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Anomalous behaviour of long-time increase in diffraction efficiency of photorefractive grating in Ce:$BaTiO_3$ in dark*

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Anomalous long-time increase of the diffraction efficiency is observed in dark-decay experiments of photorefractive gratings in Ce:$BaTiO_3$. It is deduced that a phase-conjugate beam is induced by the writing beam at acute angle to the $+c$ axis of the crystal and it interferes with the other writing beam to form a second grating which is perpendicular to the first grating formed by the interference between two writing beams. The rising behaviour of the diffraction efficiency results from the different decay rates of these two photorefractive gratings. Furthermore, a simplified model of two gratings, both induced by two deep traps, is proposed to account for this phenomenon and the fitting results agree well with the experimental results.

Keywords: dark decay, photorefractive grating, two deep-trap model, Ce:$BaTiO_3$
PACC: 4265, 4230, 4240E

1. Introduction

The dark decay of photorefractive grating refers to the decay of photorefractive space-charge field induced by the translation of thermally generated free carriers under the dark condition. The dark decay time of photorefractive grating is of significance for photorefractive data storage[1,2] and for the application of optical phase conjugation.[3] On one hand, researchers have attempts to find photorefractive materials with longer decay time constants.[4,5] On the other hand, the study of dark decay behaviours in photorefractive crystals stimulates the development of photorefractive dynamics.[6−9]

Generally, the strength of the diffracted beam of photorefractive grating decreases along with the decay of photorefractive grating under the dark condition. It was reported by Tayebati *et al*, however, that in theory, normalized diffraction efficiency could increase and even exceed 1 within the first few seconds in the dark decay process.[7] Afterwards, they presented the experimental evidence of the rising behaviours along with their theoretical interpretation in Ref.[10]. Furthermore, normalized diffraction efficiency exceeding 1 was exhibited in the experimental results of four-wave mixing in $BaTiO_3$ crystal presented by Imtiaz Ahmad Taj *et al.*[11]

Up to now, the reported ascending phenomena of the diffraction efficiency in dark decay experiments of photorefractive gratings were all explained by the shallow-deep model on such condition that the charge grating in the shallow trap has opposite sign to that in the deep trap. Additionally, the rising processes of the diffraction efficiency in previous experiments all merely lasted for a short time (a few seconds). In this article, we report for the first time, the observation of an anomalous long-time increase (several hours) of the diffraction efficiency in the dark decay experiments of Ce:$BaTiO_3$, and deduce that this phenomenon does not result from the interaction between charge gratings with opposite signs in the shallow and deep traps

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but from the interaction between two mutually perpendicular photorefractive gratings formed inside the crystal.

2. Experimental setup

The experimental arrangement (Fig.1) and procedures are depicted in Ref.[8] except that a half-wave plate $\lambda/2$ is used here to obtain either e-polarized or o-polarized beams. The polarized Ce:$BaTiO_3$ used in our experiments was 0° cut and its dimension was 7.09mm×6.42mm×4.14mm with the c-axis along the 7.09-mm-long edge (the Ce:$BaTiO_3$ crystal was purchased from the Institute of physics, Chinese Academy of Sciences). Photorefractive grating is written by two cw-532nm laser beams crossing inside the crystal. After saturation of the grating, one of the writing beams is blocked with a shutter and the other writing beam, which is attenuated by a neutral attenuator ND, and is used as the readout beam directly. So the Bragg matching condition is satisfied automatically. It is also approved experimentally that the erasure effects of the readout beam on the original grating could be ignored by controlling both the readout beam intensity and duration. The diffracted beam of the grating which transmitted in the same direction as W_1 is measured by photomultiplier PMT.

