreases to a constant steady-state value at point 2. The value of $E_1$ at point 2 is very small. The usual tentative explanation for this behavior is the following. In region A, photoinduced charge carriers create a primary photorefractive grating that is due to the inhomogeneous light illumination. We call this process the writing of the hologram. In region B the primary grating is compensated by secondary charge carriers, which move by diffusion and drift in the field created by the primary charge carriers. In other words, the secondary charge carriers create a negative replica of the primary space-charge field. We call this process the erasure of the hologram. Of course, a sharp distinction between the two processes occurring in regions A and B is an idealization. In reality, the writing of the hologram continues after point 1, and the compensation begins before this point. Nevertheless, the above definitions are useful because the dynamics shows clearly distinct features in the two regions.

Regions C and D represent the hologram erasure process. In these two parts the crystal either is left in the dark or is illuminated in a homogeneous way. After the recording process is terminated the space-charge field increases, reaches a maximum at point 3, and then decreases to zero. This behavior is observed in KNbO$_3$ independently of whether the crystal is in the dark or is illuminated homogeneously. The process occurring in region C
We can look at the process occurring in region D in two gram. The process in region D is governed mainly by the carriers. We call this process the revealing of the holo-space-charge grating by diffusion of the primary charge carriers. The partial erasure of the primary charge carriers and the subsequent revelation of the secondary grating are the underlying processes (see Ref. 15). The value of $|E_1|$ at point 2 is not to scale.

can be assigned to the partial erasure of the primary space-charge grating by diffusion of the primary charge carriers. We call this process the revealing of the hologram. The process in region D is governed mainly by the secondary charge carriers, which slowly redistribute themselves until a homogeneous distribution is reached. We can look at the process occurring in region D in two different ways, depending on the experimental conditions. If the crystal is in the dark, the process is related to the storage time of the hologram. If the crystal is illuminated for recovering the original information, it is related to the handling time of the hologram. The reason for this distinction will become clear below.

The processes observed in regions A, B, C, and D are characterized by typical exponential time rates $\Gamma$ describing the time evolution. In particular, the experimental observations in KNbO$_3$ show that the time evolution during the compensating, the storage, or the handling time of the hologram are well described by a single exponential function. When the temperature is varied, the typical time constants observed during these processes can be made much longer than the writing or the revealing time. These properties permit the fixing of the phase gratings with the procedure described in Ref. 15.

In this paper we present two models that are able to explain the phenomena shown schematically in Fig. 1. The first model is analyzed in detail in Subsection 2.A. It considers optically inactive ionic charges that drift in the space-charge field created by the primary charge carriers that are being represented by electrons. The second model is presented in Subsection 2.B and considers optically excited holes instead of ions as secondary charge carriers. It will be shown that the second model describes basically the same physics as the first one. The minor differences are discussed in Section 4.

In our models the conventional Kukhtarev equations are modified to include the contributions of the moving secondary charge carriers and are solved in the small-modulation approximation. Special attention is paid to the prediction of the grating spacing dependence of the steady-state values of the involved electric-field gratings and all the time rates that describe the charge-compensation dynamics. Furthermore, the effects originating from an externally applied electric field are discussed in detail. Analytical relations obtained for some limiting cases are compared with exact solutions derived for a set of model parameters. This permits the discussion of the domain of validity of the approximations made. By comparing the theoretical results with experiments, one can determine many properties of the primary charge carriers, such as the effective number of electron traps, the dielectric relaxation rates for electrons and ions, and the diffusion length for electrons. It is also shown that the rate at which the quasi-permanent grating finally disappears can decrease with increasing illuminating light intensity. If this effect occurs in the experiments in a given crystal sample, information on the trap-limited electronic mobility or on the number of shallow defect levels in the crystal can be gained.

The experimental observations in KNbO$_3$ crystals presented in Section 3 are consistent with the new predictions of our theoretical models. Our models are also able to explain the experimental observations in Bi$_{12}$SiO$_{30}$, Bi$_{12}$TiO$_{30}$, and BaTiO$_3$.

This is discussed in Section 4. Finally, the implications resulting from the theoretical models for the optimization of the fixing of phase holograms in highly photoconductive materials are discussed in detail.

2. THEORETICAL MODELS

A. Electron–Ion Model

In our first model we consider optically excited electrons and a background ionic conduction. Free electrons can be photoexcited only from a single-donor level in the band gap into the conduction band. No contribution of optically excited holes is considered. The photoexcited electrons move by diffusion and drift and are then retrapped, generating a concentration grating. At the same time, ions (which are assumed to have a single positive charge) drift under the effect of the total internal electric field and diffuse in their concentration gradient. The set of equations first put forward by Kukhtarev et al. must be extended with the continuity equation for the ionic charges and the contribution of the ion distribution to the Poisson equation. We neglect the contribution of photovoltaic currents to the motion of electrons, and we consider the usual one-dimensional problem with the modulation along the x axis. The complete set of equations is then written as

$$\frac{\partial n}{\partial t} = (sI + \beta)(N_{D0} - N_{D^+}) - \gamma n N_{D^+} + \frac{1}{e} \frac{\partial J_i}{\partial x},$$

$$\frac{\partial N_{D^+}}{\partial t} = (sI + \beta)(N_{D0} - N_{D^+}) - \gamma n N_{D^+},$$

$$\frac{\partial N_i}{\partial t} = - \frac{1}{e} \frac{\partial J_i}{\partial x},$$

$$J_e = \epsilon \mu_e n E + \mu_e k_T \frac{\partial n}{\partial x},$$

$$J_i = \epsilon \mu_1 n I_e - \mu_1 k_T \frac{\partial N_i}{\partial x},$$

$$\frac{\partial E}{\partial x} = \epsilon \frac{\partial E_0}{\partial x} + \epsilon \frac{\partial E_1}{\partial x},$$

$$\frac{\partial E_0}{\partial x} = e(N_{D^+} - n - N_A) + e(N_i - N_h).$$

Fig. 1. Dynamic behavior of the photorefractive space-charge field amplitude $|E_1|$ observed in KNbO$_3$ crystals at elevated temperatures ($\sim$100°C). In regions A and B the crystal is illuminated with two laser beams used for recording. In regions C and D the crystal is in the dark or is illuminated by only one homogeneous beam. Charge compensation of a primary space-charge grating by secondary charge carriers and the subsequent revealing of the hologram are well described by a single exponential function. When the temperature is varied, the typical time constants observed during these processes can be made much longer than the writing or the revealing time. These properties permit the fixing of the phase gratings with the procedure described in Ref. 15.
The symbols in the above equations are \( n \), the free-electron concentration in the conduction band; \( N_{Do} \), the donor concentration; \( N_{Do}^+ \), the concentration of ionized donors; \( N_e \), the average concentration of ionic charges; \( N_i \), the concentration of ions; \( N_A \), the concentration of ionized donors in the dark; \( J_e \), the electronic current density; \( J_i \), the ionic current density; \( E_o \), the electronic contribution to the space-charge field; \( E_i \), the ionic contribution to the space-charge field; \( E \), the total electric field; \( I \), the light intensity; \( s \), the photoionization constant; \( \beta \), the dark generation rate for electrons; \( \gamma \), the recombination constant for electrons; \( \mu_e \), the electronic mobility; \( \mu_i \), the ionic mobility; \( \varepsilon_0 \), the permittivity of vacuum; \( \varepsilon \), the effective dielectric constant; \( \varepsilon_{eff} \), the absolute value of the elementary charge; \( k_B \), the Boltzmann constant; and \( T \), the temperature.

We consider the usual case of a sinusoidally varying light intensity distribution of the form

\[
I(x) = I_0 + I_1 \exp(iKx),
\]

where \( K = 2\pi/\Lambda \) is the grating wave vector associated with the grating spacing \( \Lambda \).

Following the usual linearization procedure for small-modulation \( (I_1 \ll I_0) \), we can set the quantities \( N_{Do}, N_i, n, E, J_e, \) and \( J_i \) in a sinusoidal form of the type

\[
F(x) = F_0 + F_1 \exp(iKx),
\]

where only the real part has physical meaning. In Eq. (3) the modulated quantities \( F_1 \) for \( N_{Do}^+ \), \( N_i \), and \( n \) are small compared with the space-averaged quantities \( F_0 \). Inserting Eqs. (2) and (3) into the set of relations (1a)–(1f), one gets three coupled linear differential equations for the first-order variables \( n_1, N_{Di}^+, \) and \( N_i \). They are

\[
\frac{\partial n_1}{\partial t} = - (\Gamma_{re} + \Gamma_{de} + \Gamma_{di} - i\Gamma_{de}) n_1 + (\Gamma_{re} - \Gamma_{ki}) N_{Di}^+ + \Gamma_{di}N_i + s(N_{Do} - N_{Do}^+)I_1,
\]

(4a)

\[
\frac{\partial N_{Di}^+}{\partial t} = - \Gamma_{re}n_1 - \Gamma_{de} N_{Di}^+ + s(N_{Do} - N_{Do}^+)I_1,
\]

(4b)

\[
\frac{\partial N_i}{\partial t} = \Gamma_{di}n_1 - \Gamma_{di} N_{Di}^+ - \Gamma_{di}I_1 + \Gamma_{di} + i\Gamma_{di} I_1 N_i,
\]

(4c)

where \( s(N_{Do} - N_{Do}^+)I_1 \) is a time-independent inhomogeneous term. The time rates \( \Gamma \) in Eqs. (4a)–(4c) are defined as follows:

**Dielectric rate for electrons**

\[
\Gamma_{de} = \frac{\varepsilon \mu_e n_0}{\varepsilon e_0}.
\]

(5a)

**Sum of production and recombination rate**

\[
\Gamma_e = sI_0 + \beta + \gamma n_0.
\]

(5b)

**Recombination rate for electrons**

\[
\Gamma_{re} = \gamma(N_A + n_0). \tag{5c}
\]

**Diffusion rate for electrons**

\[
\Gamma_{de} = \frac{K^2 k_B T_{\mu e}}{e}.
\]

(5d)

In the following sections we analyze the solutions of the set of differential equations (4a)–(4c) for the steady state (Subsection 2.1) and for the dynamic evolution (Subsections 2.2 and 2.3). We need to make certain approximations in order to get analytical expressions. The theoretical relationships are illustrated with figures drawn by using the set of parameters shown in Table 1. In some cases the analytical expressions are compared with exact solutions of Eqs. (4a)–(4c) obtained with the same set of parameters.

