11-1998

Self-Enhancement of Dynamic Gratings in Photogalvanic Crystals

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(Received 1 October 1997)

We have developed a compact closed-form solution of the band transport model for high-contrast gratings in photogalvanic crystals. Our solution predicts the effect of the photoconductivity and the electric field grating enhancement due to the photogalvanic effect. We predict a pronounced dependence of the steady-state photogalvanic current on the contrast of the interference pattern and an increase of holographic storage time due to the enhancement of the photoconductivity grating contrast. In the high contrast limit and a large photogalvanic effect the refractive index grating will be shifted from the position of the intensity modulation pattern, contrary to the usually adopted model of unshifted gratings. [S1050-2947(98)03610-5]

PACS number(s): 42.65.Hw, 42.70.Nq

I. INTRODUCTION

The popular photorefractive model [1] known as the band transport model for recording of dynamic holographic gratings was originally solved for a small contrast of the interference light pattern. The small contrast approximation was very successful in description and prediction of many interesting effects such as energy transfer [2], anisotropic diffraction [3], phase conjugation, and holographic storage. However, a growing number of new experiments with high contrast interference patterns reveal the necessity for further development of the model for photorefractivity in order to describe high contrast effects. Several attempts have been made to expand the theory to high modulation using both analytical [4] and numerical approaches [5]. In this paper we expand our new compact analytical solution [8,12] to encompass the problem of high contrast gratings *including the photogalvanic current* [13].

Our solution addresses the problems of holographic current [6] and of photoconductivity modified by dynamic gratings [7], where the current is very sensitive to the degree of modulation of the photoconductivity grating. Photogalvanic current can give a major contribution to the grating recording in Fe-doped $LiNbO_3$ crystals [9] and is also pronounced in such materials as $BaTiO_3$ and $KNbO_3$ [10]. Photogalvanic effects were also measured in the cubic sillenite crystals $Bi_{12}XO_{20}$ ($X=$ Si, Ge,Ti) [11] by using an original modulation technique, which involves the detection of the alternating photogalvanic current with the change of polarization of incident light. In the first commercial applications of holographic storage for optical filters based on $LiNbO_3$:Fe, photogalvanic current was proven to be a major player in the grating recording with high contrast [14].

II. THEORETICAL MODEL

We shall start from the standard monopolar photorefractive model [1,3] with the equations for concentrations of photocarriers n, ionized N^+, and neutral N donors, and compensating centers N_A:

$$\dot{n}=\dot{N}^+-\frac{\nabla j}{e}, \tag{1}$$

$$\dot{N}=g(N-N^+)-rnN^+. \tag{2}$$

Here g is the generation rate $g=\beta+sI$, β is the thermal generation rate, s is the photon capture cross section, and $I=I_0(1+m\cos Kx)$ is the intensity pattern with contrast m. The expression for electric density current j,

$$j=e\mu nE-eD\nabla n+pI, \tag{3}$$

includes the usual conductivity ($\sim e\mu n$), diffusion ($\sim eD\nabla n$), and photogalvanic current, represented for simplicity by the main term as for $LiNbO_3$ along the C axis ($\sim pI$), with p being the effective Glass coefficient [13]. For determination of electrostatic field E we use the Poisson's equation

$$\nabla(\varepsilon E)=e(n+N_A-N^+). \tag{4}$$

Combining equations (1)–(4) we can get the expression for the total current density J:

$$J=j+\frac{\partial}{\partial t}(\varepsilon E), \tag{5}$$

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which is connected with external voltage V by Kirchhoff's law:

$$JRS + dE_0 = V, \tag{6}$$

where R is the external resistance, S is the area of contact, d is the distance between the contacts, and $E_0 = \langle E \rangle$ is the spatially averaged external electric field. These basic equations (1)–(6) describe the formation of the photoinduced grating of the electric field E, the concentration of photoinduced carriers n (or the grating of photoconductivity $\sigma = en\mu$), and also the ionized impurity grating N^+.

