Enhanced photocurrent in photo-EMF experiments in pure and doped absorbing photorefractive crystals

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Abstract

We report a mathematical formulation that successfully describes the holographic photocurrent produced, in strongly absorbing photorefractive materials, by the action of a pattern of interference fringes of light sinusoidally vibrating with large amplitude. The large vibrating amplitude produces a sensible enhancement of the photocurrent signal and in this way facilitates measurements. We also show that taking account of the bulk light absorption of the sample is essential in order to adequately describe the experiment. We measure the first temporal harmonic of the photocurrent, without externally applied field, as a function of the amplitude and the temporal frequency of the vibrating pattern of fringes and show these data to fit very well our theoretical model. From this fitting we are able to determine some material’s parameters for pure and doped photorefractive Bi$_{12}$TiO$_{20}$ (BTO) crystals.

Introduction

When an interference pattern of fringes of light, with a grating vector $\mathbf{K}$, is projected onto a photoconductive (not necessarily photorefractive) material, a space-charge electric field is built up with a characteristic time $\tau_{sc}$. The pattern of fringes also produces a periodic pattern of photoexcited electrons in the conduction band that is build up with a much smaller characteristic time $\tau_{e}$ that is the so-called electron lifetime. If the pattern of fringes is sinusoidally vibrating along the direction of $\mathbf{K}$, with amplitude $\Delta$ and (angular) frequency $\Omega$, a corresponding ac electric current is produced along $\mathbf{K}$, due to the action of the space-charge field on the free charges in the conduction band [1]. This current depends on the material (photoconductivity, density of photoactive centers, diffusion length of photogenerated electrons, etc.) parameters. It is therefore possible to characterize some of the material's parameters by adequately analyzing the data from an appropriate experiment.

The present technique has already been used to characterize some photorefractive inorganic crystals [1] as well as photorefractive organic polymers [2]. The influence of bulk light absorption was also reported before but for particular conditions and for large frequencies $\Omega\tau_{e} \gg 1$ only. Some experiments using pattern-of-fringes vibration of large amplitude were already published before [2] but they use rather rough approximations for the large amplitudes of modulation in their experiments.

Here we report an accurate formulation for the dc ($j^0$), first ($j^{\Omega}$) and second ($j^{2\Omega}$) harmonic terms in $\Omega$ of the holographic photocurrent density, for bulk absorbing materials, including the case of large amplitudes $\Delta$ of the vibrating pattern of fringes. The large $\Delta$ is intended to sensibly amplify the current signal. Bulk absorption effect on both the photoconductivity and the holographic response time is accounted for in order to adequately describe the experiment in the whole range of $\Omega\tau_{e}$.

Theory

The recorded space-charge field modulation and the free charge-carriers distribution in the conduction band of a nonphotovoltaic photorefractive material, in the absence of an externally applied electric field ($E_0 = 0$), are mutually $\pi/2$–phase shifted so that the electric current, averaged along the interelectrode distance in the direction of $\mathbf{K}$, is zero. However, if the pattern of fringes is moved fast enough, the above referred phase shift is modified and a current may appear in the form of a pulse, an ac or a dc signal, according to the way the pattern of fringes is moved. Let us assume a pattern of fringes sinusoidally oscillating along the coordinate $x$ (parallel to $\mathbf{K}$) as described by
\[ I = I_0 + \frac{k}{2} (m(t)e^{ikx} + cc) \]

where \( m(t) \) is the complex fringes modulation coefficient

\[ m(t) = |m|e^{i\phi} \sum_{l=-\infty}^{\infty} J_l(K\Delta)e^{i\Omega t} \]

where \( J_l() \) is the ordinary Bessel function of order \( l \).

Let us assume electrons to be the only charge-carriers involved. Let us assume the so-called first spatial harmonic approximation that allows one to consider the linearized expressions for the free electron concentration and the overall electric field, respectively

\[ N(x,t) = N_0 + \frac{N_0}{2} (a(t)e^{ikx} + cc) \]
\[ N_D^+(x,t) = N_0^+ + \frac{N_0^+}{2} (A(t)e^{ikx} + cc) \]
\[ E(x,t) = E_0 + \frac{1}{2} (E_{sc}(t)e^{ikx} + cc) \]

The equations above, plus the Poisson relation and the expressions for the current density and the rate equations for \( N \) and \( N_D^+ \) were shown to lead to the following relations for \( E_{sc}(t) \) and \( a(t) \) [3]

\[ E_{sc}(t) = -|m| \frac{E_0}{1+K^2l_0^2} \sum_{l=-\infty}^{\infty} \frac{J_l(K\Delta)e^{i\Omega t}}{1+\Omega^2\tau_{sc}} \]
\[ a(t) = \frac{1}{E_0} \frac{E_0(t)}{1+K^2l_0^2} \sum_{l=-\infty}^{\infty} \frac{J_l(K\Delta)e^{i\Omega t}}{1+\Omega^2\tau_{sc}} \]
\[ \tau_{sc} = \frac{\sigma_0 K^2 l_0^2}{\sigma_0 (1+K^2l_0^2)} \]

where \( l_0 \) and \( l_s \) are the diffusion and the Debye screening lengths, respectively, for the photogenerated electrons, \( \varepsilon \) is the dielectric constant, \( \varepsilon_0 \) is the electric permittivity of vacuum, \( \sigma_0 = e\mu N_0 \) and where we have assumed \( E_0 = 0 \). Its is possible to show [1] that the total current density flowing through the electrodes at the ends of the sample, along the direction of \( K \), can be written as

\[ j(t) = \frac{1}{L} \int_0^L e\mu N(x,t)E_{sc}(x,t)dx \]