In Table 1 all the values characterizing the external conditions, such as the light intensities, the effective modulation index \( m' \), and the temperature, are arbitrary but correspond to usually applied experimental conditions. The other parameters have been chosen in such a way that the most important predictions of the theoretical model can be well identified in the different figures. In particular, the most important parameter in Table 1 is the mean density of ions \( N_{Do} \). Its value must be large compared with \( N_A \) and \( N_{Do} \), for there to be appreciable compensation effects. Some of the remaining parameters that describe crystal properties are particularly related to KNbO_3 crystals, but values appropriate to BaTiO_3, Bi_{12}SiO_{20}, or other photorefractive crystals can be used without qualitative modifications in the theoretical predictions. The number \( N_A \) of ionized donors in the dark corresponds to a typical value for slightly doped KNbO_3 crystals. The small ionic mobility \( \mu_i \) is chosen in such a way that the product \( \mu_i N_{Do} \) is consistent with experimental observations in KNbO_3. The relatively small electron mobility \( \mu_e \) should be considered a trap-limited mobility and not a true conduction band mobility. Finally, the density of donors \( N_{Do} \) is estimated from normal doping concentrations. It is chosen in such a way that its combination with \( N_A, s, \beta, \gamma, \) and \( \mu_e \) through Eqs. (6) and (5a) gives dielectric relaxation rates consistent with observations in reduced KNbO_3.
For the steady state, all time derivatives in Eqs. (4a)-(4c) are zero. The value of the total space-charge field \( E_1 \) is then given by

\[
E_1 = \frac{e}{\varepsilon_0 i K} \left[ s(N_D - N_{D0}) I_1 (\Gamma_{de} + i \Gamma_{el}) (\Gamma_{de} - i \Gamma_{el}) \right] / \left[ \Gamma_{de} (\Gamma_{de} + \Gamma_{el}) (\Gamma_{de} - i \Gamma_{el}) + \Gamma_{el} (\Gamma_{de} + i \Gamma_{el}) (\Gamma_{de} - i \Gamma_{el}) \right].
\]  

For cw illumination of the crystal sample the concentration \( n_o \) of free charges in the conduction band is small compared with the concentrations \( N_{D0} \) and \( N_A \). In this case it is reasonable to assume that the sum of production and recombination rate \( \Gamma_D \) is small compared with the recombination rate \( \Gamma_{de} \) and that the dielectric rate for electrons \( \Gamma_{el} \) is small compared with the corresponding diffusion rate \( \Gamma_{de} \). The solution [Eq. (7)] can then be reduced to a simpler form:

\[
E_1 = \frac{-im E_q (E_{D0}^2 + E_0^2)}{(E_{D0} + E_0 + i E_0)(E_{D0} - i E_0) + E_q (E_{D0} + i E_0)}.
\]  

where we introduce standard quantities such as \( E_D = K k_B T / e \) for the diffusion field, \( m' = s I_1 / (s I_0 + \beta) \) for the effective modulation index, and

\[
E_q = \frac{e}{\varepsilon_0 i K} \frac{N_A (N_D - N_A)}{N_{D0}} = \frac{e}{\varepsilon_0 i K} N_{D0}
\]

for the maximum space-charge field that can be created by the electrons. The term \( N_{D0} \) in Eq. (9) defines an effective number of traps. In analogy with \( E_q \) a new limiting field was introduced in Eq. (8), that is,

\[
E_{cl} = \frac{e N_{D0}}{\varepsilon_0 K}.
\]

Table 1. Values of the Crystal Parameters and of the External Conditions Used for Modeling the Photorefractive Response

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
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<tbody>
<tr>
<td>Basic material parameters</td>
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<tr>
<td>Photoionization constant</td>
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<tr>
<td>Dark generation rate</td>
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<tr>
<td>Recombination constant</td>
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<tr>
<td>Donor concentration</td>
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<tr>
<td>Accepter concentration</td>
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<td>Ion concentration</td>
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<td>Electron mobility</td>
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<td>Ion mobility</td>
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<tr>
<td>Relative dielectric constant</td>
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<tr>
<td>External parameters</td>
<td></td>
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<tr>
<td>Light intensity during recording</td>
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<tr>
<td>Light intensity during erasure</td>
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<td>Effective modulation index</td>
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<tr>
<td>Temperature</td>
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<tr>
<td>Derived parameters</td>
<td></td>
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<tr>
<td>Electron concentration (by ( I_0 = 100 ) W/m(^2))</td>
<td></td>
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<tr>
<td>Dielectric relaxation rate for electrons (by ( \mu_e = 10^{-8} ) m(^2)V/s)</td>
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<tr>
<td>Dielectric relaxation rate for ions</td>
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<tr>
<td>Debye wave vector for electrons</td>
<td></td>
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<tr>
<td>Debye wave vector for ions</td>
<td></td>
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<tr>
<td>Inverse of diffusion length</td>
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</tbody>
</table>

1. Steady-State Solutions
For the steady state, all time derivatives in Eqs. (4a)-(4c) are zero. The value of the total space-charge field \( E_1 \) is then given by

\[
E_1 = \frac{e}{\varepsilon_0 i K} \left[ s(N_D - N_{D0}) I_1 (\Gamma_{de} + i \Gamma_{el}) (\Gamma_{de} - i \Gamma_{el}) \right] / \left[ \Gamma_{de} (\Gamma_{de} + \Gamma_{el}) (\Gamma_{de} - i \Gamma_{el}) + \Gamma_{el} (\Gamma_{de} + i \Gamma_{el}) (\Gamma_{de} - i \Gamma_{el}) \right].
\]  

For cw illumination of the crystal sample the concentration \( n_o \) of free charges in the conduction band is small compared with the concentrations \( N_{D0} \) and \( N_A \). In this case it is reasonable to assume that the sum of production and recombination rate \( \Gamma_D \) is small compared with the recombination rate \( \Gamma_{de} \) and that the dielectric rate for electrons \( \Gamma_{el} \) is small compared with the corresponding diffusion rate \( \Gamma_{de} \). The solution [Eq. (7)] can then be reduced to a simpler form:

\[
E_1 = \frac{-im E_q (E_{D0}^2 + E_0^2)}{(E_{D0} + E_0 + i E_0)(E_{D0} - i E_0) + E_q (E_{D0} + i E_0)},
\]

where we introduce standard quantities such as \( E_D = K k_B T / e \) for the diffusion field, \( m' = s I_1 / (s I_0 + \beta) \) for the effective modulation index, and

\[
E_q = \frac{e}{\varepsilon_0 i K} \frac{N_A (N_D - N_A)}{N_{D0}} = \frac{e}{\varepsilon_0 i K} N_{D0}
\]

for the maximum space-charge field that can be created by the electrons. The term \( N_{D0} \) in Eq. (9) defines an effective number of traps. In analogy with \( E_q \) a new limiting field was introduced in Eq. (8), that is,

\[
E_{cl} = \frac{e N_{D0}}{\varepsilon_0 K}.
\]

representing the maximum space-charge field that can be created by the ionic charges with density \( N_{D0} \). The total modulated space-charge field \( E_1 \) of Eq. (8) is a superposition of an electronic component \( E_{el} \) and an ionic component \( E_{cl} \), which can be expressed as

\[
E_{el} = \frac{-im E_q (E_{D0} + E_0 + i E_0)(E_D - i E_0)}{(E_{D0} + E_0 + i E_0)(E_D - i E_0) + E_q (E_D + i E_0)},
\]

\[
E_{cl} = \frac{+im E_q E_{D0}(E_D - i E_0)}{(E_{D0} + E_0 + i E_0)(E_D - i E_0) + E_q (E_D + i E_0)}.
\]

(11a)

(11b)

We first discuss the results in Eqs. (8), (11a), and (11b) in the simpler case for which no field is applied to the crystal, that is, \( E_D = 0 \). The space-charge fields \( E_1, E_{el}, \) and \( E_{cl} \) are then purely imaginary, indicating a phase shift of \( \pm \pi/2 \) with respect to the light interference fringes, and are given by

\[
E_1 = \frac{-im E_D}{1 + E_D + E_{cl}},
\]

\[
E_{el} = \frac{-im E_q E_{D0}}{E_q + E_D + E_{cl}},
\]

\[
E_{cl} = \frac{+im E_q E_{D0}}{E_q + E_D + E_{cl}}.
\]

(12a)

(12b)

(12c)

These equations show that if the number of ions \( N_{D0} \) greatly exceeds the effective number of traps \( N_{D0} \), that is, \( E_{D0} \gg E_q \), the total space-charge field \( E_1 \) in Eq. (12a) becomes small, well below the value of \( E_D \) or \( E_{D0} \). This corresponds to the steady-state value of \( E_1 \) shown in Fig. 1 at point 2 and to the experimental observations in KNbO\(_3\) at high temperatures.
The grating spacing dependencies of the steady-state values for the fields $E_1$ and $E_{d1}$ predicted by Eqs. (8), (12a), and (12b) are shown in Fig. 2 for the set of model parameters of Table 1. Since $E_1 = E_{d1} + E_{i1}$ is small, the field $E_{i1}$ (not shown in Fig. 2) takes approximately the same values as $E_{d1}$, but with opposite sign. Figure 2 shows that for no applied fields the compensation is more effective at larger grating spacing where the field $E_1$ becomes smaller. At the same time, both $E_{d1}$ and $E_{i1}$ become larger with larger grating spacing and reach the value $mE_0$ even if no electric field is applied to the sample. This is different from the conventional case in the absence of ions. There a space-charge field close to the value of the diffusion field because of the presence of the compensating ionic grating.