For the steady state assuming $N_A \gg n$, the system of equations (1)–(6) will be simplified to

$$N^+ = g(N - N_A)/(nr),$$

$$E = \frac{D}{\mu} \nabla \ln n + \frac{J}{e\mu n} - \frac{pI}{e\mu n}, \tag{7}$$

$$\nabla(\varepsilon E) = e(N_A - N^+).$$

Seeking the solution of the system (7) for $n(x)$ in the form $n = n_0[1 + a\cos(Kx + \varphi)]$ with free parameters n_0, a, φ in the one-dimensional case, we obtain

$$J = \sigma_0 \sqrt{1 - a^2}(E_0 - E_{\mathrm{pv}})$$

with

$$E_{\mathrm{pv}} = -\frac{pI_0}{\sigma_0}\left(\frac{\overline{m_c}}{a} + \frac{1 - m_c/a}{\sqrt{1 - a^2}}\right). \tag{8}$$

Here $\sigma_0 = e\mu n_0$ and $m_c = m\cos\varphi$. The justification for this form of solution is discussed in details in [12]. For short circuit conditions when $JRS \ll dE_0$,

$$E_0 = V/d. \tag{9}$$

The average field is constant and the conductivity grating influences the total electric current density J. We can see [8] that both the field grating E and the ion grating N^+ can be explicitly defined as functions of the carrier grating n. This means that the solution is dependent only on the parameters n_0, a, φ in the carrier distribution. These parameters we shall find from Poisson's equation, which after substitution of E and N^+ as a function of n takes the form

$$\lambda(\ln n)'' + \overline{\xi}\left(\frac{1}{1 + a\cos(u + \varphi)}\right)' + F\left(\frac{1 + m\cos u}{1 + a\cos(u + \varphi)}\right) = 1 - \frac{G_0}{n_0}\left(\frac{1 + \overline{m}\cos u}{1 + a\cos(u + \varphi)}\right), \tag{10}$$

where $\lambda = \varepsilon DK^2/e\mu N_A n_0 = E_D/E_Q$, $E_D = KD/\mu$ is the diffusion field, $E_Q = eN_A/\varepsilon_0\varepsilon K$ is the limiting space charge field, $\overline{\xi} = \varepsilon KJ/e^2\mu N_A n_0$ and $F = p\varepsilon KI_0/e^2\mu n_0 N_A = E^0_{\mathrm{pv}}/E_Q$, $\overline{m} \equiv m/(1 + \sigma_d/\sigma_p)$ and $\sigma_d/\sigma_p = \beta/sI_0$ is the ratio between dark and photoconductivities. We use the transformation $u = Kx$ for the spatial variable and use the symbol $(\,)'$ for the derivative with respect to u. Finally, $E^0_{\mathrm{pv}} = pI_0/\sigma_0$. After averaging Eq. (10) (charge neutrality conditions), and equating coefficients to the $\cos(u + \varphi)$ and $\sin(u + \varphi)$ terms, we have three equations:

$$\frac{n_0}{G_0} = \frac{\overline{m_c}}{a} + \frac{a - \overline{m_c}}{a\sqrt{1 - a^2}},$$

$$\lambda - \frac{Fm_s}{a} = \frac{G_0(\overline{m_c} - a)}{n_0 a\sqrt{1 - a^2}}, \tag{11}$$

$$\frac{\overline{\xi}}{\sqrt{1 - a^2}} + \frac{F(a - m_c)}{a\sqrt{1 - a^2}} = -\frac{G_0\overline{m_c}}{n_0 a},$$

where $G_0 = (\beta + sI_0)(N - N_A)/\gamma_R N_A$, $\overline{m_c} = \overline{m}\cos\varphi$. The system of equations (11) can be solved for two limiting cases of practical importance: for small and large photogalvanic field.

Let us start with small photogalvanic field. For a small photogalvanic field, $E^0_{\mathrm{pv}} \ll E_D\, E_Q$ we can obtain the following solution from the system of equations (11):

$$n_0 = \frac{G_0(1 + M^2\delta^2)}{(1 + \lambda)[1 - \delta\sqrt{1 + M^2(\delta^2 - 1)}]},$$

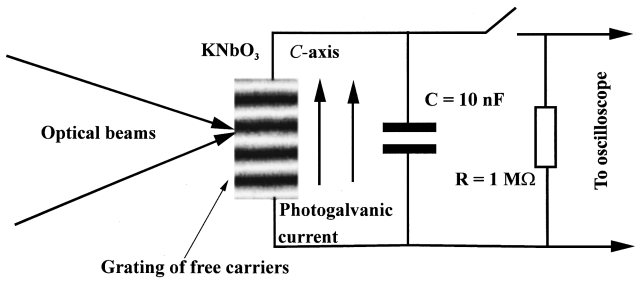
$$a = \frac{M}{(1 + M^2\delta^2)}\{1 - \delta\sqrt{1 + M^2(\delta^2 - 1)}\}, \tag{12}$$

where $\xi = E_0/E_Q$, $\delta = \lambda/(1 + \lambda)$, $M = \overline{m}(1 + \lambda)/\sqrt{\xi^2 + (1 + \lambda)^2}$, and $\tan\varphi = -\xi/(1 + \lambda)$. This solution is similar to that derived by us in [12] and shows that even the averaged concentration of photocarriers n_0 depends on contrast, grating period, and external field E_0. For the electric field we have the expression

