The recording of static holograms in photorefractive crystals by moving interference patterns

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The mechanism for the recording of a static hologram by a moving interference pattern in a photorefractive crystal has been studied theoretically and experimentally. The recording is carried out in an alternating external field whose frequency is equal to the difference between the frequencies of the writing laser beams. Analytic expressions are derived for the amplitude of the space-charge-field grating and for the time required to record the hologram. Experiments were carried out on a $Bi_{12}TiO_{20}$ crystal. The effect of the self-diffraction of the writing beams on the writing of the hologram has been found to be different from that corresponding to known mechanisms.

1. INTRODUCTION

Mechanisms involving a separation of space charge are used to write phase holograms in photorefractive crystals. These mechanisms are based on a drift of photoelectrons in an external or internal photovoltaic field and also on diffusion.[1,2] A variation of the refractive index, $\delta n = -(n^3r/2)E_{sc}$, arises as a result of the linear electrooptic effect. Here n is the refractive index, r is the electrooptic coefficient, and E_{sc} is the field of the nonuniformly distributed space charge.

The efficiency of these recording mechanisms is reduced substantially if the frequency difference between the signal beam and the reference beam, Ω, exceeds the reciprocal of the hologram recording time τ_{sc}^{-1}, which is determined primarily by the Maxwellian relaxation time $\tau_M = \varepsilon\varepsilon_0/\sigma$. Here σ is the conductivity induced in the crystal by the light, ε is the permittivity of the medium, and ε_0 is the permittivity of free space. The efficiency is lowered because there is not enough time for the hologram to be rewritten in the new position, as a result of the motion of the interference pattern formed by the waves of different frequency. An exceptional case is that of a drift mechanism in a static external field E_0, in which case the period of the grating hologram, Λ, is shorter than the electron drift length $E_0\mu\tau$, where μ is the mobility of the photoelectrons, and τ is their lifetime in the conduction band. In this case the hologram which is written is a moving hologram,[3] and the efficiency of the recording process increasing in a resonant fashion when the velocity of the grating hologram and that of the writing interference pattern become equal.

In this paper we are reporting a theoretical and experimental study of the mechanism for the writing of a static hologram by a moving interference pattern (by beams of different frequencies). This mechanism, which operates as the result of a drift of photelectrons in an alternating external electric field. This mechanism, operates in crystals with an arbitrary drift length, was first demonstrated experimentally in Ref. 4, for the particular case of the photorefractive $Bi_{12}TiO_{20}$ crystal. At a qualitative level, the writing mechanism can be explained most clearly in the case in which the drift length is short in comparison with the grating period Λ. At the time of the maximum external field, the photoelectrons generated at the maxima of the interference pattern are retrapped by trapping centers on the left-hand slopes of the pattern (Fig. 1a). After a quarter of a period, a maximum in the interference pattern coincides with a maximum of the density of the retrapped electrons. If the field frequency Ω is equal to the difference between the frequencies of the writing beams, the field vanishes at this time, preventing erasure (Fig. 1b). After another quarter of a period, the field changes sign, and the electrons accumulate on the right-hand slopes (Fig. 1c), thereby increasing the separation of space charge created in the initial stage of the process. After yet another quarter of a period (Fig. 1d), the field crosses zero again, preventing an erasure of the existing grating of space charge density. The process then repeats itself. The moving interference pattern thus forms a fixed distribution of the electric field of the space charge in an alternating field, so it produces a fixed-phase-grating hologram by virtue of the electrooptic effect.

If the drift lengths are large, the photoelectrons are retrapped uniformly over the volume of the crystal. As a result

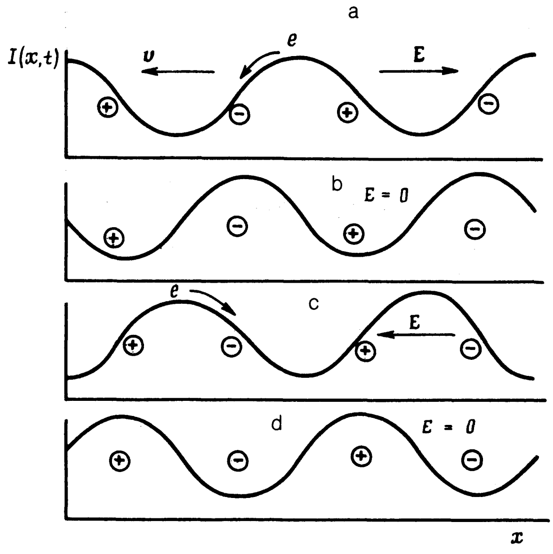

FIG. 1. Qualitative mechanism for the writing of a static hologram by an interference pattern moving at a velocity $\mathbf{v} = (\Omega/q^2)\mathbf{q}$.

