STUDY OF THE EQUIVALENT ELECTRON DRIFT FIELD CHARACTERISTICS IN LiNbO$_3$ BY PHASE HOLOGRAPHY

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STUDY OF THE EQUIVALENT ELECTRON DRIFT FIELD CHARACTERISTICS IN LiNbO$_3$ BY PHASE HOLOGRAPHY

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An analysis of the diffraction efficiency of photorefractive holograms in ferroelectric crystals is shown to provide a novel technique for determining the nature and magnitude of the equivalent electron drift field. For a Fe doped lithium niobate crystal (0.05% per mole) we find that the total field consists of an intensity independent internal field of 8.5 KV/cm and a photo-generated field given by the conversion factor $1.4 \times 10^6$ V-cm/W.
Electron drift has been shown to be the main mechanism for photorefractive volume phase hologram formation in electro-optic crystals.\textsuperscript{1,2} The nature and origin of the drift field has been the subject of considerable discussion. There exist in the literature two approaches: (i) the drift field is an intensity independent internal field due to the ferroelectric nature of the crystal,\textsuperscript{1} and (ii) the field is an effective, intensity dependent field produced by the bulk photovoltaic effect.\textsuperscript{3} In addition to the obvious physical interest, the relevance of such a discussion lies in that the sensitivity for hologram formation in an electro-optic crystal is largely determined by the drift field.\textsuperscript{1,2,4}

In this letter we report an independent and novel approach for analyzing the nature of the equivalent electron drift field. We show that the diffraction efficiency of a photorefractive hologram can be utilized as a sensitive test for determining the relative roles played by the two theories. The above mentioned two theories are based on the measurements of either the change of refractive index\textsuperscript{1} or the photocurrent\textsuperscript{3} due to spatially uniform illumination. Our approach utilizes a quantity routinely measured in holographic experiments and applies directly to the case of a spatially modulated light pattern. An advantage of this new scheme lies in its simplicity and hence easy applicability to other ferroelectric crystals.

The basic scheme of our approach is as follows. The coupled wave analysis\textsuperscript{5} of the diffraction efficiency $\eta$ shows that $\eta$ oscillates as a function of the coupling constant, which in turn depends on the writing time. The total number of oscillations is determined by the maximum value of the coupling strength, which is given by the asymptotic value (in time) of the space charge
field $\mathcal{E}$ or of the depth of the refractive index modulation. Regardless of the nature of the drift field, our nonlinear dynamic theory\textsuperscript{4} predicts that $\mathcal{E}$ saturates and reaches the value of the total drift field itself in a time scale determined by the light intensity (see Eqs. (5), (6) in Ref. 4). Thus if $\eta$ is measured for a given crystal with different writing intensities, three possible situations may ensue: (i) the drift field is internal and hence independent of the intensity, in which case $\eta$ should undergo the same number of oscillations but with different time scales; (ii) the field is an effective field due to the bulk photovoltaic effect and consequently intensity dependent, in which case the total number of oscillations is directly proportional to the intensity used; (iii) the field is a combination of the internal and photovoltaic components, in which case $\eta$ should undergo different number of oscillations but the number of oscillations is not in the same proportion as the corresponding intensities.

The photorefractive diffraction efficiency $\eta$ is measured in the usual way.\textsuperscript{6,7} The experimental parameters are: the writing half angle $\sim 6^\circ$, writing and reading wavelength 4880 Å, the crystal c-axis is in the plane of the writing beams, and the field polarization perpendicular to the writing plane. $\eta$ is measured by briefly interrupting the writing process at regular intervals of time. Beam coupling effects during the writing process are found to be negligible by continuously monitoring the intensities of the two beams. The crystal used for these measurement is a 1x1 cm$^2$ and 1 mm thick Fe:LiNbO$_3$ (0.05% mole) crystal with a linear attenuation coefficient, $\alpha = 12\text{ cm}^{-1}$ at 4880 Å.
In Fig. 1a we show the diffraction efficiency (as a function of writing time $t$) for four different intensities. The diffraction efficiency $\eta$ is defined as the ratio of the diffracted beam intensity to the initially transmitted read beam intensity. Note that each $\eta$-curve undergoes a remarkably similar pattern of oscillations in time, the main difference between curves being the time scale. This is a definite confirmation of our theory,\cite{4} which predicts that the space charge field evolves in time mainly determined by writing intensity. (See Eq. (5d) in Ref. 4.) This similarity in the oscillation pattern is seen more clearly in Fig. 1b, in which we plot the same $\eta$-curves in time scaled w.r.t. the intensity, i.e., exposure. Furthermore, it is evident from Figs. 1a and 1b that with increasing intensity $\eta$ goes through an increasingly large number of oscillations, implying a higher saturated $\mathcal{E}$ value or equivalent drift field (see Eq. (5) in Ref. 4). We emphasize that by measuring $\eta$ for a sufficiently long time, $\eta$ is, in fact, observed to reach a steady state value. This trend inevitably leads us to the conclusion that the equivalent drift field contains an intensity dependent component.