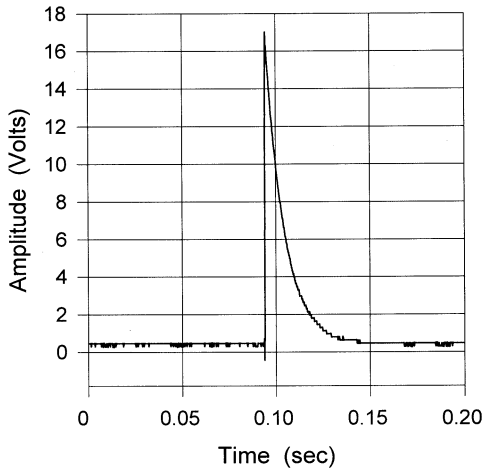
$$E = -E_D\frac{a\sin(u + \varphi)}{1 + a\cos(u + \varphi)} + \frac{\sqrt{1 - a^2}(E_0 - E_{\mathrm{pv}})}{1 + a\cos(u + \varphi)} + \frac{E_{\mathrm{pv}}(1 + m\cos u)}{1 + a\cos(u + \varphi)}, \tag{13}$$

where the two first terms correspond to diffusion and drift contribution and the third term is related to the photogalvanic current. It can be seen from Eq. (13) that the photogalvanic term will generate spatial harmonics of the electric field with a phase shift between harmonics dependent on the initial phase shift φ and the fundamental harmonics of $n(x)$ and $E(x)$. This means that due to the dependence of φ on E_0 and grating period (12) the phase shift between spatial harmonics of the electric field grating can be adjusted with the value of E_0 and the period of the interference pattern.

For the case of large photogalvanic field $E^0_{\mathrm{pv}} \gg E_D, E_0$ we get from Eq. (11) the solution for n_0, φ, and a by neglecting the dark photoconductivity:



(a)



(b)

FIG. 1. (a) Configuration for current measurements in photorefractive $KNbO_3$. (b) Typical trace of the discharge of capacitor after 10 min of illumination with light of intensity 30 mW/cm^2.

$$n_0=\frac{G_0 m\sqrt{1+F^2}}{a},$$

$$a=\frac{m\sqrt{1+F^2}}{1+F^2+m^2F^4}\{1+F^2\sqrt{1-m^2(1-F^2)}\}, \tag{14}$$

where $F=\tan\varphi=E_{\rm pv}^0/E_Q=pI_0/e\mu G_0E_Q$. In this limit the average concentration n_0 and the maximal value of conductivity grating modulation a grow with photogalvanic field $E_{\rm pv}$. The dependence of the conductivity grating contrast a on the interference pattern modulation m calculated from Eq. (14) is shown in Fig. 2 for three different values of F. It is interesting to note that in this limit of large photogalvanic field the contrast of the conductivity grating a can exceed the modulation of the interference pattern: from Eq. (14) it follows that for $F\gg 1$, m^{-1} we obtain

$$a=1-0.5(mF)^{-2}. \tag{15}$$

This contrast enhancement effect for the photogalvanic current will be pronounced in this limiting case. From the expressions (8), (15) we obtain for the current

$$J=\sigma_0E_QSm^{-1}, \tag{16}$$

where S is the area of the electrodes. This relation still holds also for small contrast (provided that $E_{\rm pv}^0$ is large enough $E_{\rm pv}^0\gg E_Qm^{-1}$). From Eq. (16) we can see that the current will be smaller than in a homogeneously illuminated crystal (with the same intensity) with a pronounced dependence on the contrast m.

This effect of contrast enhancement for the photogalvanic current may have an important practical application for holographic storage due to the dependence of storage time on photoconductivity. It is quite unusual to have contrast enhancement. We can compare it with the previous limiting case of small photogalvanic field, when $a\leqslant m$ is always the case due to the charge screening by diffusion and drift. The influence of the photoconductivity contrast on the holographic storage time may be explained by taking into account the fact that the storage time τ depends on the ionic conductivity [12]: $\tau\propto\sigma^{-1}$. When the ionic conductivity is spatially modulated with contrast m, then the storage time also will be spatially modulated within the range $\tau_{s0}\leqslant\tau_s\leqslant\tau_{s0}/(1-m_i)$ where τ_{s0} is the small-contrast (averaged) value of the storage time.