where \( L \) is the interelectrode distance. After substituting \( a(t) \) and \( E_{sc}(t) \) and rearranging terms we got the following expression for the dc component, for the first harmonic and the second harmonic term for \( j(t) \)

\[ j^0 = j^{2\Omega} = 0 \]
\[ j^\Omega = -\sigma_0 |m|^2 \frac{J_2(K\Delta)J_2(K\Delta)}{1+\Omega^2\tau_{sc}} E_0 \tau_{sc} - \sigma_0 |m|^2 \frac{J_2(K\Delta)J_2(K\Delta)}{1+\Omega^2\tau_{sc}} \frac{3\Omega\tau_{sc}}{(1-\Omega^2\tau_{sc})(1+2\Omega^2\tau_{sc})} \]

where \( \phi = 0 \). The expression of \( j^\Omega \) does depend on two groups of material parameters only, one being \( \tau_{sc} \) and the other being

\[ A \equiv \frac{\sigma_0 \tau_{sc}}{(1+K^2l_0^2)(1+K^2l_s^2)} \]

From \( A \) it is possible to compute \( l_s \) (assuming that \( \varepsilon \) is known). From \( A/\tau_{sc}^2 \) instead, it is possible to compute \( \sigma_0/(1+K^2l_0^2) \) that allows one to find out \( \sigma_0 \) provided that \( l_0 \) is known.
The physical meaningful value is the real part of the first harmonic term
\[ \frac{i}{\pi} e^{i\Delta t} + \frac{(i\Omega)^{p}}{2} e^{-i\Omega t} \]

Because of bulk absorption, \( \sigma_0(z) = \sigma_0(0) e^{-\alpha z} \) and \( \tau_0(z) = \tau_0(0) e^{-\beta z} \) and the experimentally measured photocurrent value is (H is the height and d is the thickness of the sample)
\[ |i^{\Omega}| = H \int_{0}^{d} j^{\Omega}(z) dz \]

**Experiment**

The Setup is schematically shown in Fig.1 where a sinusoidal pattern of fringes is projected onto the (110) crystallographic plane of the BTO sample with its [001]-axis perpendicular to the plane of incidence and to \( \mathbf{K} \). The angle between the interfering beams (in air) is 51° and the wavelength is \( \lambda=0.5145\mu\text{m} \). A piezoelectric supported mirror (PZT), placed in one of the interfering beams, is driven by a sinusoidal voltage \( v(t) = v_d \sin \Omega t \) that produces a corresponding phase modulation of amplitude (in radians) \( K\Delta = K_{\text{PZT}} v_d \), where \( K_{\text{PZT}} \) is the response of the piezoelectric. We deduce that for \( \Omega \geq 50 \text{ Hz} \) it should be \( |i^{\Omega}| = |i^{\Omega}_{\text{max}}| \) because all curves have approximately the same maxima (that correspond to the maximum for the product \( Jo(K\Delta) J_1(K\Delta) \) that occurs for \( K\Delta = 1.1 \text{ rad} \)). The abscissa of these maxima however are different because of the varying frequency response of the PZT. We used this fact in order to calibrate the PZT. We convert \( v_d \) into its corresponding amplitude \( K\Delta \) of the phase modulation and therefore to plot all the experimental data in Figs. 2, in terms of \( K\Delta \) as shown in the corresponding Figs. 3.

**Fig. 1 Experimental Setup**

**Fig. 2:** The first harmonic amplitude value \( |i^{\Omega}| \) as a function of \( \Phi_{\text{MOD}} \) for different fixed values of \( \Omega \) are shown.

**Fig. 3:** The curves represent the best fit of the theoretical equations to data (spots) shown in fig.2.
Table 1: Best fit parameters

<table>
<thead>
<tr>
<th></th>
<th>BTO</th>
<th>BTO:Ce</th>
<th>BTO:Pb</th>
<th>BTO:Pb a</th>
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<tr>
<td>Io(W/m²)</td>
<td>396</td>
<td>153</td>
<td>382</td>
<td>382</td>
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<tr>
<td>A/τₗₜ(0)(F/sm) 10⁹</td>
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<td>29</td>
<td>13.5</td>
<td>24.5</td>
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<td>A(F/m) 10⁻¹⁰</td>
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<td>τₗₜ(0)(ms)</td>
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<td>-</td>
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<td>7.6</td>
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<tr>
<td>lₗ (µm)</td>
<td>0.051</td>
<td>-</td>
<td>0.2</td>
<td>0.067</td>
</tr>
<tr>
<td>(σ₀(0)/I(0))/(1+K²L₀²) (mwΩ*10⁻⁶)</td>
<td>1.1</td>
<td>-</td>
<td>2.18</td>
<td>1.14</td>
</tr>
</tbody>
</table>

a: neglecting bulk absorption

Data analysis and Conclusions

Data (spots) in Fig.4 are fit to theoretical equation by |i₅|, where bulk absorption is accounted for with their corresponding best fitting parameters being shown in Table1. The lₗ and σ₀(0)/I(0) here reported for the BTO is in rough agreement with those from holographic erasure experiments [4] for this sample. The data reported here and the few data available in the literature for BTO:Ce and BTO:Pb are still not enough to allow one to draw some definite conclusions about the effect of these dopants in BTO. We have shown the interest of using a large modulation amplitude for the phase modulation in the experiment, in order to considerably improve the size of the signal. In this paper we also report an accurate theoretical formulation for this large-amplitude operation condition and draw the attention to the misleading use of some approximations. We have also shown the importance of taking into account the bulk light absorption in these materials.

References