We now discuss the case with $E_0 \neq 0$. The dashed-dotted curve in Fig. 2 shows that the steady-state value of the total space-charge field $E_1$ is no longer so strongly dependent on the grating spacing $A$ as for $E_0 = 0$. For large enough $A$ and $E_0$, $E_1$ is proportional to $mE_0/E_{d0}$ and is essentially in phase with the light interference fringes. On the other side, unless the applied field $E_0$ exceeds $E_{d0}$, the unshifted (that is, real) component of the electronic and the ionic contribution to the space-charge field are still small compared with the $\pm \pi/2$ phase-shifted components. Nevertheless, the resulting field $E_1$ is formed by these unshifted components that have opposite signs and different magnitude for electrons and ions.

2. Dynamic Behavior with No Applied Field
When we set $E_0 = 0$, the set of Eqs. (4a)–(4c) can be solved exactly, giving the grating dynamics in the small-modulation approximation. In this case the time evolution of $N_{i0}^+$, $N_{i1}$, and $n_1$ takes a triple exponential form of the type

$$G_1(t) = G_{1(\Gamma_1)} \exp(-\Gamma_1 t) + G_{1(\Gamma_2)} \exp(-\Gamma_2 t) + G_{1(\Gamma_3)} \exp(-\Gamma_3 t) + G_{\text{sat}}.$$  

(13)

The $G_{1(\Gamma_i)}$ are nine real amplitudes for $N_{i0}^+$, $N_{i1}$, and $n_1$, and the constant terms $G_{\text{sat}}$ account for the inhomogeneity of the set of differential Eqs. (4a)–(4c). The three time rates $\Gamma_1$, $\Gamma_2$, and $\Gamma_3$ are obtained from a cubic characteristic equation derived from Eqs. (4a)–(4c). This is

$$\Gamma^3 - (\Gamma_{\text{tot}})^2 \Gamma - (\Gamma_{\text{tot}})^3 = 0,$$

(14)

where the constant terms $\Gamma_{\text{tot}}$, $\Gamma_{\text{tot}}^2$, and $\Gamma_{\text{tot}}^3$ contain the characteristic time rates listed in Eqs. (5a)–(5h) and are defined below. Since the fields $E_1$, $E_{d1}$, and $E_{i1}$ are linearly related to the amplitudes of the modulated concentrations $N_{i0}^+$, $N_{i1}$, and $n_1$, they also have the form of Eq. (13) but are merely imaginary, showing the expected $\pi/2$ phase shift. The amplitudes $G_{1(\Gamma_i)}$ are determined by the requirement that Eq. (13) fulfill the appropriate initial conditions. The constant term $G_{\text{sat}}$ vanishes in the erasure process ($I_1 = 0$) and assumes the steady-state values of Eqs. (12a)–(12c) for the recording process.

We now want to formulate an analytical expression for the three time rates $\Gamma_1$, $\Gamma_2$, and $\Gamma_3$ involved in the dynamics of the space-charge fields. In the case with no applied field they are all real and positive. The three coefficients in Eq. (14), which is used for obtaining the three solutions $\Gamma_1$, $\Gamma_2$, and $\Gamma_3$, are as follows. The first coefficient is equal to

$$\Gamma_{\text{sat}} = \Gamma_{\text{dir}} + \Gamma_{\text{el}} + \Gamma_{\text{Re}} + \Gamma_{\text{di}} + \Gamma_{\text{ei}} + \Gamma_{\text{diff}},$$

(15)

which is a total rate given by the sum of all the nonvanishing rates in Eqs. (5a)–(5h); the second coefficient is equal to

$$\Gamma_2 = \left[ \Gamma_{\text{dir}}(\Gamma_{\text{di}} + \Gamma_{\text{el}} + \Gamma_{\text{Re}}) + \Gamma_{\text{di}}(\Gamma_{\text{di}} + \Gamma_{\text{el}} + \Gamma_{\text{Re}}) + \Gamma_{\text{dir}}(\Gamma_{\text{di}} + \Gamma_{\text{el}} + \Gamma_{\text{Re}}) \right]^{1/2};$$

(16)

and the third one is

$$\Gamma_3 = \left[ \Gamma_{\text{dir}}(\Gamma_{\text{di}} + \Gamma_{\text{el}} + \Gamma_{\text{Re}}) + \Gamma_{\text{di}}(\Gamma_{\text{di}} + \Gamma_{\text{el}} + \Gamma_{\text{Re}}) \right]^{1/3}. $$

(17)

It can be verified that $\Gamma_{\text{tot}} > \Gamma_1$, $\Gamma_2$, and the solutions of Eq. (14) can be developed in a series in terms of $(\Gamma_1/\Gamma_{\text{tot}})^2$ and $(\Gamma_2/\Gamma_{\text{tot}})^3$, leading in the first approximation to the following expressions:

$$\Gamma_1 = \Gamma_{\text{tot}},$$

(18a)

$$\Gamma_2 = \frac{(\Gamma_1)^2}{\Gamma_{\text{tot}}} - \frac{(\Gamma_1)^3}{(\Gamma_{\text{tot}})^2} \frac{\Gamma_{\text{tot}}(\Gamma_1)^4}{(\Gamma_{\text{tot}})^2} - 2 \frac{(\Gamma_1)^2(\Gamma_1)^5}{(\Gamma_{\text{tot}})^2} \frac{(\Gamma_1)^6}{(\Gamma_{\text{tot}})^2} + \ldots = \frac{(\Gamma_1)^2}{\Gamma_{\text{tot}}} - \Gamma_3,$$

(18b)

$$\Gamma_3 = \frac{(\Gamma_1)^3}{(\Gamma_{\text{tot}})^2} + \frac{\Gamma_{\text{tot}}(\Gamma_1)^5}{(\Gamma_{\text{tot}})^2} + 2 \frac{\Gamma_{\text{tot}}(\Gamma_1)^5(\Gamma_1)^6}{(\Gamma_{\text{tot}})^2} + \frac{(\Gamma_{\text{tot}})(\Gamma_1)^7}{(\Gamma_{\text{tot}})^2} + \ldots. $$

(18c)

The time rate $\Gamma_1$ describes mainly the dynamics of the free-electron grating in the conduction band. The experiments show that the related time constant $1/\Gamma_1$ is normally in the microsecond region. For cw illumination it can be shown that the space-charge field amplitudes accompanying this time constant are small. Therefore, unless the grating has been formed by intensive pulsed light, the term containing $\Gamma_1$ in the time evolution of the space-charge field [Eq. (13)] can be neglected.
We concentrate further on the two other time rates, \( \Gamma_2 \) and \( \Gamma_3 \). In most of the cases the leading terms in Eqs. (18b) and (18c) dominate, so that \( \Gamma_2 \) and \( \Gamma_3 \) can be written as

\[
\Gamma_2 \approx \left( \frac{\Gamma_{\text{tot}}}{\Gamma_1} \right)^2, \tag{19a}
\]

\[
\Gamma_3 \approx \left( \frac{\Gamma_{\text{tot}}}{\Gamma_2} \right)^2. \tag{19b}
\]

It is usually also true that the recombination rate \( \Gamma_{Re} \) and the diffusion rate \( \Gamma_{De} \) for electrons are much larger than all other time rates in Eqs. (5a)-(5h), and relations (19a) and (19b) can be put into more useful forms; that is,

\[
\Gamma_3 = \Gamma_{\text{dif}} \left[ 1 + \frac{K^2}{K_{0f}^2} \right] + \Gamma_{\text{diff}} \left[ 1 + \frac{K^2}{K_{00}^2} \right], \tag{20}
\]

\[
\Gamma_3 = \Gamma_{\text{dif}} \left[ \frac{K^2}{K_{0f}^2} + \frac{K^2}{K_{00}^2} \right] + \Gamma_{\text{diff}} \left[ 1 + \frac{K^2}{K_{0f}^2} (1 + \frac{K^2}{K_{00}^2}) \right], \tag{21}
\]

where

\[
K_{0f}^2 = \frac{e^2 N_A (N_{De} - N_A)}{\varepsilon \varepsilon_0 k_B T N_{De}} = \frac{e^2 N_{\text{diff}}}{\varepsilon \varepsilon_0 k_B T}, \tag{22}
\]

is the square of the Debye wave vector for the electrons,

\[
K_{00}^2 = \frac{e^2 N_A}{\varepsilon \varepsilon_0 k_B T}, \tag{23}
\]

is the square of the corresponding wave vector for ions, and

\[
K_r^2 = \frac{e_N A}{\mu_r k_B T}, \tag{24}
\]

is the square of the inverse diffusion length.