Next we describe an analysis of the experimental $\eta$-curves reported in Figs. 1a and 1b. $\eta$ as a function of writing time, $t$ and the crystal thickness that is scaled w.r.t. the attenuation constant, viz., $\xi = \alpha z$ is obtained by solving the coupled equations:

$$\frac{\partial r(\xi,t)}{\partial \xi} = -j\Gamma(\xi,t)s(\xi,t)$$

$$\frac{\partial s(\xi,t)}{\partial \xi} = -j\Gamma^*(\xi,t)r(\xi,t)$$

where the coupling constant
\[ \Gamma(\zeta, t) = (2\pi n_e^3 e_{33}/\alpha \lambda \cos \theta) A(\zeta, t) \exp[-j\varphi_g(\zeta, t)] \]  

(2)

is given in terms of the extraordinary index of refraction, \( n_e = 2.27 \), the electro-optic coefficient, \( e_{33} = 3.08 \times 10^{-11} \text{ m/V} \), the writing wavelength (\( \lambda \)) and half angle (\( \theta \)) and the space charge field, \( \mathcal{E} \). \( \mathcal{E} \) evolves nonlinearly in time and can be obtained from our theory as

\[ \mathcal{E}(\zeta, t) = A(\zeta, t) \cos[kx - \varphi_g(\zeta, t)] \]  

(3a)

\[ A(\zeta, t) = E(\zeta) [1 + e^{-2u} - 2e^{-u} \cos(au)]^{\frac{1}{2}} \]  

(3b)

\[ \varphi_g(\zeta, t) = \tan^{-1}\left( e^{-u} \sin(au)/[1 - e^{-u} \cos(au)] \right) \]  

(3c)

\[ u = (n_\mu g \tau / \epsilon) \left[ e^{-\zeta}/(1 + \alpha^2) \right], \quad \alpha = K \mu \tau G(\zeta) \]  

(3d)

where the equivalent drift field \( E(\zeta) \) depends on intensity \( I \) as

\[ E(\zeta) = E_{\text{int}} + \alpha I e^{-\zeta} \]  

(3e)

Here \( K, \kappa, g, \mu, \tau \) are, respectively, the grating wavevector, the photovoltaic conversion factor, electron generation rate, mobility, and trapping time. In Eq. (1), the attenuation of the incident reference (R) and signal (S) beams are accounted for by introducing \( r = R \exp(\frac{3}{2} \zeta), s = S \exp(\frac{3}{2} \zeta) \), both of which depend parametrically on time. The saturation time of the effective photovoltaic field is taken to be very short (\( \sim 10^{-3} \text{ sec} \)) compared with the reading or writing time. A nonessential simplification is introduced by neglecting the diffusion term in \( \mathcal{E} \), which is valid for an equivalent drift field much greater than \( DK/\mu \). Due to the high absorption of the crystal (30% transmission) one needs to include the \( \zeta \)-dependence of the quantities involved. The net total dependence of \( A(\zeta, t) \) on \( \zeta \) can be approximated by \( \exp^{-\zeta} \). The phase \( \varphi_g \) of the space charge field relative to the light modulation pattern decreases monotonically with \( \zeta \) and can be approximated by
\( \varphi_g(\zeta, t) = \varphi_0 - \varphi_1 \zeta \) \hfill (4)

Thus our analytical discussion of \( \eta \) is, apart from the parametric dependence of the quantities involved on time, reduced to the \( \eta \) considered by Uchida.\(^8\)