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of the succeeding exposures to the maxima of the interference pattern and of the drift in the external field, however, the only electrons which survive are those which are trapped on the left-hand slopes of the interference pattern (we mean the position of the pattern at zero time). The efficiency of the writing is obviously lower in this case than when the drift length is short. In addition, the writing time increases under otherwise equal conditions. The reason is that in this case an electron goes through many events of photoionization and trapping by trapping centers before it reaches the necessary position.

2. THEORY

For a quantitative analysis of the writing process, we adopt the most popular model of a photorefractive crystal. According to this model, there is one donor level, with a density N_D, in the band gap of the crystal. There is a partial compensation for this level by virtue of the release of an electron to an acceptor level with a density $N_A \ll N_D$. Only the donor levels participate in the writing process. Working from the discussion above, and ignoring the dark conductivity, we start with the following system of equations to describe the writing process [these are the kinetic equation for the generation of photoelectrons, Ohm's law (the diffusion current is taken into account), the continuity equation, the Poisson equation, and a balance equation for the number of particles]:

$$\begin{aligned} &\partial n_e/\partial t-\nabla j/e=sIN-\gamma n_e N^+,\\ &j=e\mu n_e E+k_B T\mu\nabla n_e,\\ &\partial(n_e-N^+)/\partial t=\nabla j/e,\\ &\nabla E=-e(n_e+N_A-N^+)/\varepsilon\varepsilon_0,\\ &N_D=N+N^+. \end{aligned} \quad (1)$$

Here n_e, N^+, and N are the densities of respectively electrons, ionized donors, and non-ionized donors; I is the light intensity; s and γ are the constants of photoionization and electron-ion recombination, respectively; E is the electric field; e is the electron charge; k_B is Boltzmann's constant; T is the temperature; and j is the electron current. The hole current and the photovoltaic current are not considered in (1). This case corresponds to the actual situation for most photorefractive crystals, with the exception of $LiNbO_3$:Fe (Ref. 1).

Eliminating N, N^+, and j, we reduce system of Eqs. (1) to a system of two nonlinear equations for n_e and E. Here we are using

$$\begin{aligned} &I=I_0\left(1+\frac{m}{2}e^{iqx}+\text{c.c.}\right),\\ &n_e=n_0+{}^1/_2 n_1 e^{iqx}+\text{c.c.},\\ &E=E_0\cos\Omega t+{}^1/_2 E_{sc}e^{iqx}+\text{c.c.}, \end{aligned}$$

where "c.c." means the complex conjugate, I_0 is the average intensity, $m=m_0 e^{-\Omega t}$ is the contrast of the moving interference pattern, E_0 is the amplitude of the alternating external field, Ω is its frequency, $E_{sc}\ll E_0$ is the amplitude of the space charge field, n_0 is the average density of photoelectrons, $n_1\ll n_0$ is the amplitude of the photoelectron-density grating, and q is the spatial frequency of the grating. Under the condition $m_0\ll 1$, the equations linearize and reduce to a single second-order differential equation for E_{sc}. Under the condition

$$\tau_M=\varepsilon\varepsilon_0/(e\mu n_0)\gg\tau=(\gamma N_A)^{-1}$$

we can ignore the term $d^2E_{sc}/dt^2\sim E_{sc}^{st}\tau_M^{-2}$; we then obtain a first-order equation for E_{sc}:

$$dE_{sc}/dt+\Gamma(t)E_{sc}=-m_0F(t), \quad (2)$$

where

$$\Gamma(t)=\left(1+\frac{E_D}{E_q}-\frac{iE_0\cos\Omega t}{E_q}+\frac{\Omega\tau\sin\Omega t}{\cos\Omega t+iE_D/E_0}\right)\Big/ \left[\tau_M\left(1+\frac{E_D}{E_\mu}-\frac{iE_0\cos\Omega t}{E_\mu}+\frac{\Omega t\sin\Omega t}{\cos\Omega t+iE_D/E_0}\right)\right], \quad (3)$$

$$F(t)=(E_0\cos^2\Omega t+iE_D\cos\Omega t)\Big/ \left[\tau_M\left(1+\frac{E_D}{E_\mu}-\frac{iE_0\cos\Omega t}{E_\mu}+\frac{\Omega\tau\sin\Omega t}{\cos\Omega t+iE_D/E_0}\right)\right]. \quad (4)$$

In expressions (3) and (4), $E_D=qk_BT/e$ is the so-called diffusion field, $E_q=eN_A/(\varepsilon\varepsilon_0 q)$ is the space-charge saturation field,[1,2] and $E_\mu=(q\tau\mu)^{-1}$ is the drift field. It is clear on physical grounds that under the condition $E_0>E_\mu$ the electron drift length is greater than $\Lambda/(2\pi)$. Equation (2) differs from Kuchtarev's equation[5] in the term in (3) and (4) with contains the dimensionless parameter $\Omega\tau$. If $\Omega\ll(E_D/E_0)\tau^{-1}$, this term can be ignored. Under the conditions $q>60\ \text{cm}^{-1}$, $E_0<10^4$ V/cm, and $\tau<10^{-8}$ s, this inequality holds for frequencies $\Omega<10^4\ \text{s}^{-1}$. At $\Omega\gg\tau_{sc}^{-1}$ we can take an average over the field period $2\pi\Omega^{-1}$ in (2), ignoring the rapidly oscillating part of E_{sc} in the process. This rapidly processing part is on the order of $(\Omega\tau_{sc})^{-1}$. As a result Eq. (2) becomes

$$\frac{dE_{sc}}{dt}+\bar\Gamma E_{sc}=-m_0\bar F, \quad (5)$$

where E_{sc} is the slowly varying amplitude of the grating of the space charge field, and $\bar\Gamma$ and $\bar F$ are the average values of $\Gamma(t)$ and $F(t)$. Taking an average over the period $2\pi\Omega^{-1}$, we find the following expressions for the steady-state value of the amplitude E_{sc} and for the relaxation time:

$$E_{sc}^{st}=-m_0\frac{\bar F}{\bar\Gamma}=-m_0\frac{E_qE_\mu\{[(E_\mu+E_D)^2+E_0^2]^{1/2}-E_\mu-E_D\}}{E_0\{[(E_\mu+E_D)^2+E_0^2]^{1/2}+E_q-E_\mu\}} \quad (6)$$

$$\tau_{sc}=\bar\Gamma^{-1}=\frac{\tau_M E_q[(E_\mu+E_D)^2+E_0^2]^{1/2}}{E_\mu\{[(E_\mu+E_D)^2+E_0^2]^{1/2}+E_q-E_\mu\}}. \quad (7)$$

The negative sign of E_{sc}^{st} means that the E_{sc} grating is out of phase with the interference pattern at zero time, in agreement with the qualitative analysis above (Fig. 1).

Expression (6) has two limiting cases. In the first, which corresponds to the inequality

$$E_0\lesssim E_\mu+E_D, \quad (8)$$

we have

$$E^{st}\approx-\frac{m_0E_0}{2}\left[1+\frac{E_D(E_\mu+E_q)}{E_\mu E_q}+\frac{E_0^2}{2E_\mu E_q}+\frac{E_D^2}{E_\mu E_q}\right]^{-1}. \quad (9)$$

Estimates based on the values of $\mu\tau$ in the literature[2] show that condition (8) holds for photorefractive crystals with high values of $\mu\tau$ (sillenites and semiconductors) under the condition $E_0 \lesssim 10$ kV/cm at small values of q, with $E_0 < E_\mu$, and also at large values of q, with $E_0 < E_D$. At small values of q we find

$$E_{sc}^{st} \approx -m_0 E_0/2.$$

This result is analogous to that for writing in a static external field.[5] The reason for the factor of 1/2 is that the effective writing field is weaker than the amplitude of the alternating field by a factor of 2.