The grating spacing dependencies of \( \Gamma_2 \) and \( \Gamma_3 \) are illustrated in Figs. 3 and 4. The curves are obtained from analytical approximations (20) and (21) and by exactly solving Eq. (14). The rate \( \Gamma_2 \) given in Eq. (20) corresponds to the rate constant found in the one charge carrier model augmented by the term containing \( \Gamma_{\text{dif}} \). It can be generally ascribed to the time evolution of the primary electronic grating and describes the dynamics during the writing (region A of Fig. 1) and revealing (region C of Fig. 1) times of the hologram. Only few ions are being redistributed in a time scale given by this rate.

Equation (21) describes the grating spacing dependence of the time rate \( \Gamma_3 \). This is the rate describing the dynamics during the storage time (in the dark), the handling time (under homogeneous illumination), or the compensating time (under inhomogeneous illumination) of a grating in ionic compensating materials. Both ions and electrons can redistribute in a time scale given by \( \Gamma_3 \).

We now discuss the grating spacing dependence of \( \Gamma_3 \) predicted by Eq. (21) and shown in Fig. 4. We consider the case in which the crystal contains a number of ions \( N_{De} \), which is large compared with the effective number of traps \( N_{\text{diff}} \); that is, \( K_{0f}^2 \gg K_{00}^2 \). We assume further that the temperature is not too high, so that \( \Gamma_{\text{dif}} \ll \Gamma_{\text{diff}} \) is fulfilled. This can be easily realized by adapting the experimental conditions. Equation (21) can then be simplified to

\[
\Gamma_3 \approx \frac{\Gamma_{\text{dif}} K^2}{K_{00}^2 + K^2}. \tag{25}
\]

Therefore, for small grating spacings \( (K \to \infty) \), \( \Gamma_3 \) is given by the dielectric relaxation time for the ions \( \Gamma_{\text{dif}} \). This represents the dielectric limit. It is interesting to remark that this is in contrast with the case of the time rate \( \Gamma_2 \) related to the primary grating. There the dielec-
tric limit is found for large grating spacings (Fig. 3). Equation (25) predicts for large grating spacings ($K \to 0$) a quadratic dependence of $\Gamma_3$ on $K$; that is, compensating, storage, and handling time increase with the square of the grating spacing $\Lambda$.

The information contained in Figs. 3 and 4 allows us also to discuss the domain of validity of approximations (20) and (21), which are derived from relations (19a) and (19b). For the set of parameters used to draw the upper curves in the two figures the dielectric relaxation rates for electrons and ions have the same value, $\Gamma_{d/e} = 6.58 \text{s}^{-1}$. The figures show that in such a case the agreement between the exact solution and the analytical expressions (20) and (21) is not very good for grating spacings smaller than 1 $\mu$m. The reason is that the higher-order terms in the development of Eqs. (18b) and (18c) also become important and cannot be neglected. In contrast, if the dielectric relaxation rate for the ions is small enough (lower curves in Figs. 3 and 4), the higher-order terms are negligible. In this case a measurement of the time rate $\Gamma_3$ as a function of the grating spacing permits the determination of the Debye wave vector $K_d$ by fitting the results to Eq. (25). This gives a powerful new method for determination of the effective number of acceptors $N_{\text{eff}}$ from Eq. (22).

We have seen that, since the term evolving with the time rate $\Gamma_3$ in Eq. (13) can be neglected, the dynamics is essentially given by a double exponential function. Figures 5(a) and 5(b) show the exact solutions of the set of Eqs. (4a)–(4c) during the full dynamic evolution. Figure 5(a) is for a grating spacing $\Lambda = 0.5 \, \mu$m, and Fig. 5(b) is for $\Lambda = 2 \, \mu$m. The parameters used are the ones of Table 1, except for a 10-times larger mobility for the ions $[\mu_I = 2 \times 10^{-14} \text{m}^2(\text{V s})^{-1}]$. The experimentally observed dynamics in KNbO$_3$ (drawn schematically in Fig. 1) is well reproduced by the exact solutions shown in Figs. 5(a) and 5(b). This shows that, unlike in LiNbO$_3$, the fixing of volume-phase holograms in KNbO$_3$ can be explained without the need for photovoltaic currents, because this contribution is not included in our model.

For the set of parameters used for Fig. 5(a) and 5(b) the dielectric relaxation times for electrons and ions are equal. Nevertheless, the time rate $\Gamma_3$ is smaller than $\Gamma_2$ because the fields driving the erasure process are smaller than the compensating electronic and ionic contributions to the space-charge field. In addition, the figures show that the compensation is slower for $\Lambda = 2 \, \mu$m than for $\Lambda = 0.5 \, \mu$m. This is expected from Eq. (21). It is interesting to note that, if only ions would contribute to the grating observed during the storage or handling time of the hologram, the time rate $\Gamma_2$ would never be smaller than the dielectric relaxation rate $\Gamma_{d/e}$. Therefore the fact that $\Gamma_2$ decreases at large $\Lambda$ values can be explained only by the simultaneous presence of the electronic charges. Similar effects in which the interaction between different charges influences the time evolution have been observed in the field of thermoelectrets. It was shown that a system of homocharges and heterocharges decays much more slowly than a homocharge or a heterocharge alone.

The information contained in Figs. 5(a) and 5(b) allows us also to discuss the behavior of the amplitudes $E_{1(0)}$, $E_{d/e(0)}$, and $E_{1(1)}$ in Eq. (13). By comparison of the two figures one can see that the steady-state values of the two components $E_{d/e}$ and $E_{1(1)}$ are larger for $\Lambda = 2 \, \mu$m. This is expected from Eqs. (12a)–(12c) and from Fig. 2. One might suggest that the field $E_{d/e}$ would evolve in time only with the time rate $\Gamma_3$. In this case we would expect an increase in the height of the transient peaks of $E_1$ (corresponding to points 1 and 3 in Fig. 1) for larger grating spacings. However, this contradicts both the experimental observations and the predictions of our model represented in Figs. 5(a) and 5(b).

The reason is that at larger grating spacings an increasing part of the primary grating $E_{d/e}$ evolves with the time rate $\Gamma_3$, which is generally

Fig. 5. Exact solution of the dynamic Eqs. (4a)–(4c) for the model parameters of Table 1 except for a 10-times increased ionic mobility $[\mu_I = 2 \times 10^{-14} \text{m}^2(\text{V s})^{-1}]$. Solid curves, total space-charge field $E_1 = E_{d/e} + E_{1(1)}$; dashed curves, electronic (primary) space-charge field $E_{d/e}$; dotted–dashed curves, ionic (secondary) space-charge field $E_{1(1)}$. Note the different scales for both the $x$ and $y$ axes in (a) and (b). The numerical values of all the parameters describing the solution for the field $E_1(t)$ following Eq. (13) are given. (a) Grating spacing $\Lambda = 0.5 \, \mu$m. Recording: $\Gamma_1 = 1.0 \times 10^7 \text{s}^{-1}$; $\Gamma_2 = 23.25 \, \text{s}^{-1}$; $\Gamma_3 = 9.7 \, \text{s}^{-1}$; $E_{1(0)} = 0.07 \text{V/m}$; $E_{0(0)} = -41784 \text{V/m}$; $E_{1(1)} = +41060 \text{V/m}$; $E_{s}(0) = 723 \, \text{V/m}$; Erasure: $\Gamma_1 = 1.0 \times 10^7 \text{s}^{-1}$; $\Gamma_2 = 17.57 \, \text{s}^{-1}$; $\Gamma_3 = 4.78 \, \text{s}^{-1}$; $E_{1(0)} = -0.05 \text{V/m}$; $E_{1(1)} = +43402 \text{V/m}$; $E_{s}(0) = -42679 \, \text{V/m}$; $E_{s}(0) = 0$. (b) Grating spacing $\Lambda = 0.5 \, \mu$m. Recording: $\Gamma_1 = 1.0 \times 10^7 \text{s}^{-1}$; $\Gamma_2 = 23.25 \, \text{s}^{-1}$; $\Gamma_3 = 9.7 \, \text{s}^{-1}$; $E_{1(0)} = 0.07 \text{V/m}$; $E_{0(0)} = -41784 \text{V/m}$; $E_{1(1)} = +41060 \text{V/m}$; $E_{s}(0) = 723 \, \text{V/m}$; Erasure: $\Gamma_1 = 1.0 \times 10^7 \text{s}^{-1}$; $\Gamma_2 = 17.57 \, \text{s}^{-1}$; $\Gamma_3 = 4.78 \, \text{s}^{-1}$; $E_{1(0)} = -0.05 \text{V/m}$; $E_{1(1)} = +43402 \text{V/m}$; $E_{s}(0) = -42679 \, \text{V/m}$; $E_{s}(0) = 0$. (b) Grating spacing $\Lambda = 2 \, \mu$m. Recording: $\Gamma_1 = 5.3 \times 10^6 \text{s}^{-1}$; $\Gamma_2 = 16.31 \, \text{s}^{-1}$; $\Gamma_3 = 0.85 \, \text{s}^{-1}$; $E_{1(0)} = 0.05 \text{V/m}$; $E_{0(0)} = -24271 \text{V/m}$; $E_{1(1)} = +24089 \text{V/m}$; $E_{s}(0) = 154 \text{V/m}$; $E_{s}(0) = 13.53 \, \text{s}^{-1}$; $E_{1(0)} = -0.05 \text{V/m}$; $E_{1(1)} = +21342 \text{V/m}$; $E_{s}(0) = -21158 \text{V/m}$; $E_{s}(0) = 0$. 
neglect the first term in Eq. (20), and than the corresponding rate for electrons. We can then of the time rates interrupted before saturation is reached. 