Upon expressing the Bessel functions appearing in Ref. 8, Eq. (6b), in terms of ascending series, we find

\[
\eta(t) = |C(\Gamma e^{-\zeta})S(\Gamma) - e^{j \varphi_1 \zeta} C(\Gamma)S(\Gamma e^{-\zeta})|^2 \hfill (5a)
\]

\[
C(z) = 1 + \sum_{k=1}^{\infty} \frac{(-)^z 2^k}{k!^2 (1+j \varphi_1)^{k+2} (1+j \varphi_1)^{-2}} \hfill (5b)
\]

\[
S(z) = \sum_{k=0}^{\infty} \frac{(-)^z 2^k}{k!^2 (1-j \varphi_1)^{k+2} (1-j \varphi_1)^{-2}} \hfill (5c)
\]

In Fig. 1c are plotted the theoretical curves for \( \eta \) based on Eqs. (2-5) as a function of time for the same intensities used in the experiment. The values of constants we use, in addition to those already mentioned are: \( \tau = 10^{-11} \) sec., \( \mu = 15 \times 10^{-4} \text{ m}^2/\text{V-sec.} \), and \( \varepsilon = 2.83 \times 10^{-10} \text{ F/m} \). No attempt has been made to curve fit the experimental data. However, the general agreement between the experimental (Fig. 1a) and analytically obtained (Fig. 1c) curves for \( \eta \) is very good, especially in terms of the intensity dependent time scale and number of oscillations. A better fit to the experimental \( \eta \)-curves can perhaps be obtained by including higher order dispersion of \( \varphi_g \) w.r.t. \( \zeta \). Effects on \( \eta \) of beam coupling in the writing process\(^9,10\), when they exist, generation of higher harmonics of \( \varepsilon \), nonuniform beam intensity across the beam diameter,\(^11\) and the change in the crystal temperature\(^12\) have been considered but the phase dispersion appears to be the dominant factor. In the limit where \( \varphi_1 = 0 \), \( \eta \) oscillates between 0 and 1 as in the theory by Kogelnik.\(^5\)
By measuring the number of oscillations in the η-curve for each intensity, one can find the corresponding saturated ε-value, i.e., the equivalent drift field. (See Eqs. (2) and (3a)-(3e).) In Fig. 2 the field is plotted as a function of intensity. The field $E$ increases linearly with intensity for the range of intensities used. The photovoltaic conversion factor $\kappa$ and the internal field $E_{\text{int}}$, as determined from the slope and intercept respectively, are found to be

$$E_{\text{int}} = 8.5 \text{ KV/cm}, \kappa = 1.4 \times 10^6 \text{ V-cm/W}.$$ 

In conclusion, we have shown that (1) the equivalent drift field has both an intensity dependent and intensity independent component, (2) the photovoltaic conversion factor is consistent with the values reported in Ref. 3 using an entirely different technique, (3) the fact that the η-curves invariably reach a final steady state value is a confirmation of our nonlinear dynamic theory\(^4\) for photorefractive phase hologram formation in that $\varepsilon$ reaches an asymptotic value, (4) also the fact that the η-curves have essentially the same pattern of oscillations verifies a prediction of our theory\(^4\), viz., $\varepsilon$ evolves in a time determined by the writing intensity.
REFERENCES AND FOOTNOTES


FIGURE CAPTIONS

FIG. 1. (a) Experimental curves of diffraction efficiency $\eta$ as a function of writing time for four different intensities: $1_1 = 21_2 = 41_3 = 81_4 = 12 \text{ mW/cm}^2$.

(b) The same $\eta$-curves plotted as a function of exposure ($W = 1t$).

(c) Theoretical diffraction efficiency curves plotted as a function of time for the four intensities used in (a). The phase dispersion factor $\varphi_1$ is taken to be 1.1.

FIG. 2. The equivalent drift field plotted as a function of the intensity and the corresponding number of oscillations in the diffraction efficiency curves. The slope is the photovoltaic conversion factor, $\kappa$ and the intercept the internal field, $E_{\text{int}}$. The slight ambiguity involved in judging the number of oscillations is accounted for by inserting error bars.
NUMBER OF OSCILLATIONS IN $\eta$-CURVE

SLOPE = $1.41 \times 10^6$ V-cm/W
INTERCEPT = 8.5 kV/cm