For photorefractive crystals with small values of $\mu\tau$ (photoelectrics), condition (8) holds over the entire q range which can be achieved by means of waves in the optical range. In this case, for small values of q, we have $E_{sc}^{st} \approx -m_0 E_0/2$ again. As q increases, the amplitude decreases, because of the terms proportional to q^2 and q^4 in the denominator in (9). The physical cause of the decrease in E_{sc}^{st} is a saturation of the trapping centers or a diffusive transport of photoelectrons over distances greater than Λ. The relative contributions of these effects depend on the specific values of E_μ, E_D, E_0, and E_q, and they differ for different ferroelectric photorefractive crystals. The same factors lead to a decrease in E_{sc}^{st} in photorefractive crystals with high values of $\mu\tau$ at large values of q, at which we have $E_0 < E_D$, so condition (8) holds.

The second of these two limiting cases corresponds to the inequality

$$E_0 \gg E_\mu + E_D, \tag{10}$$

in which case we have

$$E_{sc}^{st} = -m_0 \frac{E_\mu E_q}{E_0 + E_q} = \begin{cases} -m_0 E_\mu, & E_0 < E_q, \quad (11a) \\ -m_0 \dfrac{E_\mu E_q}{E_0}, & E_0 > E_q. \quad (11b) \end{cases}$$

Estimates show that condition (10) holds for $E_0 \lesssim 10$ kV/cm for photorefractive crystals with high values of $\mu\tau$ (sillenites and semiconductors at intermediate values of q). In contrast with $E_{sc} \approx -m_0 E_0/2$ (as $q \to 0$), the grating amplitude initially falls off as q^{-1} [see (11a)]. The reason is that the drift length is greater than the grating period, and the electrons are not captured by the trapping centers on the left-hand slopes of the interference pattern (we mean the position of this pattern at zero time). With a further increase in q, the decrease in E_{sc}^{st} is accentuated by the saturation of the trapping centers [see (11b)]. In this case we have $E_{sc}^{st} \propto q^{-2}$.

Summarizing this analysis of limiting cases, we repeat that the first case generally holds for arbitrary q for ferroelectrics. For sillenites and semiconductors, the two values of q which separate the different limiting cases are found from the condition $E_0 = E_\mu + E_D$. For all photorefractive crystals, without exception, we have $E_{sc}^{st} \to -m_0 E_0/2$ as $q \to 0$. In photorefractive crystals with a small value of the parameter $\mu\tau$, the decrease in E_{sc}^{st} occurs at larger values of q than in photorefractive crystals with high values of $\mu\tau$ (under otherwise equal conditions). It thus becomes possible to predict that these crystals will have a greater bandwidth (in the spatial frequency of the grating) for the writing of holograms by the mechanism which we are discussing here.

For the time required to write the hologram we have, in the first of these limiting cases,

$$\tau_{sc} \approx \tau_M \frac{1 + E_D/E_\mu}{1 + E_D/E_q}. \tag{12}$$

It can be seen from (12) that in this case the writing time does not depend on the amplitude of the external field, E_0. The behavior of the time τ_{sc} as a function of q is the same as that of the writing time for the case of the diffusion mechanism in the absence of an external field.[2]

In the second limiting case, we have the following expression for the writing time:

$$\tau_{sc} = \tau_M \frac{E_0 E_q}{(E_0 + E_q) E_\mu} = \begin{cases} \tau_M E_0/E_\mu, & E_0 \ll E_q, \quad (13a) \\ \tau_M E_q/E_\mu, & E_0 \gg E_q. \quad (13b) \end{cases}$$

In deriving (13) we made use of the condition $E_\mu \ll E_q$, which holds for photorefractive crystals with high values of $\mu\tau$. As we mentioned earlier, it is for photorefractive crystals of this sort that the second of our limiting cases holds. In this case, τ_{sc} increases linearly with E_0 or q, up to a limiting value $\tau_M E_q/E_\mu \gg \tau_M$. The physical reason for the increase in τ_{sc} is (as mentioned above) that a photoelectron will undergo many events of photoionization and drift transport before it reaches the necessary position, which is determined by the position of the static hologram. Expression (13) is analogous to that in the case of writing by single-frequency beams in an alternating field.[2]