It must be mentioned that the dynamics shown in Fig. 6 for the erasure process is only partial. It can be recognized from Fig. 6 that during the erasure process the field $E_1$ crosses zero at the point where the ionic and the electronic contributions have equal amplitude. Afterward $E_1$ recovers in part but with the opposite sign. This behavior is found also in the situation of Figs. 5(a) and 5(b). There the zero-crossing point occurs at the beginning of the erasure process. It must be mentioned that the dynamics shown in Fig. 6 for the erasure process is found also for full compensating crystals having $N_0 >> N_{eff}$ if the recording process is interrupted before saturation is reached.

We want now to discuss the light intensity dependence of the time rates $\Gamma_3$ and $\Gamma_4$. We limit ourselves to the case in which the dielectric relaxation rate for ions is smaller than the corresponding rate for electrons. We can then neglect the first term in Eq. (20), and $\Gamma_3$ becomes linearly dependent on $\Gamma_{d_1}$. Therefore its value increases linearly with illuminating intensity $I_0$, as is expected from the conventional Kukhtarev model.23

The case of the time rate $\Gamma_4$ is more complicated. Equation (21) indicates a monotone increase of $\Gamma_4$ with light intensity $I_0$ because $\partial I_4/\partial I_0 > 0$. This increase is very small as far as $I_{d_1}$ sufficiently exceeds $I_{d_1}$. Experimentally, we have observed such a slight increase in most of our KNbO$_3$ samples. However, in one KNbO$_3$ crystal showing small trap-limited electronic mobility,26 we have observed that $\Gamma_3$ becomes smaller with increasing intensity. An example of such an observation is reported in Fig. 5 of Ref. 15. As we said above, such a decrease cannot be explained by Eq. (21). In the derivation of Eq. (21) it was assumed that the electronic dielectric relaxation rate $\Gamma_{d_e}$ was small compared with the diffusion rate $\Gamma_{d_1}$. If this assumption is abandoned, an additional term must be considered in the denominator of Eq. (21). We give this correction in the special case previously described by expression (25), that is, in the limit $N_{d_1} >> N_{eff}$ and $\Gamma_{d_1} >> \Gamma_{d_e}$. Equation (25) is then transformed into

$$\Gamma_3 = \frac{K^2}{K_{d_1}^2 + K^2 + K \frac{\Gamma_{d_1}}{\Gamma_{d_e}}}.$$

Thus a large decrease of $\Gamma_3$ with light intensity can be expected when $K^2 \Gamma_{d_e}/\Gamma_{d_1}$, which equals $e^{2N_0}/ee{k_B}T$, becomes comparable with $K_{d_1}^2$, that is, if the free-electron concentration $n_0$ becomes comparable with the effective number of traps $N_{eff}$.

We illustrate this effect in Fig. 7, which shows the intensity dependence of the normalized time rate $\Gamma_3(I_0 \neq 0)/\Gamma_3(I_0 = 0)$. The solid curve is drawn following Eq. (19b) for the parameters of Table 1. The other curves in the figure are drawn by changing the indicated parameters (one at a time) with respect to the ones of Table 1. The smaller intensities required for the drop of $\Gamma_3$ with decreasing recombination constant $\gamma$ are due to the increased electron density $n_0$, whereas the decrease for smaller $N_0$ is due mainly to the decrease of $K_{d_1}^2$. 

Fig. 7. Intensity dependence of the time rate $\Gamma_3$ as predicted from Eq. (19b) for $\Lambda = 2 \mu m$. The values are divided with the value of $\Gamma_3$ in the dark. The solid curve is drawn with the parameters of Table 1. The other curves are obtained by modifying only the indicated parameter to the given value.
3. Dynamic Behavior with Applied Field

Equation (13), which describes the time evolution of the amplitude $F_1$ in Eq. (3), is still valid, but the relaxation rates $\Gamma_i$ are now complex. For later use we call the complex rates $\Gamma_1^*$, $\Gamma_2^*$, and $\Gamma_3^*$. The imaginary parts of the three time rates produce a time-dependent change of the phase shift between the light fringes and the three components of the grating. As long as the steady state is not reached, every component of the grating in Eq. (13) is running inside the crystal with velocity $\Im(\Gamma_3^*)/K$. Therefore the imaginary part of the time rates produces oscillations in the dynamics that can be observed experimentally in a two-beam coupling configuration in which the beam coupling gain is sensitive to phase changes. However, the observation of such oscillations is possible only if the oscillatory terms $\Im(\Gamma_1^*)$ are not too small with respect to the exponential decay rates $\Re(\Gamma_1^*)$. Hence, $\Gamma_1^*$ is the wave vector for which the applied field equals the diffusional wave vector $K_D$, this is the exponential decay rate.

We concentrate now on the three complex time rates $\Gamma_1^*$, $\Gamma_2^*$, and $\Gamma_3^*$. They are the three solutions of the cubic equation (14), this time with complex coefficients $\Gamma_{1\text{tot}}^*$, $\Gamma_{2\text{tot}}^*$, and $\Gamma_{3\text{tot}}^*$. The first coefficient is defined as

$$\Gamma_{1\text{tot}}^* = \Gamma_{1\text{tot}} + i(\Gamma_{1\text{lf}} - \Gamma_{1\text{lf}}),$$

where $\Gamma_{1\text{lf}}$ and $\Gamma_{1\text{lf}}$ are the drift rates for ions and electrons defined in Eqs. (5b) and (5e) and $\Gamma_{1\text{tot}}$ is given in Eq. (15). The second constant is

$$(\Gamma_{2\text{tot}}^*)^2 = \Gamma_0^2 + \Gamma_{\text{rel}} \Gamma_{\text{rel}} + i(\Gamma_{\text{rel}} \Gamma_{\text{rel}} + \Gamma_{\text{rel}} + \Gamma_{\text{rel}} + \Gamma_{\text{rel}})$$

and the third is

$$(\Gamma_{3\text{tot}}^*)^3 = \Gamma_0^3 + \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} + i(\Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} + \Gamma_{\text{rel}} + \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} - \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} + \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} - \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} - \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} - \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} - \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}} - \Gamma_{\text{rel}} \Gamma_{\text{rel}} \Gamma_{\text{rel}})$$

The solutions $\Gamma_i^*$ of Eq. (14) can still be expressed in good approximation by Eqs. (18a)–(18c) by replacing all the real constants with the corresponding complex constants. As in the derivation of Eqs. (20) and (21) from relations (19a) and (19b), we consider the usual case with the diffusion rate $\Gamma_{\text{rel}}$, the recombination rate $\Gamma_{\text{rel}}$, and the drift rate $\Gamma_{\text{rel}}$ for electrons to be larger than the other rates defined in Eqs. (5a)–(5h). We assume further that $\Gamma_{\text{rel}} \gg \Gamma_{\text{rel}}$. The quantities $\Gamma_0^*$, $\Gamma_{\text{rel}}^*$, and $\Gamma_{\text{rel}}^*$ can then be obtained from relations (18a), (19a), and (19b) if the real variables $\Gamma_{1\text{tot}}^*$, $\Gamma_{2\text{tot}}^*$, and $\Gamma_{3\text{tot}}^*$ in these three expressions are replaced by the corresponding complex variables. However, the expressions are rather lengthy, and we give only the values for $\Gamma_2^*$. From relation (19a) we get

$$\Re(\Gamma_2^*) \equiv \Gamma_{\text{rel}} \left[ 1 + \frac{K_D^2}{K_{\text{rel}}^2} \right]$$

for its real part and

$$\Im(\Gamma_2^*) \equiv \Gamma_{\text{rel}} \left[ -\frac{K_D^2}{K_{\text{rel}}^2} \right]$$

for its imaginary part, where

$$K_D = \frac{eE_0}{K_{\text{rel}} T}$$

is the wave vector for which the applied field equals the diffusion field. The expected dependencies of $\Re(\Gamma_2^*)$ and $\Im(\Gamma_2^*)$ on the externally applied field are shown in Fig. 8 for $\Lambda = 0.5$, 2, and 10 $\mu$m. The influence of the field on the real part of $\Gamma_2^*$ is larger for large grating spacing $\Lambda$, this is the same value obtained without the field for small grating spacings $\Lambda \rightarrow \infty$ in Eq. (20). Figure 8 shows that if the ionic mobility is small, the imaginary part [relation (31)] vanishes both for zero and for large electric fields and reaches a maximum for an intermediate value given by $E_0 = (k_B T/e)(K^2 + K_D^2)/K$. For this value of $E_0$ one observes the fastest oscillations during the writing or the revealing time of the hologram.

