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Dark decay behaviours of photorefractive grating induced by two deep-trap levels in Ce:$BaTiO_3$*

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Dark decay behaviours of photorefractive grating in Ce:$BaTiO_3$ crystal are studied experimentally. It is observed that two deep-trap levels, i.e. Fe-ion deep level and Ce-ion deep level, both participate in the photorefractive process. A simplified two-deep model is presented based on the one-deep trap model and the shallow-deep trap model, with which we analyse quantitatively the contribution of each deep level to the whole space-electric field and thus the photorefractive grating varying with the different grating wave vectors and different writing intensities.

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

1. Introduction

The dark decay of photorefractive grating refers to the decay of photorefractive space-electric field which is 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]

The dark decay-time constant of $BaTiO_3$ is closely interrelated with the position of the trap level in the band gap of the crystal, which is often determined by measuring the dark decay characteristics of photorefractive grating.[4] It is commonly thought that a Fe-ion deep trap level and a shallow trap level exist in pure $BaTiO_3$.[5,6] The levels of doped $BaTiO_3$ are, however, more complex since more than two trap levels exist in the energy band. For example, in Refs.[7] and [8], the positions of one deep trap level (Ce-ion deep trap) and two shallow levels were determined in Ce:$BaTiO_3$.

The previous experimental results on the photorefractive effect in Ce:$BaTiO_3$ crystal indicated that there was only one deep level as well as one shallow level (or several shallow levels) involved in the building of photorefractive grating,[4,7,8] whose dark decay characteristics could be explained by the shallow-deep model[5,6,9] (the effects of several shallow levels could be regarded as one equivalent shallow level), and the dark decay-time constant at room temperature could be as long as thousands of years.[4]

In our experiments of the dark decay characteristics of photorefractive grating with Ce:$BaTiO_3$, it is observed that the two deep traps (Fe-ion and Ce-ion deep trap) make contributions to the forming of photorefractive grating simultaneously. On the basis of one-deep model and shallow-deep model, a simplified kinetic model with two deep traps (two deep-trap model) is presented and the experimental phenomena are analysed.

2. Simplified photorefractive kinetic model with two deep trap levels

The theoretic foundation of analysing the microscopic mechanism of photorefractive effect is the photorefractive kinetic model. The earliest photorefractive kinetic model is one-deep model,[10] which is convinced that there is only one deep level in the band gap of the crystal. So under the dark condition the photorefractive grating will decay in a single exponential

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form. Later, as the one-deep model could not be used to explain the phenomena of several decay constants existing in the experimental results, Brost *et al*[5] and Tayebati and Mahgerefteh[6] presented the shallow–deep trap model: in the band gap, there is a shallow level which is very close to the conduction band (or valence band) besides a deep level. This model considers that in the dark decay process, the carriers on the shallow level are thermally excited firstly, then not only is the charge grating in the shallow level erased entirely, but also the charge grating in the deep level is erased partly or even totally, due to the carriers on the shallow level returning to the deep level. The remaining charge grating in the deep level will decay much slowly as its thermal-excitation rate is very small. In brief, a strong coupling exists between the shallow and the deep levels in the dark decay process. The shallow–deep model has been used to explain kinds of experimental phenomena preferably. But if two deep levels participate in the building of the photorefractive grating, the one-deep model and the shallow–deep model would be unfit for explaining the phenomena.

For the photorefractive process with two deep levels, even in the thermal equilibrium under the dark condition, each deep level has its own carriers. Furthermore, since the thermal-excitation rates are both very small, the coupling between them is much weaker than that between the shallow and the deep level. Assuming the coupling between the two deep levels and the contribution of shallow level to the whole electric field could be ignored, the two deep levels respond to the incident light separately and then their respective electric fields are linearly superposed to form the whole electric field. Therefore the photorefractive process with two deep traps could be described by the superposition of two one-deep model. Namely, when the crystal is under the dark condition and there is no external electric field, the decay of electric field on each deep level is in a single exponential form:

$$E_i^{\rm sc}(t) = E_i^{\rm sc}(0)\mathrm{e}^{-t/\tau_i}, \quad i = 1, 2, \tag{1}$$

where i represents the first (i=1) deep level or the second (i=2) deep level, the decay-time constant of the level is $\tau_i = \varepsilon/\mu q n_i$, n is the number density of free carriers, q is the electric-charge of a carrier, μ is the mobility of free carrier, ε is the dielectric constant, $E^{\rm sc}$ is the intensity of space-electric field under the dark condition, $E^{\rm sc}(0)$ is the intensity of space-electric field when the grating is in the steady state.

The whole space-electric field is

$$E_{\rm sc}(t) = E_1^{\rm sc}(0)\mathrm{e}^{-t/\tau_1} + E_2^{\rm sc}(0)\mathrm{e}^{-t/\tau_2}, \tag{2}$$

so under the dark condition, the decay of whole space-electric field consists of two exponentials.