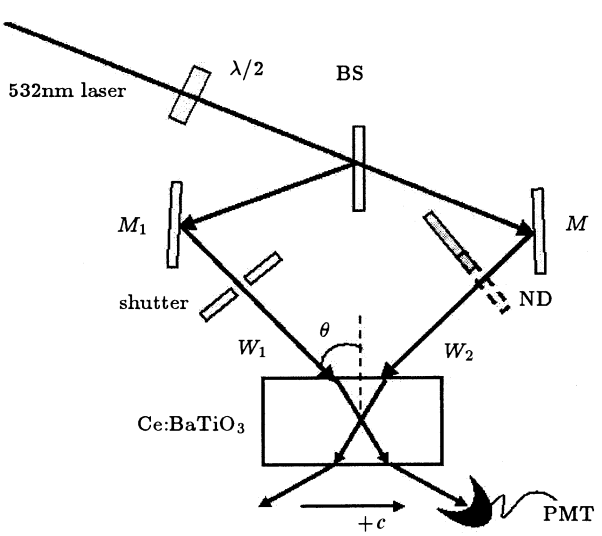


Fig.1. Experimental arrangement for measuring the dark-decay characteristics of photorefractive grating.

3. Results and discussions

3.1. Experimental results

In the two experiments which we carried out, the powers of the two writing beams are both equal to 10mW and the incident angle θ=25°. The grating wave vector $K = 2\pi/\Lambda = 10(\mu m^{-1})$, where the grating interval $\Lambda = \lambda/2n\sin\theta' = \lambda/2\sin\theta$, λ is the wavelength of the writing beams in vacuum, 2θ and $2\theta'$ are the angles included between the two writing beams on the surface and inside the crystal respectively, n is the refractive index of the crystal.

In the first experiment, both of the writing beams are e-polarized. The curve of the normalized diffraction efficiency versus time (the increasing curve) is plotted in Fig.2, from which we can see that the diffraction efficiency decays a little for about 10min in the beginning and then keeps rising for hours. In the second experiment, by contrast, both the writing beams are o-polarized. The curve of the normalized diffraction efficiency (the decreasing curve) is also plotted in Fig.2. Note that the diffraction efficiency does not exhibit a rising behaviour as observed in the e-polarized experiment.

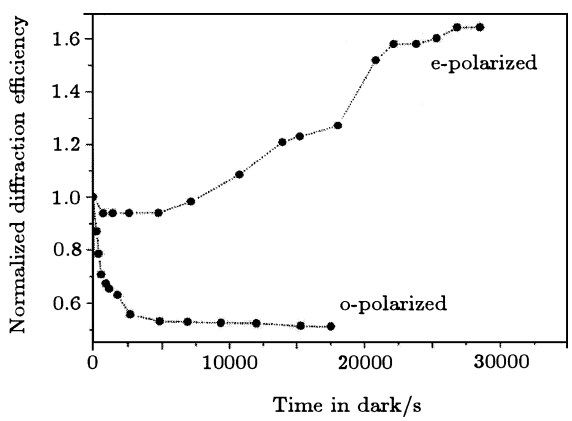


Fig.2. Traces of diffraction efficiency when both the writing beams are e-polarized or o-polarized (normalized to its initial value at t=0).

3.2. Discussions

According to the shallow-deep model, if the charge gratings formed during illumination in the deep and the shallow traps have opposite signs and the two traps decay at different rates, the resulting space-charge field and thus the diffraction efficiency might initially increase. As the decay time constant related to the shallow trap in pure or doped $BaTiO_3$ crystals is usually in the order of seconds, the rising process also ends in a few seconds.[7,10,11] However the rising process of the diffraction efficiency in our e-polarized experiment lasts for more than 10h, which is much longer than those reported in the previous researches. Therefore, the mechanism based on the shallow–deep

model could not be used to explain the long-time increase of the diffraction efficiency observed in our experiment.