Without giving the analytical expressions, we also discuss the electric-field dependencies of the real and the imaginary parts of the time rate $\Gamma_3^*$. Figure 9 shows $\Re(\Gamma_3^*)$ and $\Im(\Gamma_3^*)$ for different values of the grating spacing $\Lambda$ as they are obtained from Eq. (19b) with the parameters of Table 1. For small $\Lambda$ the electric field has only little influence on the real part of $\Gamma_3^*$. In this case the value of $\Re(\Gamma_3^*)$ is close to $\Gamma_{\text{rel}}$, the dielectric limit predicted from Eq. (21). In contrast, for large grating spacings where $\Re(\Gamma_3^*)$ in absence of an applied field is smaller, $E_0$ contributes to increase it to approximately the same value (close to $\Gamma_{\text{rel}}$) found for small fringe spacings. The region where $\Re(\Gamma_3^*)$ increases faster is found around electric-field values that fulfill $K_D(E_0) = K_{\text{rel}}(N_{\text{eff}})$. That is, if the crystal contains a larger number of traps, a larger electric field is needed to speed up the time rate $\Re(\Gamma_3^*)$.

Fig. 8. Real and imaginary parts of the time rate $\Gamma_2^*$ as a function of the externally applied electric field $E_0$. Solid curves, real part from relation (30); dashed curves, imaginary part from relation (31). The usual parameters of Table 1 are taken, and the curves for the three indicated values of the grating spacing $\Lambda$ are shown.
Carriers can be excited by absorption of a photon. Here they considered the case in which both kinds of charge carriers can be excited by absorption of a photon. Here we analyze the case in which one kind of charge carrier (for instance holes) cannot be excited optically. Then this model describes essentially the same physics as the one introduced in Subsection 2.A. We limit ourselves to the results for the case with no applied fields.

The solutions for the steady state and for the dynamic evolution of the space-charge field \( E_1 \) are obtained in the same way as in Subsection 2.A, by solving the appropriate set of equations, which is similar to Eqs. (1a)–(1f). The complete set of equations has been given in Ref. 29. We consider in our case that the photoexcitation constant for holes \( s_h \) is zero and that their thermal excitation rate \( \beta_h \) is different from zero. In analogy with Eq. (9), one may introduce the two limiting space-charge fields

\[
E_{eq} = \frac{(e/\varepsilon_0 K)[N_{D0}^+(N_{D0}^- - N_{D0}^+)/N_{D0}]}{E_D + E_{eq} + E_{ph}},
\]

for electrons and holes, respectively. In the above expressions, \( N_{D0}^+ \) and \( N_{A0}^- \) are the densities of empty electron acceptors and empty hole acceptors in the dark, respectively (Fig. 10). The steady-state solution for \( E_1 \) is given by

\[
E_1 = \frac{-imE_D E_{eq}}{E_D + E_{eq} + E_{ph}},
\]

where \( m' \) is equal to \( s_h I_i/(s_e I_0 + \beta_e) \). When \( E_{eq} \) is substituted for \( E_{ph} \) this result is fully equivalent to Eq. (12a). Therefore the effects shown in Fig. 1 can also be explained by charge compensation through thermally excited holes if the condition \( E_{eq} \gg E_{ph} \) is fulfilled; that is, the number of hole acceptors \( N_{A0}^- \) and of hole donors \( N_{A0}^+ \) should be much larger than \( N_{D0}^- \) and \( N_{D0}^+ \).

It is interesting to compare the result of Eq. (33) with the one given in literature\(^{28-31} \) for the case of allowed hole photoexcitation. There \( E_1 \) is proportional to \( (E_{eq} - E_{ph}) \). A complete compensation in the steady state would therefore be possible only if \( E_{eq} = E_{ph} \), that is, if the effective number of electron acceptors equals the effective number of hole acceptors. If \( E_{eq} \) is larger than \( E_{ph} \) the space-charge field \( E_1 \) changes sign during the recording process because the holes can be moved in an active way. In contrast, in our case the redistribution of holes is driven only by the presence of the field generated by the electronic charges. Therefore the hole contribution to the space-charge field cannot reach a magnitude larger than the one of the electronic contribution. This gives rise to the compensated space-charge field given in Eq. (33).

For the two-level model the dynamic behavior can be solved in a way similar to that for the electron–ion model. The set of Eqs. (4a)–(4c) is replaced by a set of four coupled linear differential equations in the variables \( N_{D0}^+ \), \( n_1 \), \( N_{A0}^- \), and \( p_1 \), where \( p(x) = p_0 + p_1 \exp(iKx) \) is the hole concentration in the valence band. Similar to the case for Eq. (13), the solution of this set of equations is a sum of four exponential terms with time rates \( \Gamma_0 \), \( \Gamma_1 \), \( \Gamma_2 \), and \( \Gamma_3 \). Again, two of these time rates (for instance \( \Gamma_0 \) and \( \Gamma_1 \)) are large and are connected with small amplitudes. They can be neglected for cw-laser illumination. We give the solutions for the other two time rates. The solutions are
given in the first approximation by
\[
\Gamma_2 = \frac{\Gamma_{dih}}{1 + \frac{K_h^2}{K_{oa}^2}} + \frac{\Gamma_{dir}}{1 + \frac{K_h^2}{K_{oe}^2}}
\]
for the faster of the two rates and by
\[
\Gamma_2 = \frac{\Gamma_{dir}}{1 + \frac{K_h^2}{K_{oa}^2}} + \frac{\Gamma_{dih}}{1 + \frac{K_h^2}{K_{oe}^2}}
\]
for the slower one. The new wave vectors \( K_{oh} \) and \( K_h \) appearing in Eqs. (34) and (35) are defined as
\[
K_{oh}^2 = \frac{\epsilon^2 N_{ao} (N_{ao} - N_{ao}^-)}{\epsilon \mu_h k_B T N_{ao}}
\]
for the square of the Debye wave vector for the holes and
\[
K_h^2 = \frac{\epsilon \mu_h N_{ao}^-}{\mu_h k_B T}
\]
for the square of the inverse diffusion length for holes. The dielectric relaxation rate \( \Gamma_{dih} \) for the holes is defined as
\[
\Gamma_{dih} = \frac{\epsilon \mu_h \mu_h}{\epsilon \epsilon_0}
\]
where \( \mu_h \) is the hole mobility. Equations (20) and (21) for the time rates in the ionic model are obtained from Eqs. (34) and (35) by setting \( 1 + \frac{K_h^2}{K_{oa}^2} = 1 \), \( \Gamma_{dih} = \Gamma_{dir} \), and \( K_{oh} = K_{oe} \). Therefore the only difference between the two models is given by the terms containing the hole-diffusion length \( (K_h)^{-1} \) in the second model. The definition of such a diffusion length in the electron-ion model is meaningless because excitation-recombination of ions does not take place. Equations (34) and (35) hold not only in the case of purely thermal excitation of holes but also if optical excitation is allowed (\( \theta_h \neq 0 \)).

3. EXPERIMENTS

We verify some of the predictions of the above theory in the framework of the electron-ion model of Subsection 2.A, which was treated more in detail. The analogies with the second model are discussed in Section 4. For this purpose we used an undoped KNbO\(_3\) crystal cut along the orthorhombic axis with dimensions \( a \times b \times c = 1.43 \text{ mm} \times 4.58 \text{ mm} \times 4.40 \text{ mm} \). The two largest surfaces were polished to optical quality. We placed the sample in a furnace with large aperture and heated it to increase the ionic conductivity. The measurements were performed at the temperature \( T = 107^\circ\text{C} \). An Ar\(^+\)-laser beam with wavelength \( \lambda = 514 \text{ nm} \) was split into two plane recording waves. Their polarization was horizontal and parallel to the ac plane of the crystal. For some measurements, electric fields as high as \( 2 \text{ kV/cm} \) were applied parallel to the positive \( c \) axis of the crystal. At the temperature of \( 107^\circ\text{C} \) and intensities of \( 40 \text{ mW/cm}^2 \) the dielectric relaxation rate \( \Gamma_{dir} \) for the electrons is of the order of \( 2 \text{ s}^{-1} \), which is large compared with the ionic dielectric relaxation rate \( \Gamma_{dih} \) of the order of \( 0.15 \text{ s}^{-1} \).

The recording-erasure cycle for our undoped KNbO\(_3\) is similar to the one shown in Fig. 1 and Ref. 15 with a fast increase of the diffraction efficiency in regions A and C and a slow decrease in regions B and D. We focus our attention first on the experimental verification of Eq. (21). This relationship describes the grating spacing dependence of the compensating, handling, or storage time of the grating and is therefore of great importance for the fixing characteristics of a crystal. For this purpose we switched off one of the recording beams after the compensation of the two gratings had reached saturation. The diffracted intensity of the remaining beam was then recorded by a photomultiplier tube and transferred to a computer. We then fitted the decay in region D by a single exponential function to obtain a value for the decay time constant \( \tau_0 = 1/\Gamma_3 \). This procedure was repeated at the same temperature for different values of the grating spacing \( \Lambda \). Figure 11 shows the values of \( \tau_0 \) for the case \( E_0 = 0 \), where the solid curve shows the best fit according to Eq. (21). The expected quadratic increase for longer grating spacings as well as the saturation at shorter grating spacings is well reproduced. From the fitted curve we get \( \Gamma_{dih} = (6.5 \pm 0.7) \text{ s} \) and \( K_{oe} = (2.65 \pm 0.25) \times 10^6 \text{ m}^{-1} \). With Eq. (22) we determine an effective number of traps \( N_{eff} = (0.75 \pm 0.15) \times 10^{15} \text{ cm}^{-3} \) for our crystal.