As a result of diffraction by the written hologram, the reference wave, of frequency $\omega + \Omega$, makes a contribution at this frequency to the signal wave, which has a frequency ω. Since the diffractive part of the reference wave also interacts with the signal wave, the latter will have an intensity which oscillates in time after it passes through the crystal. This result could be interpreted in a different way. As a result of the motion of the interference pattern, the phase shift between this pattern and the grating hologram takes on values from 0 to 2π. We know[1,2] that in this case there are changes in the direction and the magnitude of the energy exchange between the interfering beams, as a result of the interaction at the grating hologram. During the writing of a static hologram in an alternating field by beams of different frequencies, the energy exchange between these beams is therefore a time-varying process. An analysis shows that the phase shift between the oscillations of the intensity of the signal beam and the alternating voltage applied to the crystal is $\pm \pi/2$, depending on the sign of the electrooptic effect.

3. EXPERIMENTAL PROCEDURE AND RESULTS

We have carried out an experimental study of the mechanism for the writing of a grating by beams of different frequency in the cubic photorefractive crystal $Bi_{12}TiO_{20}$ (BTO). The experimental layout is shown in Fig. 2a. A 2-mW He–Ne laser ($\lambda = 0.63$ μm) was used. The signal beam was tapped off with the help of a beam splitter 5. The frequency of the reference wave, I_0, was shifted by a piezoelectric mirror 4, to which a sawtooth voltage was applied. The frequency shift, $\Omega(2\pi)^{-1} = 50$ Hz, was determined by the ac line and was monitored with the help of an auxiliary Michelson interferometer, which consisted of parts 1–4 and photodetector D 1. The intensity ratio of the signal wave and the reference wave, β, was varied over a wide range with the help of filters F. The beam polarizations were set by polarizers P. Figure 2b shows the orientation of the crystal. An alternating external voltage with a frequency of 50 Hz was

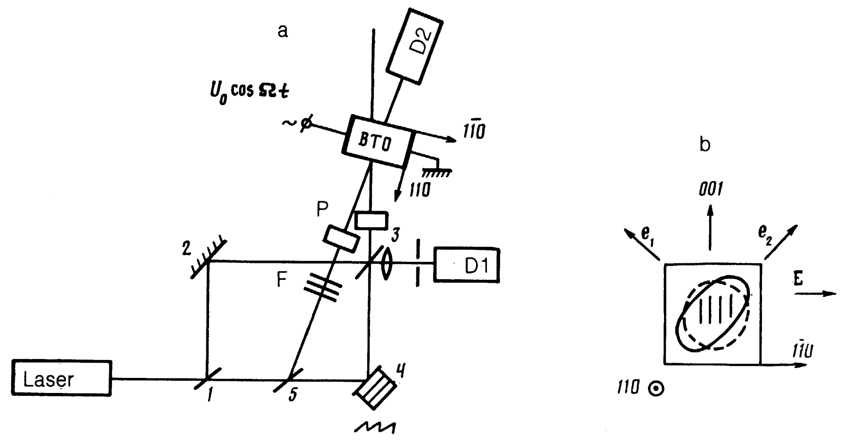

FIG. 2. a—Optical layout of the experiments; b—deformation of the index ellipsoid of a cubic photorefractive crystal in an external field **E**. Here $\mathbf{e}_1$ and $\mathbf{e}_2$ are the polarizations of the natural waves if the optical activity is ignored.

applied to the $1\bar{1}0$ faces and could be varied up to $U_0 = 5.5$ kV. The thickness of the BTO crystal was $d = 4.5$ mm, and the distance between the electrodes was $L = 5.6$ mm. The intensity of the signal wave behind the crystal was measured with photodetector $D2$. From the amplitude of the oscillations in the signal from $D2$, we determined the diffraction efficiency of the hologram, η, with the help of the formula

$$I_c = |E_{c0} \pm \eta^{1/2} E_0|^2,$$

where I_c is the intensity of the signal wave at the exit from the crystal, E_{c0} is the amplitude of this wave at the entrance to the crystal, E_0 is the amplitude of the reference wave at the entrance, and $\beta = E_{c0}^2/E_0^2$. The plus sign corresponds to a maximum of I_c, and the minus sign to a minimum.