In Ref.[8], it has been observed that for the Ce:$BaTiO_3$ crystal, the contribution of the shallow trap to the whole charge grating could be ignored and that the Ce-ion and Fe-ion deep traps are the two main traps involved in the forming of photorefractive grating, whose behaviour can be interpreted by the two deep-trap model. As in the shallow–deep model, the diffraction efficiency could exhibit a long-time increase if the signs of the charge gratings in the two deep traps are opposite. This explanation, however, fails to account for the decay curves shown in Fig.2, because the temporal behaviour of the diffraction efficiency with o-polarized writing beams should be similar to that with e-polarized writing beams, which is obviously inconsistent with our experimental results.

Noting the dramatic distinction between the temporal behaviours of the diffraction efficiency in e-polarized and o-polarized experiment, we then focus our attention on the different effects caused by different polarizations of the incident lights. As the effective electro-optic coefficient for the e-polarized incident beam is much larger than that for the o-polarized incident beam, the e-polarized writing beam is more likely to induce self-pumped phase-conjugate beam than the o-polarized beam is. So we deduce that it is the formation and existence of the phase-conjugate beam that leads to the increasing behaviour of the diffraction efficiency under the dark condition. The mechanism could be described with Fig.3. In the e-polarized experiment, two mutually perpendicular gratings, in which grating 1 is induced by the interference between two writing beams W_1 and W_2, and grating 2 is induced by W_2 and the phase-conjugate beam PC of W_1, coexist inside the crystal. When the crystal is illuminated by a weak readout beam R transmitting in the same direction as W_2, i.e., R is Bragg-matched with both grating 1 and grating 2, two 1st order diffracted beams are formed, in which O_1 is from grating 1 in the direction of W_1 and O_2 is from grating 2 in the direction of PC, respectively. As the intensity of the diffracted beam (O_1) is positively related to the strength of grating 1 and negatively related to grating 2, the decay of grating 1 will cause the intensity of O_1 to decrease whereas the decay of grating 2 will cause it to increase. So under the dark condition, if grating 2 decays faster than grating 1 does, the intensity of the diffracted beam O_1 could ascend. In the o-polarized experiment, however, since the phase-conjugate beam has not been built up at all within the illumination duration or its intensity is so weak that its effects can be ignored, only one photorefractive grating (grating 1) exists inside the crystal. Therefore, the diffraction efficiency does not ascend but decays according to the two deep-trap model.

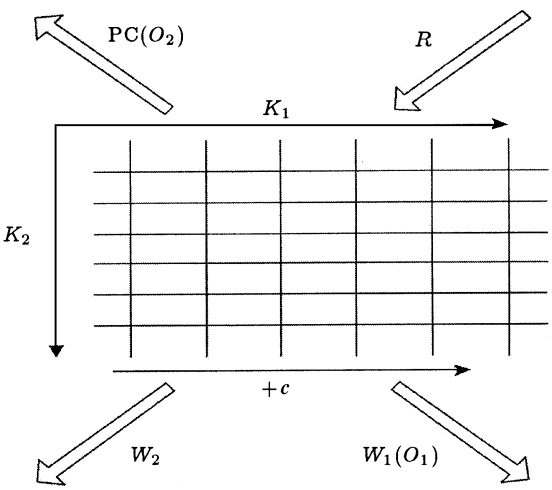


Fig.3. Mutually perpendicular gratings in the crystal (grating 1 is induced by the interference between writing beams W_1 and W_2, and grating 2 is induced by W_2 and the phase conjugate beam PC).