From the general solution for the photoinduced electric field (13) we can obtain the solution for the fundamental harmonic $E_1=E_{1c}\cos u+E_{1s}\sin u$ that determines beam coupling (self-diffraction) and diffraction efficiency: For the components E_{1c} and E_{1s} we can write

$$E_{1c}=T\left\{a(F-\xi)-F(Fm_s-\lambda a)+\frac{F(m_c-a)}{\sqrt{1-a^2}}\right\},$$

$$E_{1c}=T\left\{aF(F-\xi)+(Fm_s-\lambda a)+\frac{F^2(a-m_c)}{\sqrt{1-a^2}}\right\} \tag{17}$$

where $T=E_Q(1-\sqrt{1-a^2})/(a^2\sqrt{1+F^2})$. For large enough photogalvanic field, $E_{\rm pv}^0\gg E_Qm^{-1}$, E_D, we get from Eq. (17):

$$E_{1C}\cong -E_{\rm pv},$$

$$E_{1s}\approx mFE_{\rm pv}^0\approx m(E_{\rm pv}^0)^2E_Q^{-1}. \tag{18}$$

In this limit of high photogalvanic effect, the shifted component of the E field (E_{is}) responsible for the energy exchange will be proportional to the grating vector K. This means that small period gratings may be effectively recorded without being limited by the space-charge screening as in the case of the diffusion dominated mechanism of recording. We also want to underline that with increasing grating vector, the refractive index grating, will be shifted from the position of intensity modulation, contrary to the accepted model [1,2] based on the small contrast approximation. This shift will lead to energy exchange between writing beams and an absence of ''phase exchange'' that introduces instability in the recording process.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

To compare our compact solution for Eq. (14) with experiment, we performed a set of experiments to estimate the photogalvanic current in $KNbO_3$. The dimensions of the crystal were $5\times5\times6$ mm. To obtain uniform illumination of the face crystal surface an Ar-ion laser beam polarized in the plane of incidence at a wavelength of 488 nm passed through a telescopic system consisting of two lenses (3 and 30 cm,

respectively) and a pin hole at 15 mm placed between them. This configuration ensured a fairly uniform intensity distribution (plane wave) of output light beam. The beam was divided on a beam splitter on two separate coherent beams to create interference pattern of desired spacing through conventional two-wave-mixing configuration. Those plane-wave beams were incident on a sample of $KNbO_3$. The C axis of the sample was along the wave vector K of the grating. The interference pattern had a spacing 24 μm. This configuration guaranteed the maximum photogalvanic current along the C axis. The sample was placed between two electrodes connected to capacitor C of 10 nF. A sketch of the experiment is given in Fig. 1(a). The photogalvanic current flows along the C axis and charges the capacitor. After a fixed period of time T_0 (in our case usually 10–30 min) the capacitor discharges through a resistance of 1 MΩ, and the discharge trace is captured with a digital oscilloscope. The measuring procedure is similar to that performed in [15]. A typical discharge trace is shown in Fig. 1(b). The photogalvanic current is evaluated as: $J=CU_{\max}/T_0$, where $U_{\max}$ is the peak value of the discharge voltage. This simple configuration allows one to measure the photogalvanic current with a high degree of accuracy.

We studied the dependence of the current J on the modulation of the interference pattern $m=2\sqrt{I_1 I_2}/(I_1+I_2)$, where I_1 and I_2 are intensities of the beams. As predicted by theory we found a pronounced dependence of the current on m. This also gives information about the contrast of the grating recorded in the sample a and the relation between a and m. As shown in [12] the current must have its maximum value when there is no modulation of the light incident on the sample, i.e., when the contrast a equals zero. A modulation of the light results in a modulation of the free carrier distribution leading to a decrease of the net photoconductivity and of the current. To erase the interference pattern we used a phase modulation technique [16]. A phase modulation is applied to one of the beams with a piezoelectric mirror driven by a sinusoidal voltage. By this means the interference pattern (and photorefractive grating in the sample) can be selectively switched on and off without changing the total intensity of the light. Thus by measuring the current with and without modulation of the incident light we obtain the ratio $J/J_{\max}=\sqrt{1-a^2}$, which gives the value of the contrast of the grating. Performing this procedure for different values of m we get an experimental relation between a and m. The experimental dependence is shown in Fig. 2 (filled circles). To compare experiment and theory we use formula (14) with a fitting parameter F. The result of the calculations is also shown in Fig. 2 for three different values of the parameter F.