The polarization vectors of the incident waves made an angle of $+30°$ or $-60°$ with the plane of incidence. In these cases, the beams which have passed through half the thickness of the crystal have polarization vectors directed at $\pm 45°$ with respect to the plane of incidence. The waves polarized in this fashion would be the natural waves for the geometry of these experiments in an optically inactive cubic crystal. The electrooptic effects for these waves are identical in magnitude but opposite in sign[6] (Fig. 2b). The rotation of the polarization plane during propagation through the crystal was caused by the optical activity of BTO and amounted to 6 deg/mm (Ref. 2). For this choice of polarization directions, the energy exchange between the interacting beams is therefore maximized.

Figure 3 shows the experimental results on $\eta^{1/2}$ as a function of the amplitude of the external field, $E_0 = U_0/L$, and the spatial frequency of the grating, q. For diffraction by a thick hologram[1] we would have

$$\eta^{1/2} \approx (\omega/c) n^3 r E_{sc} d/2, \tag{14}$$

where $r = 5.8$ pm/V (Ref. 2) is the electrooptic coefficient of BTO. These results agree reasonably well with the results of the theoretical analysis above. At small values of q, the quantity $\eta^{1/2} \propto E_{sc}$ does not depend on q, in agreement with (9). As q increases, it falls off in accordance with (11a) and (11b), since BTO has a relatively high value of $\mu\tau$, and at $E_0 \approx 10$ kV/cm the drift length becomes greater than the grating period even at $\Lambda < 10\ \mu$m (Ref. 7). We believe that the difference between the values of $\eta^{1/2}$ for the different polarizations is a consequence of self-diffraction, which was

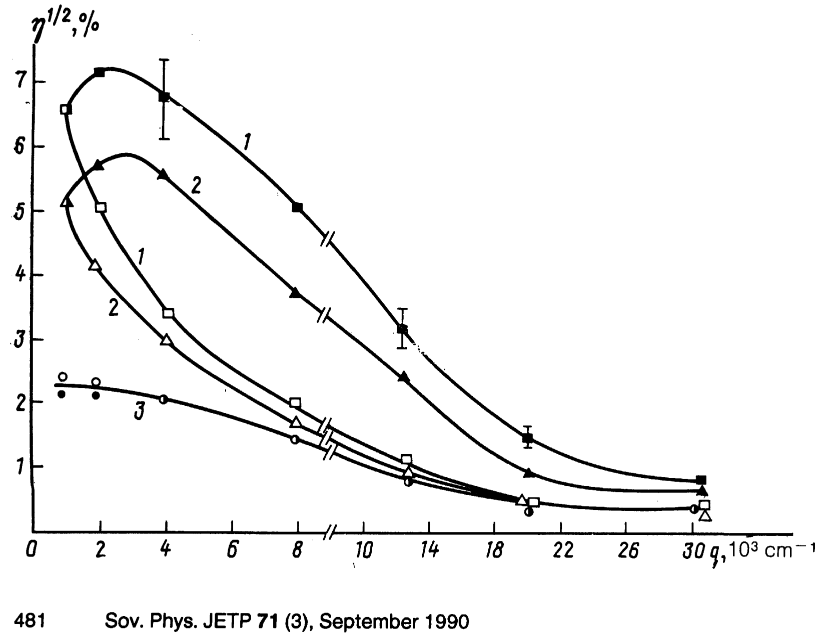

FIG. 3. Experimental results on the diffraction efficiency η as a function of the spatial frequency of the grating, q, for various amplitudes of the external field E_0 (kV/cm): 1—$E_0 = 9.6$; 2—7.9; 3—3.5. ●, ▲, ■) The polarization vector makes an angle of $+30°$ with the plane of incidence; ○, △, □) an angle of $-60°$. Here $\beta = E_{c0}^2/E_0^2 = 10^{-2}$.

ignored in the theoretical analysis. During diffraction by a static hologram, the reference wave generates a secondary wave at the frequency $\omega + \Omega$ which is collinear with the signal wave. This wave, interacting with the reference wave, writes a grating hologram in the alternating field by a time-varying mechanism which operates under the condition $E_0 > E_\mu$ (Ref. 2).