As the diffraction efficiency of photorefractive grating $\eta \propto |E_{\rm sc}(t)|^2$, the normalized diffraction efficiency is:

$$\begin{aligned}\eta_{\rm nor}(t) &= \left|\frac{E_{\rm sc}(t)}{E_{\rm sc}(0)}\right|^2 \\ &= A^2\mathrm{e}^{-2t/\tau_1} + B^2\mathrm{e}^{-2t/\tau_2} \\ &\quad + 2AB\mathrm{e}^{-t(1/\tau_1+1/\tau_2)},\end{aligned} \tag{3}$$

where

$$A = \frac{E_1^{\rm sc}(0)}{E_1^{\rm sc}(0) + E_2^{\rm sc}(0)},$$

and

$$B = \frac{E_2^{\rm sc}(0)}{E_1^{\rm sc}(0) + E_2^{\rm sc}(0)}.$$

Regarding the photorefractive crystal Ce:$BaTiO_3$, if both Fe-ion and Ce-ion deep traps are involved in the photorefractive process, then the deep level of Ce-ion can be defined as the first deep level and the deep level of Fe-ion as the second. Then A and B represent the proportion of the charge grating in Ce-ion level and that in Fe-ion level in the whole steady-state space-electric field, respectively.

3. Experimental setup

The experimental arrangement for investigating the dark decay characteristics of photorefractive grating is shown in Fig.1. A CW 532nm laser as a writing light passed through a beam splitter BS and was split into two beams. After reflected by M_1 and M_2, two o-polarized incident beams entered the Ce:$BaTiO_3$ crystal, forming light-intensity space-modulated fringes by interference. The direction of the wave vector of photorefractive grating was along the c-axis of the crystal. Each time before grating writing, it was necessary to erase all original grating with homogeneous light. 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).

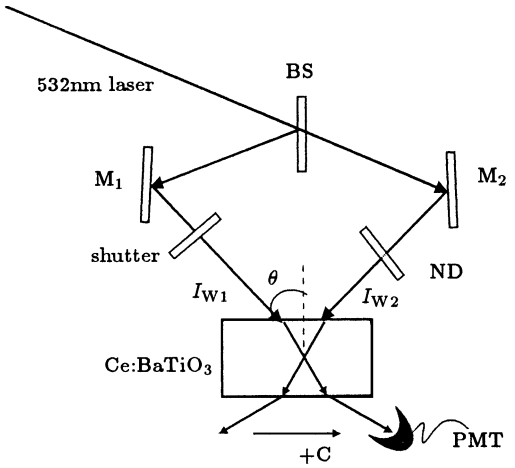


Fig.1. Experimental arrangement for measuring the dark decay characteristics of photorefractive grating.

In the experiments of the dark decay characteristics of photorefractive grating, after saturation of the grating, a Bragg-matched weak probe beam (its wavelength might be different from that of the writing light) was usually used to detect the diffraction signal of the grating. But in our experiments, for convenience, one of the writing beams was blocked with a shutter after saturation of the grating. The other writing beam, which was attenuated by a neutral attenuator ND, was used as a readout light directly. So the Bragg matching condition was satisfied automatically. The diffraction signal of grating was measured by a photomultiplier PMT.

The readout beam should be very weak and the readout duration should be short enough so as to diminish the erasure effect of the readout beam on the photorefractive grating. The intensity of readout beam was 0.07μW (when $I_{W1} = I_{W2}$=10mW) or 0.035μW (when I_{W1}=I_{W2}=5mW) in our experiments. It was approved experimentally that the erasure effects of the readout beam on the original grating could be ignored by controlling the readout duration.

4. Results and discussions

At room temperature (20°C), when the powers of the two writing beams are both equal to 10mW (the total incident power I_{total}=20mW), the dark decay curves of the normalized diffraction efficiency at different grating wave vectors are plotted in Fig.2. The dashed lines connect the experimental dots and the real lines are the fitting curves.

The grating wave vector $K = 2\pi/\Lambda(\mu m^{-1})$, where the grating interval $\Lambda = \lambda/2n\sin\theta' = \lambda/2\sin\theta$, λ is the wavelength of writing beam (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, $n = n_o$ when the writing beam is o-polarized.

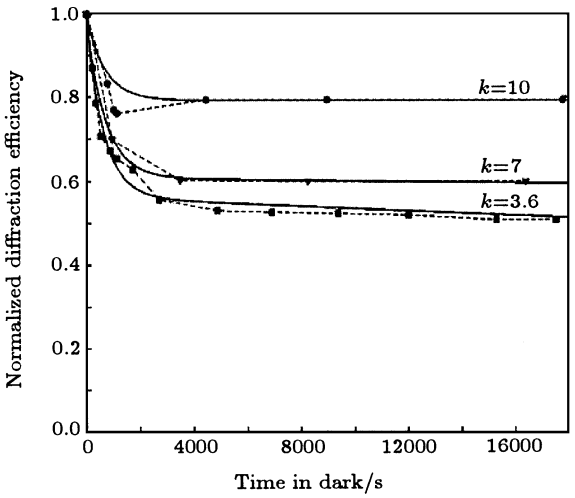


Fig.2. Traces of dark decay on different grating wave vectors (normalized to its steady state value at t=0).