Our experimental results allow us to estimate the strength of the photogalvanic field in the $KNbO_3$:Fe. The material parameters for this $KNbO_3$ are as follows. The donor concentration is $N_D=10^{25}$ m^{-3}, the absorption coefficient is $\alpha=1$ cm^{-1} and $\varepsilon_{33}=50$. Those values are taken from [17]. The acceptor concentration for this particular sample is $N_A=1.9\times10^{20}$ m^{-3}. We evaluate the photogalvanic field using the relation between E_{pv}^0 and the limiting field E_Q: $F=E_{\mathrm{pv}}^0/E_Q$. To evaluate this field we need only the values of acceptor concentration and dielectric constant specific for our sample and also the value of spacing for grating recorded. To fit our experimental points we used formula (14)

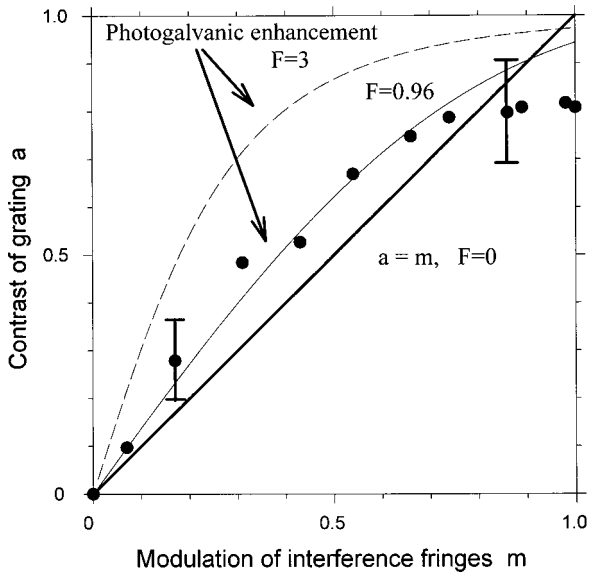


FIG. 2. The dependence of grating contrast a on modulation of interference pattern m. Curves 1–3 calculated by using formula (14) for the case of large photogalvanic field. Filled circles: experiment.

with different values of parameter F. The best coincidence occurs at $F=0.96$. Therefore we can obtain for E_{pv}^0: $E_{\mathrm{pv}}^0=0.96eN_A\Lambda/(2\pi\varepsilon\varepsilon_0)=2.48$ kV/cm. This value is very close to that given in [17].

We have used our compact, analytical method [12] to solve the problem of the description of high contrast gratings in photorefractive, photogalvanic crystals like $LiNbO_3$:Fe and $KNbO_3$. The photogalvanic mechanism of recording dominates in the realization of the first commercially available holographic in $LiNbO_3$ with thermal fixing. Our calculations reveal, unexpectedly, an interesting effect of contrast enhancement: the contrast of the photoconductivity grating can be larger than the contrast of the initial interference pattern. Our experimental results show that this effect is very pronounced for values of m between 0.5 and 0.8. At first glance it looks very unusual: we should remind the reader that drift and diffusion currents result in conductivity gratings with diminished contrast. However, we must take into account that the photogalvanic current by virtue of it being a local effect with a current proportional to the light intensity does not depend on the gradients of the electric potential and concentration like drift and diffusion currents do. This essential difference between the currents explains the effect of contrast enhancement in the photogalvanic crystals.

The fundamental spatial harmonic of the electric field (and of the refractive index grating) for a large photogalvanic field also has some new features such as an absence of space charge screening for small periods and a contrast-dependent phase shift. Contrary to the accepted model, the index grating will be shifted away from the position of light intensity when the contrast is large. In this case there will be no phase exchange introducing instability in the recording process.

We have also programmed the reduced system of equations (1)–(4) and for $j=0$ assuming predominantly photovoltaic contribution to the photorefractive effect. This was done using MATLAB to solve the resulting first-order nonlinear differential equation for the electrostatic field. The results (not plotted here) show the phase difference between

the intensity profile, electrostatic field, and the electron density, in agreement with analytical results above. Furthermore, our computations preserve all spatial harmonics for the electrostatic field and the electron density.

To observe contrast enhancement, the most direct method is to measure the photogalvanic current for a small contrast of the interference pattern. Provided the photogalvanic effect is large enough, we observe a pronounced dependence of the current on the interference pattern contrast m (even for small values of m). The experimental dependence of a as a function of m clearly shows the enhancement of a with respect to m for m between 0 and 0.4.

Contrast enhancement may have practical applications such as increasing the holographic storage time in Fe-doped crystals of $LiNbO_3$ and allowing recording of short-period gratings without space-charge screening.

Sergei Lyuksyutov would like to thank the Danish Technical/Research Council for STVF research Grant No. 9700255. Nickolai Kukhtarev wishes to acknowledge the support from The Technical University of Denmark and from the NSF under Grant No. HRD-9353548. Tatiana Kukhtarev acknowledges the support from AAMU, SHBCU/MI.

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