4. Simplified model and fitting for the experimental results

4.1. Model

Assume that the two mutually perpendicular refractive-index gratings formed in the crystal are independent to each other and they can be written as

$$n_1 = n_0 + \frac{1}{2}\Delta n_1 \exp(-\mathrm{i}\boldsymbol{K}_1 z) + \frac{1}{2}\Delta n_1 \exp(+\mathrm{i}\boldsymbol{K}_1 z), \quad (1)$$

$$n_2 = n_0 + \frac{1}{2}\Delta n_2 \exp(-\mathrm{i}\boldsymbol{K}_2 z) + \frac{1}{2}\Delta n_2 \exp(+\mathrm{i}\boldsymbol{K}_2 z), \quad (2)$$

where n_1 and n_2 are the refractive indices of grating 1 and grating 2 respectively. n_0 is the refractive index of the crystal without illumination, Δn_1, Δn_2 are the refractive index modulations induced by the photorefractive effects, $\boldsymbol{K}_1$, $\boldsymbol{K}_2$ are grating wave vectors of grating 1 and grating 2 respectively and $\boldsymbol{K}_1 \perp \boldsymbol{K}_2$.

When an individual photorefractive grating in a crystal is illuminated by a Bragg-matched readout beam, the diffraction efficiency can be written as

$\eta = \dfrac{I_O}{I_i} \propto |E^{\rm sc}|^2 \propto (\Delta n)^2$ if the refractive index modulation Δn of the grating is very small.[7] Here, $E^{\rm sc}$ is the intensity of space-charge field formed by the writing beams in the crystal, $I_{\rm i}$ is the power of the readout beam and I_O the power of the diffracted beam. Therefore, $I_O \propto I_i(\Delta n)^2$.

When two photorefractive gratings coexist inside the crystal and both of them are Bragg-matched with the readout beam, two diffracted beams O_1, O_2 would be produced. Due to the interaction between the two gratings, the relationship between the powers of the diffracted beams I_{O1}, I_{O2} and the power of the readout beam $I_{\rm i}$ should also be modified, which can be written as $I_{O1} \propto \alpha_1 I_i(\Delta n_1)^2$, $I_{O2} \propto \alpha_2 I_i(\Delta n_2)^2$ where α_1 and α_2 are modified coefficients for I_{O1} and I_{O2} respectively. Firstly, since α_1 and α_2 represent the effects of the coexistence of two gratings, they must be functions of Δn_1 and Δn_2. Secondly, α_1 and α_2 should obey the rule that the stronger the grating is, the greater the power of the diffracted beam related to it will be. Finally, α_1 and α_2 must be able to represent the situation of an individual grating. For example, if only grating 1 exists ($\Delta n_2 = 0$), α_1 must equal to 1 and thus $I_{O1} \propto I_i(\Delta n_1)^2$. Taking into account these three prerequisites which α_1 and α_2 must satisfy, we postulate that $\alpha_1 = \dfrac{(\Delta n_1)^2}{(\Delta n_1)^2 + (\Delta n_2)^2}$ and $\alpha_2 = \dfrac{(\Delta n_2)^2}{(\Delta n_1)^2 + (\Delta n_2)^2}$. Then I_{O1} and I_{O2} can be rewritten as

$$I_{O1} \propto \alpha_1 I_{\rm i}(\Delta n_1)^2 = \frac{(\Delta n_1)^4}{(\Delta n_1)^2 + (\Delta n_2)^2} I_{\rm i}, \quad (3)$$

$$I_{O2} \propto \alpha_2 I_{\rm i}(\Delta n_2)^2 = \frac{(\Delta n_2)^4}{(\Delta n_1)^2 + (\Delta n_2)^2} I_{\rm i}. \quad (4)$$

In the previous dark-decay experiments of photorefractive grating in Ce:$BaTiO_3$,[8] it was observed that two deep traps, i.e. Fe-ion deep trap and Ce-ion deep trap, both participate in the photorefractive process. According to the two deep-trap model, when the crystal is under the dark condition and there is no external electric field, the two deep traps respond to the incident light separately and their respective electric fields are linearly superposed to form the whole electric field which is given by

$$E^{\rm sc}(t) = E^{\rm sc}_{\rm C}(0)\exp\left(-\frac{t}{\tau_{\rm C}}\right) + E^{\rm sc}_{\rm F}(0)\exp\left(-\frac{t}{\tau_{\rm F}}\right), \quad (5)$$