![Fig. 11. Grating spacing dependence of the handling time \( \tau_0 = (\Gamma_3)^{-1} \) in a pure KNbO3 sample measured at the temperature \( T = 107^\circ\text{C} \) with an erasing intensity \( I = 40 \text{ mW/cm}^2 \). The solid curve indicates the best fit to Eq. (21). From the fitted parameters we determine that \( \tau_{dih} = (6.5 \pm 0.7) \text{ s} \) and \( N_{eff} = (0.75 \pm 0.15) \times 10^{15} \text{ cm}^{-3} \).](image-url)
diffraction efficiency by applying a field chosen. For \( A = 18 \) and approaches the value as expected, the decay time constant decreases with that the decay is well described by a single exponential.

Figure 13 shows the electric-field dependence of the diffraction efficiency \( \eta \) measured during the handling process at the grating spacing \( \Lambda = 2.96 \mu m \). During the recording and the beginning of the erasure process the electric field \( E_0 \) is on. After the grating compensation has reached saturation, one of the recording beams was switched off, and the erasure proceeded as shown in regions C and D in Fig. 1. The curve shown compare the decay rates for three values of the applied field \( E_0 \) at the grating spacing \( \Lambda = 2.96 \mu m \). The values for the decay time constants \( \tau_0 = 1/\gamma_0 \) are \( \tau_0(E_0 = 0) = 18 s \), \( \tau_0(E_0 = 1 kV/cm) = 13 s \), \( \tau_0(E_0 = 2 kV/cm) = 8.2 s \).

Figure 12 shows the decay dynamics of the gratings in region D for the grating spacing \( \Lambda = 18 \mu m \). During the recording and the beginning of the erasure process the electric field \( E_0 \) is on. After the grating compensation has reached saturation, one of the recording beams is switched off at time \( t = 5 s \). Later, the field is switched off (dashed curves) and on again (solid curves) several times in order to show the jumps in diffraction efficiency. The inset shows the same kind of measurement but for the grating spacing \( \Lambda = 0.7 \mu m \).

Figure 13 shows the possibility of modulating the hologram diffraction efficiency by applying a field \( E_0 \). It shows an erasure process with one Ar+ laser beam with intensity \( I = 40 mW/cm^2 \) at \( \Lambda = 18 \mu m \). The space-charge gratings were previously recorded with \( I_0 = 66 mW/cm^2 \) and an applied field \( E_0 = 2 kV/cm \). At the beginning of the erasure process the applied field is kept on (solid curve). After a while the field is periodically switched off (dashed curves) and on again (solid curves). When the field is removed, the diffraction efficiency \( \eta \) drops to a much lower value with a ratio \( \eta(E_0 \text{ off})/\eta(E_0 \text{ on}) \) of \( \approx 0.0015 \). When the field is switched on again, the diffraction efficiency recovers in large extent. The inset in Fig. 13 shows the same measurement but for a grating spacing \( \Lambda = 0.7 \mu m \) and an initially applied field \( E_0 = 1 kV/cm \). Again, at a certain point the field is switched off (dashed curve), with only a small change in diffraction efficiency and in the time constant \( \tau_0 \). Note also the big difference in the value of the diffraction efficiency, which is approximately 100 times bigger for \( \Lambda = 18 \mu m \) than for \( \Lambda = 0.7 \mu m \).

4. DISCUSSION

We discuss first the experiments presented in Section 3 for K\textsubscript{3}N\textsubscript{O\textsubscript{3}} in the framework of the theoretical model of Subsection 2.A.

The fit of the measurements to Eq. (21), shown in Fig. 11, indicates an effective number of traps \( N_{\text{eff}} = (0.75 \pm 0.15) \times 10^{15} \) cm\textsuperscript{-3} in our crystal. We can compare this value with the one we have obtained from room-temperature two-beam coupling measurements in the same crystal. There we found \( N_{\text{eff}} = (1.1 \pm 0.3) \times 10^{15} \) cm\textsuperscript{-3}, which is in good agreement with the above value. This shows that the effective number of traps in photorefractive materials can be also determined by purely dynamic measurements and not only by two-beam coupling experiments. It is also possible to determine \( N_{\text{eff}} \) from dynamics measurements in the absence of compensation by fitting the time rate \( \Gamma_0 \) to Eq. (20). However, in this case, three parameters, \( \Gamma_{\text{Str}} \), \( K_{\text{eff}} \), and \( K_{\text{ref}} \), have to be fitted simultaneously. In contrast, if the optimal experimental conditions are chosen in compensating crystals, the curve of Fig. 11 is influenced only by the values of the two parameters \( \Gamma_{\text{Str}} \) and \( K_{\text{ref}} \) [Eq. (25)].

The measurements of Fig. 12 confirm the increase of the time rate \( \text{Re}(\Gamma_0^*) \) under an applied field, as expected from the dependencies shown in Fig. 9. In the measurements shown in Fig. 12 there are no oscillations in the dynamics because the described Bragg-diffraction experiments are not sensitive to the relative phase between light fringes and refractive index grating. This is valid until the diffraction efficiency does not become so big that the space-charge grating is modified as a result of beam-coupling effects between incident and diffracted beams.24

Now we explain the observations shown in Fig. 13. We remarked in Section 2 that the electric field \( E_0 \) generates a phase shift between electronic and ionic grating. As a consequence, the total space-charge field \( E_{\text{t}} \) at saturation becomes larger, as shown in Fig. 2. Also, during the time evolution \( E_{\text{t}}(t) \) is larger for \( E_0 \neq 0 \) than for \( E_0 = 0 \). This is especially true for large \( \Lambda \), where both \( E_{\text{t}}(t) \) and \( E_{\text{r}}(t) \) can exceed \( E_{\text{t}}(t) \) by orders of magnitude. In the experiment presented in Fig. 13 the electronic and the ionic contributions to the space-charge field remain their relative phase shift of \( \pi \) predicted by Eqs. (12b) and (12c)
every time the field is switched off, that is, when \( E_1 \) decreases. This is the explanation for the observed drop of the diffraction efficiency \( \eta \). If the field is switched on again, the two components shift in phase again, giving rise to the new increase of \( \eta \). It is evident that, since only one beam illuminates the crystal during this measurement, the described effects cannot be understood without assuming the simultaneous presence of two complementary gratings. If in region D only the ionic charges would contribute to \( E_1 \) (i.e., \( E_{\text{int}} = 0, E_0 = 0 \)), the electric field could not influence the diffraction efficiency. In fact, for small grating spacings such a case is nearly realized. This is the reason for the small influence of the field in the measurement shown in the inset of Fig. 13.

In Subsection 2.A.2 we treated the intensity dependence of the time rate \( \Gamma_0 \), and we obtained relation (26). The parameters \( \Gamma_{\text{int}}, \Gamma_{\text{int}}, K_1, \) and \( K_\alpha \) appearing in relation (26) can all be determined independently, for instance by fitting the grating spacing dependencies of the measured time rates \( \Gamma_0 \) and \( \Gamma_1 \) to Eqs. (20) and (21). Therefore a measurement of the intensity dependence of the time rate \( \Gamma_0 \) gives information on \( \Gamma_0 \) and thus on the electron mobility \( \mu \) through relation (5d).

We discuss this with the help of a further experiment not presented in Section 3. In a KNbO\(_3\) crystal doped with lithium and sodium\(^{15}\) we observe a decrease of \( \Gamma_0 \) by \( \sim 30\% \) if the crystal is illuminated with an intensity of 250 mW/cm\(^2\) with respect to its value in the dark. According to relation (26), such a decrease is compatible with a mobility of the order of \( 10^{-4}\) cm\(^2\)/V s\(^{-1}\). Such values for \( \mu \) are much smaller than the ones previously reported in the literature for KNbO\(_3\), which are in the region 0.01-0.5 cm\(^2\)/V s\(^{-1}\).\(^{23,32,33}\) These values can be understood only if the mobility is trap limited\(^{36}\), i.e., limited by many trapping and reexcitation processes in shallow traps placed close to the conduction or valence band.\(^{34}\) It should be remarked that in the same crystal we also observed strongly nonexponential decays of the primary space-charge grating at room temperature. These dark decays were also dependent on the writing intensity and on the recent illumination history of the sample. As was shown by Strohhendl,\(^{35}\) such effects are expected for crystals containing a large number of shallow traps. The effect of such shallow trap levels is not explicitly included in our electron–ion model. However, without modifying it we may also consider that the electrons trapped in shallow traps contribute to the free charge concentration \( n_0 \) but that only the electrons in the conduction band are free to move. This leads to a decreased effective mobility \( \mu_{\text{eff}} = \mu(n_{\text{free}}/n_0) \), where \( n_{\text{free}} \) is the true electron concentration in the conduction band and \( \mu \) is the true conduction band mobility. As stated in Subsection 2.A.2, a large \( n_0 \) represents the necessary condition for the observation of the decrease of \( \Gamma_0 \) with light intensity. Thus our observations can be explained if \( n_{\text{free}} \ll n_0 \). All the above considerations clearly demonstrate the problems connected with the determination of the true mobility in crystals containing a number of different defect levels that can act as traps for the charge carriers. Such problems have been discussed in detail by Partanen et al.\(^{36}\) A method to overcome them may be represented by the investigation of the grating dynamics after excitation with short pulses in the picosecond range.\(^{34}\)

The behavior of KNbO\(_3\) crystals during high-temperature charge compensation could also be explained by the second model described in Subsection 2.B. In this case one has to assume that \( E_{\text{ph}} \) greatly exceeds \( E_{\text{st}} \), that is, the effective number of hole acceptors \( N_{\text{eff},h} = (N_{\text{h0}}/N_{\text{h0}}) (N_{\text{h0}} - N_{\text{h0}}^-) \) is again large compared with the effective number of electron acceptors \( N_{\text{eff},e} = (N_{\text{h0}}/N_{\text{h0}}) (N_{\text{h0}} - N_{\text{h0}}^-) \), similar to the assumption \( N_{\text{h0}} \gg N_{\text{eff}} \). However, in KNbO\(_3\) this electron–hole mechanism can be excluded owing to the fact that compensation takes place independently in crystals with different primary charge carriers.\(^{15}\)

The activation energy \( \Delta E = 1.0\) eV for the conductivity of the compensating charge carriers remains unchanged even if the type of photoconduction is changed in the same crystal sample from hole conduction to electron conduction by means of a reduction treatment.