As the decay-time constant related to the shallow level is in the order of seconds at room temperature, it is found that the charge grating in the shallow level and its effect on the whole space-electric field are both very weak. On the other hand, the Fe-ion deep trap level decays with a time constant of tens of minutes to a few hours whereas the Ce-ion deep level has a decay-time constant of tens of hours to thousands of years.[4,6,11] From Fig.2 we can see that the dark decay process consists of a fast stage and a slow stage, which indicates that there are two deep levels involved in the forming of photorefractive grating. According to the decay-time constants of different levels, it can be determined that the fast decay and the slow decay processes are attributed to the charge grating in the Fe-ion and Ce-ion deep levels, respectively. The faster the grating decay is, the greater the contribution of Fe-ion to the whole space-electric field will be.

As the contribution of charge grating in the shallow level to the whole space-electric field is so small that the experimental results could be analysed quantitatively with the simplified two-deep level model. Using the least-squares method to fit Eq.(3), the fast decay-time constant τ_2 of 714s (about 12 minutes) is obtained. The slow decay-time constant, however, could not be obtained correctly as the measurement duration is so short compared with the slow decay-time constant τ_1. But when the slow decay-time constant $\tau_1 > 2\times10^6$s, the variation of τ_1 does not affect the values of A and B, furthermore, the fitting curves

have almost no change in our measurement range. So it can be deduced that the slow decay-time constant τ_1 is larger than 2×10^6s (about 23 days). Slow decay-time constant could be exactly obtained by measuring the decay process at elevated temperatures.[4]

When the direction of grating wave vector k is certain and its value is much less than that of Debye screening wave vector k_0 (Ce:$BaTiO_3$ has a large k_0[12]), the decay-time constant only relates to the position of the level. Consequently, the variety of k does not influence the decay-time constant, but changes the value of $E_1^{sc}(0)/E_2^{sc}(0)$ $(=A/B)$: when k=3.6μm^{-1}, then A=0.75 and B=0.25; when k=7μm^{-1}, then A=0.78 and B=0.22; when k=10μm^{-1}, then A=0.89 and B=0.11. It can be seen from the above that A is greater than B, this is because the trap density of Ce level is generally greater than that of Fe level in Ce-doped $BaTiO_3$, the contribution of the charge grating in Ce-ion level to the whole grating is greater than that of Fe-ion level. Furthermore, the smaller the k is, the less the proportion of the charge grating in Ce-ion deep level in the whole space-electric field is. The reason is probably that the diffusion length of Ce-ion is shorter than that of Fe-ion, and the smaller k corresponds to a larger grating interval. When the diffusion length is small, it is uneasy to form charge grating via diffusion which is the dominant mechanism for pure or doped $BaTiO_3$ crystals to establish charge grating.

The experimental results also demonstrate that not only the value of k, but also the power of the writing beam influence the ratio $E_1^{sc}(0)/E_2^{sc}(0)$. Figure 3 shows the dark decay curves when k=3.6μm^{-1} and the writing powers I_{total} are 10mW and 20mW, respectively. The dashed lines connect the experimental dots and the real lines are the fitting curves. By using the two-deep model, we can obtain A=0.97 and B=0.03 when I_{total}=10mW; A=0.75 and B=0.25 when I_{total}=20mW. The larger the power of the writing beam is, the faster the grating decays, thus the stronger the charge grating in the Fe-ion level is, which is consistent with the experimental results in Ref.[3].

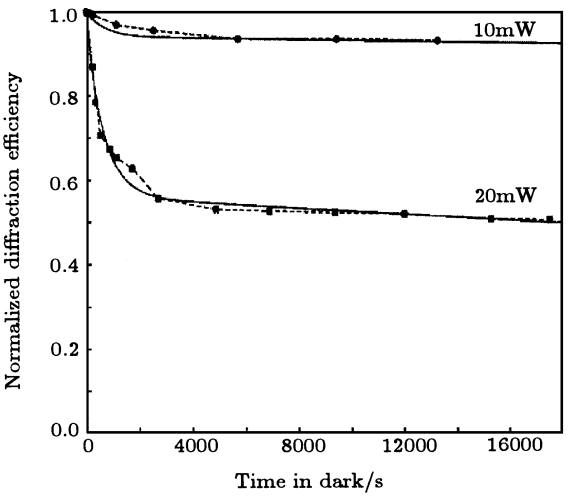


Fig.3. Traces of dark decay at different writing powers I_{total}, when k=3.6μm^{-1} (normalized to its steady state value at t=0).

5. Conclusions

It is observed that Fe-ion deep level and Ce-ion deep level both make contributions to the photorefractive process in the experiments of the dark decay behaviours of photorefractive grating with Ce:$BaTiO_3$. Using the simplified two-deep model, we have analysed quantitatively the contribution of each deep level to the whole space-electric field with different grating wave vectors and different writing beam intensities. As the Fe-ion deep level participates in the forming of photorefractive grating, the dark decay process is expedited greatly, which is unfavorable for the application of Ce:$BaTiO_3$ to optical data storage.

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