where $E^{\rm sc}_{\rm C}(0)$ and $E^{\rm sc}_{\rm F}(0)$ are the steady-state intensities of the space-charge fields in the Ce-ion deep trap and Fe-ion deep trap respectively (subscript C and F stand for Ce and Fe respectively). The decay-time constants of the two deep traps are $\tau_{\rm C} = \dfrac{\varepsilon}{\mu q N_{\rm C}}$ and $\tau_{\rm F} = \dfrac{\varepsilon}{\mu q N_{\rm F}}$, where ε is the dielectric constant, μ is the hole mobility, q is the electric quantity of a carrier, $N_{\rm C}$ and $N_{\rm F}$ are the hole concentrations in the two deep traps respectively, and it is known that $\tau_{\rm C} \gg \tau_{\rm F}$ due to the deeper energy level of Ce-ion in bandgap. Because $\Delta n(t) \propto E^{\rm sc}(t)$, the refractive index modulation of the grating also decays with two exponentials which can be written as

$$\Delta n_1(t) = \Delta n_1(0)\left[a_{\rm C}\exp\left(-\frac{t}{\tau_{\rm C}}\right) + a_{\rm F}\exp\left(-\frac{t}{\tau_{\rm F}}\right)\right], \quad (6)$$

$$\Delta n_2(t) = \Delta n_2(0)\left[b_{\rm C}\exp\left(-\frac{t}{\tau_{\rm C}}\right) + b_{\rm F}\exp\left(-\frac{t}{\tau_{\rm F}}\right)\right], \quad (7)$$

where $\Delta n_1(0)$ and $\Delta n_2(0)$ are the refractive index modulations of the two gratings in the steady state respectively. $a_{\rm C}$, $a_{\rm F}$ and $b_{\rm C}$, $b_{\rm F}$ represent the proportion of the space-charge field in each deep trap to the whole steady-state space-charge field for grating 1 and 2 respectively. Substituting 0 for t in Eq.(6) and (7) yields $a_{\rm C} + a_{\rm F} = 1$ and $b_{\rm C} + b_{\rm F} = 1$. Supposing $\Delta n_2(0) = \beta \Delta n_1(0)$, we can rewrite Eqs.(6) and (7) as

$$\Delta n_1(t) = \Delta n_1(0)\left[a_{\rm C}\exp\left(-\frac{t}{\tau_{\rm C}}\right) + (1 - a_{\rm C})\exp\left(-\frac{t}{\tau_{\rm F}}\right)\right], \quad (8)$$

$$\Delta n_2(t) = \beta\Delta n_1(0)\left[b_{\rm C}\exp\left(-\frac{t}{\tau_{\rm C}}\right) + (1 - b_{\rm C})\exp\left(-\frac{t}{\tau_{\rm F}}\right)\right]. \quad (9)$$

In our experiments, the intensity of the diffracted beam O_1 was measured and the normalized diffraction efficiency related to it is given by

$$\begin{aligned}\eta_{\rm nor1} &= \frac{I_{O1}(t)/I_{\rm i}}{I_{O1}(0)/I_{\rm i}} = \frac{I_{O1}(t)}{I_{O1}(0)} \\ &= \frac{\dfrac{[\Delta n_1(t)]^4}{[\Delta n_1(t)]^2 + [\Delta n_2(t)]^2}}{\dfrac{[\Delta n_1(0)]^2}{1 + \beta^2}} \\ &= \frac{[\Delta n_1(t)]^4}{[\Delta n_1(t)]^2 + [\Delta n_2(t)]^2} \times \frac{1 + \beta^2}{[\Delta n_1(0)]^2}. \end{aligned} \quad (10)$$

4.2. Fitting for the experimental results

In Fig.4, the rising curve is the fitting result to the e-polarized experimental data in Fig.2 with Eq.(10) (the dots show the experimental data and the solid line is the fitting curve).