One can conclude that in KNbO\(_3\) the 1-eV activation energy is associated to the ionic mobility \( \mu \); this is thermally activated by an Arrhenius-type process described by \( \mu(T) = \mu_0 \exp(-\Delta E/k_B T) \). At room temperature the value of the ionic mobility is so small that the charge-compensation process of Fig. 1 is not observed. As shown previously,\(^{15}\) the low ionic conductivity at room temperature permits us to fix holographic gratings by a temperature cycling method.

In general, with the ionic model the ion mobility \( \mu \) can be obtained from the dielectric relaxation rate \( \Gamma_{\text{int}} \) [Eq. (5f)] if the number of ions \( N_{\text{b}} \) is known. The exact determination of \( N_{\text{b}} \) was not possible in KNbO\(_3\) because this number exceeds the effective number of traps \( N_{\text{eff}} \) too greatly. In crystals for which the dynamics shows only a partial compensation, similar to the one shown in Fig. 6,\(^{3,19-21}\) the concentrations \( N_{\text{b}} \) (or \( N_{\text{eff},h} \) for the model of Subsection 2.B) can be determined by the fitting of the steady-state values of \( E_1 \) measured at large grating spacings for different values of the applied field \( E_0 \) to Eq. (8).

We have already mentioned that the applicability of the described models is not limited only to KNbO\(_3\) crystals. Observations similar to the one of Refs. 14 and 15 in other photorefractive materials have been reported recently. In Bi\(_{12}\)SiO\(_{20}\), charge compensation takes place at temperatures of \( \sim 260\)°C\(^{8}\) or at room temperature.\(^{16,18,20}\) Room-temperature charge compensation has also been observed in Bi\(_{12}\)TiO\(_{20}\).\(^{19}\) Recently, Kirillov and Feinberg reported on thermal hologram fixing in BaTiO\(_3\) at temperatures close to 100°C.\(^{21}\) Their observations are similar to our previous ones in KNbO\(_3\) and the ones by Arizmendi in Bi\(_{12}\)SiO\(_{20}\).\(^3\) They can be explained with the ionic model of Subsection 2.A. The cited room-temperature charge compensation in Bi\(_{12}\)SiO\(_{20}\) and Bi\(_{12}\)TiO\(_{20}\) may also be understood by the models of this paper or by an analogous model to the one of Subsection 2.B but with allowed photoexcitation of holes.\(^{26-31}\)

The key point to distinguish between our models and the one reported in Refs. 28–31 is the light intensity dependence of the time rate \( \Gamma_0 \) measured during the handling process of the hologram. While our models predict a weak dependence like the one shown in Fig. 7, the alternative model predicts a much stronger dependence because the rate \( \Gamma_{\text{int}} \) in Eq. (35) increases with light intensity. There is a second possibility, to exclude experimentally the electron–hole model with photoexcitation of both charge carriers. This is possible if in the steady state the mutually compensating
contributions to the grating are of equal magnitude and the diffraction efficiency drops to nearly zero as for KNB03. As indicated in Subsection 2.B, in this model such a case would only occur for $E_{pe} = E_{ph}$, a highly improbable accident.

We have already remarked that the distinction between the model of Subsection 2.A including moving ions and the model in Subsection 2.B with purely thermally excited holes as secondary charge carriers is less evident. The results for the steady state are the same, and thus the two models are indistinguishable in this respect. In the equations describing the dynamic behavior there is a slight difference given by the term $(1 + K^2/K^2_0)$ that appears in the denominators of Eqs. (34) and (35) for the time rates $\Gamma_2$ and $\Gamma_3$. If the dielectric rate $\Gamma_{die}$ for holes is small compared with $\Gamma_{die}$ and $K_0$ is not too large, this term produces a slight decrease of the decay rate $\Gamma_3$ for large spatial frequencies $K$. Such a decrease is not predicted in the ionic case [Eq. (21)]. However, for large hole-acceptor concentration $N_{accept}$ and small $\Gamma_{die}$ (small $\mu_4$ or small $p_0$), the wave vector $K_4$ given in Eq. (37) is more likely to be large, so that the observation of such a decrease of $\Gamma_3$ is difficult for experimentally accessible spatial frequencies $K$. Thus an ultimate determination of the microscopic mechanism can be gained only by alternative physical investigations.

We want now to draw some consequences from the presented theoretical models for quasi-permanent hologram storage in photorefractive crystals. Since the electron-hole model and the electron-ion model lead essentially to the same conclusions, we discuss only the latter case. We may consider the following thermal fixing scheme. The hologram is recorded at elevated temperature until the steady-state space-charge field $E_1$ given by Eq. (8) is reached. Afterward the recording beams are switched off, and the crystal is cooled down so fast that the grating is not erased during this process. This can be achieved if the crystal reaches a temperature where the ions are essentially frozen in a time shorter than the inverse time rate $(\Gamma_3)^{-1}$. At room temperature the ionic mobility is small, and the dielectric rate $\Gamma_{die}$ becomes much smaller than the corresponding rate for the electrons $\Gamma_{die}$. We use Eq. (25) to calculate the storage and handling time of the hologram at room temperature, which is given by $\tau = (\Gamma_3)^{-1}$. This time increases quadratically with the grating spacing $\Lambda$. Therefore we can get longer storage times by recording the hologram with large grating spacings. For KNB03, room-temperature storage times $\tau$ of more than 1 year can be expected for the grating spacing $\Lambda = 10 \mu m$. The use of such large grating spacing can apparently have two disadvantages. First, the observed diffraction efficiencies at room temperature are smaller than for smaller grating spacings. Second, besides the storage and handling time, the compensating time needed for recording the hologram at high-temperature also increases. Both of these problems can be overcome in the same way, that is, by applying an external field. The field $E_0$ increases the time rate Re($\Gamma_3^*)$ and the diffraction efficiency. Thus an electric revelation of the grating can be realized.

We now discuss the difference between handling and storage times of the hologram. Both of these times are given by the inverse of the time rate $\Gamma_3$ given in Eq. (21), but different values of the dielectric relaxation rate for the electrons $\Gamma_{die}$ should be used in the two cases. For the handling time we must use the value of $\Gamma_{die}$ under light illumination, while for the storage time we need the value in the dark. The ideal situation is presented by the case where $\Gamma_{die} = 0$ in the dark, which would mean no thermal excitation of electrons. In this case the storage time is infinite, bu the revealing of the grating (C) does not take place. Thus in this case the observation of the grating is possible only under light illuminating and the handling time depends on the illumination intensity, with $\Gamma_3$ first increasing linearly with intensity, reaching a maximum and then decreasing again in accordance with the curves with Fig. 7.

In the case of KNB03 the value of $\Gamma_{die}$ is also relatively large in the dark and lies in the region 0.1-100 s$^{-1}$, depending on crystal sample and temperature. At temperatures below 100°C the ionic dielectric rate $\Gamma_{die}$ is smaller than $\Gamma_{die}$, and the rate $\Gamma_3$ does not depend strongly on light intensity. Therefore the handling time is approximately equal to the storage time and can even become longer for high intensities (Fig. 7). This can be regarded as an advantage because the illuminating light does not contribute to a faster erasure of the hologram.

### 5. CONCLUSIONS

We have presented two theoretical models of the photorefractive effect when the electronic space-charge grating is compensated by secondary charge carriers that cannot be photoexcited. In the first model the secondary carriers are moving ionic defects. In the second they are thermally excited holes in the valence band. Approximate analytical solutions, as well as exact solutions in the modulation-approximation approximation, have been presented. The two models predict the same results if the number of compensating carriers is large. This limit is always valid in the ionic case, while in the case of hole conduction a reduced concentration of hole acceptors leads to a slightly different grating spacing dependence of the time rates describing the dynamics. The unusual dynamic behavior observed in KNB03 at elevated temperature and reported in Ref. 15 is exactly predicted by the ionic model. For KNB03 the electron-hole model cannot be applied because the compensation takes place independently in crystals with different primary charge carriers. However, both models are useful for explaining the experimental observations on quasi-permanent hologram storage reported in different photorefractive materials. Some implications of our models for the optimization of the fixing procedures and conditions with respect to grating spacing and applied electric field have been discussed.

The determination of a number of relevant photorefractive parameters such as the effective number of electron traps $N_{effect}$, the inverse diffusion length $K_0$, and the dielectric rates $\Gamma_{die}$ and $\Gamma_{die}$ for electrons and ions, respectively, is obtained by fitting the experimental results to the derived theoretical relations. Thus the use of the models presented for the investigation of the dynamic behavior during charge compensation is a powerful new method for gaining more insights into the mechanisms governing the photorefractive effect.
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REFERENCES