Analysis shows that the difference between the signs of the electrooptic effect (Fig. 2b) in the case in which the polarization vector of the interacting beams makes an angle of 30° with the plane of incidence causes this secondary grating to amplify the primary grating, recorded by the beams of different frequencies. In the case of an angle of $-60°$, on the other hand, the secondary grating weakens the primary grating. In other words, it is in phase or out of phase, respectively, with the primary grating. The efficiency of the writing of this secondary grating falls off at small and large values of q (Refs. 2 and 7), explaining why the values of $\eta^{1/2}$ for the different polarizations move closer together at small and large values of q. The value calculated for $\eta^{1/2}$ from (9) for small values of q is 11% with $\beta = 10^{-2}$ and $E_0 = 9.6$ kV/cm and agrees reasonably well with the experimental value of 7% (Fig. 3). A correct theoretical description of the writing mechanism at intermediate values of q will require consideration of the secondary grating which is written as a result of the self-diffraction.

The maximum diffraction efficiency, $\eta = 6\%$, was achieved at $m = 0.9$, $U_0 = 5.5$ kV, and $\Lambda = 20$ μm. The phase difference between the intensity oscillations of the signal wave behind the crystal and the external voltage is $\pm(90° \pm 10°)$. The choice of sign here depends on the sign of the electrooptic effect, which is in turn determined by the polarization of the beams (Fig. 2b). This result agrees with the qualitative arguments above.

The time required to write the holograms at the power used was 1–10 s.

4. DISCUSSION

The process described above is one case of a process with $\langle E^3 \rangle \neq 0$, where the angle brackets mean a time average.[8] Physically, this process is the flow of a photoconductivity current $j \propto E\,|E_l|^2 + \text{c.c.}$, where E is the low-frequency electric field, and $E_l = E_1 + E_2$ is the resultant field of the light wave, which determines the photoconductivity of the crystal. The current j has a term

$$\propto (EE_1E_2^* + \text{c.c.}) \propto (e^{-i\Omega t}e^{-i\omega t}e^{i(\omega+\Omega)t} + \text{c.c.}),$$

which does not have a zero time average but which is spatially nonuniform $\{\propto \exp[i(\mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}]\}$. This component of the current is the reason why a static hologram can be written.

Trofimov and Stepanov[9] have studied the flow of a time-varying, spatially uniform photoconductivity current which arises when a static grating E_{sc}, written by single-frequency beams, is exposed to a moving interference pattern. This case is the opposite of that described above.

Three waves take part in the writing of the primary static hologram: two light waves, $E_1 \exp[i(\mathbf{k}_1\mathbf{r} - \omega t)]$ and $E_2 \exp i[\mathbf{k}_2\mathbf{r} - (\omega + \Omega)t]$, and one low-frequency wave, $E_0 \exp(-i\Omega t)$, i.e., the external field. We call the latter a "wave with a known degree of conditionality." During diffraction of the writing beams by the static hologram, two new light waves arise: $E_1' \exp\{i[\mathbf{k}_1\mathbf{r} - (\omega + \Omega)t]\}$ and $E_2' \exp(i(\mathbf{k}_2\mathbf{r} - \omega t))$. These waves satisfy the Bragg condition $\mathbf{k}_1 - \mathbf{k}_2 = \mathbf{q}$. As a result, this process could with equal justification be called a "three-wave" or "five-wave" mixing.

In our opinion, the primary advantage of this mechanism for the interaction of light waves in a photorefractive crystal is the presence of a time-varying energy exchange, which occurs even at small beam interaction angles. The frequency of the oscillations in the energy exchange is the same as the frequency of the external field. This effect might find applications in the interferometry of vibrating objects. As was mentioned above, crystals with low values of $\mu\tau$ would appear to be the best choice for the writing of holograms by this mechanism, because such crystals would be expected to have a large bandwidth in terms of the spatial frequency of the grating. In our opinion, the mechanism of the writing of a static hologram by beams differing in frequency deserves a detailed experimental study in various photorefractive crystals.

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