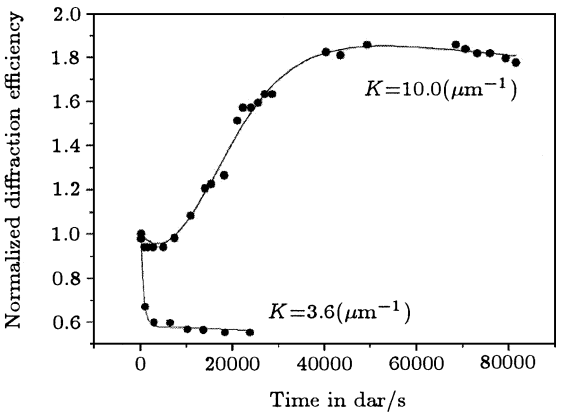


Fig.4. Traces of diffraction efficiency when the writing beams are both e-polarized but with different incident angles (normalized to its initial value at t=0).

From the fitting, the fast decay time constant τ_F of 9×10^3s is obtained. The slow decay time constant, however, could not be obtained correctly as the measurement duration is so short compared with τ_C. But when $\tau_C > 1.7 \times 10^6$s, the variation of τ_C has almost no effect on the fitting curve which indicates that τ_C is larger than 1.7×10^6s. The values of τ_C and τ_F are consistent with the results of our previous experiments.[8]

According to the two deep-trap model, the greater the contribution of the Fe-ion to the whole space-charge field is, the faster the grating decay will be. In our e-polarized experiment, the fitting value of a_C is 0.53 and b_C is 0.15, i.e. $a_F = 0.47$ and $b_F = 0.85$. Therefore the contribution of the Fe-ion to the whole space-charge field in grating 2 is greater than that in grating 1 and thus grating 2 decays faster than grating 1 does. As $\eta_{nor1} \propto \frac{[\Delta n_1(t)]^4}{[\Delta n_1(t)]^2 + [\Delta n_2(t)]^2}$, when Δn_2 falls faster than Δn_1, η_{nor1} could exhibit an increasing behaviour, which is shown by the rising curve in Fig.4.

The decreasing data in Fig.4 is the result of another e-polarized experiment in which the incident angle θ=9°($K = 3.6\mu m^{-1}$). It is obvious that the diffraction efficiency does not show a large increase as observed in the previous e-polarized experiment, this distinction results from the different incident angles of writing beams in the experiments. It has been demonstrated that for the Ce:$BaTiO_3$ crystal used in our experiment, the more the incident angle θ of the writing beam approaches 55°, the larger the effective electro-optic coefficient is and thus the greater the intensity of the induced phase-conjugate beam is.[3] Due to the small incident angle θ=9° and thus the small effective electro-optic coefficient, the phase-conjugate beam induced in the crystal is so weak that the second grating (grating 2) is negligible. According to the two deep-trap model, in the presence of an individual photorefractive grating, the intensity of the diffracted beam will decay under the dark condition. The decreasing data in Fig.4 is fitted with Eq. (10) as β is fixed as 0, which means that grating 2 is neglected. Similarly, the decreasing data in Fig.2 which is obtained from the o-polarized experiment can be fitted by this method as well and the fitting curves agree well with our experimental results.

5. Conclusion

In this paper, we report, for the first time, the observation of an anomalous long-time increase of the diffraction efficiency in the dark decay experiments of Ce:$BaTiO_3$. We deduce that the writing beam at acute angle to the $+c$ axis of the crystal induces a phase-conjugate beam, which interferes with the other writing beam to form a second grating, and this grating is perpendicular to the first grating formed by the interference between the two writing beams. There are different decay rates for these two photorefractive gratings which result in the rising behaviour of the diffraction efficiency. In addition, a simplified model based on the two-deep trap model is presented to account for this phenomenon and the fitting curves are in good agreement with our experimental results.

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