

# Light-induced absorption in photorefractive BaTiO<sub>3</sub> crystals

Ravinder Kumar Banyal and B. Raghavendra Prasad<sup>a)</sup>

*Indian Institute of Astrophysics, Bangalore 560034, India*

(Received 3 January 2003; accepted 8 April 2003)

We present the experimental results on the measurement of temporal and steady-state light-induced absorption change in undoped and rhodium-doped barium titanate (Rh:BaTiO<sub>3</sub>) crystals at different wavelengths and intensities. Theoretical calculations based on a two-center charge transport model agree well with the experimental results which supplement the earlier studies carried out using photorefractive BaTiO<sub>3</sub> crystals. We also report a wavelength-specific light-induced effect that modifies the absorption dynamics in a peculiar manner. This can be attributed to light-induced thermal effects. However, the possibility of additional photorefractive centers becoming active cannot be ruled out. © 2003 American Institute of Physics. [DOI: 10.1063/1.1578533]

## I. INTRODUCTION

Photorefractive (PR) materials show intensity-dependent refractive index and absorption changes that can be exploited in a variety of applications<sup>1</sup> such as optical information processing, holographic data storage, coherent image amplification, phase conjugation, etc.

BaTiO<sub>3</sub> is one of the most widely used PR materials owing to the very high value of its electro-optic coefficient, faster response time, and excellent phase conjugate reflectivity. Applications like real-time holography, optical switching, and phase conjugation require a very fast response of the crystal. Response time depends on intensity of the input beams, however, the beam fanning, light-induced absorption (LIA), and thermal effects can seriously degrade the PR performance of the crystal at high light intensity.

In this article, we examine the steady-state and temporal behavior of LIA of a probe beam passing through the crystal that is subjected to pump beam illumination at different wavelengths and intensities. The observed behavior is interpreted in terms of a two-center charge transport model for PR materials. An anomalous temporal evolution of absorption at 633 nm probe and 514 nm pump illumination was also observed in Rh:BaTiO<sub>3</sub> crystal. Different crystal parameters like thermal excitation rate, absorption build up, and decay rate have been obtained and compared with the earlier studies.

First, we briefly outline the absorption mechanism qualitatively in the light of the two-center charge transport model. Details of the experiments, results, and, finally, the discussion and conclusions are presented in the following.

## II. BACKGROUND

The presence of impurity atoms like Fe, Mn, Cu, etc. (also called primary or deep centers) is thought to be responsible for the photorefractive effect. A nonuniform illumination of the crystal by coherent light photoexcite the charges from impurity sites to the conduction band. Under the influence of diffusion or drift, charges are migrated from brighter

regions and subsequently trapped in darker regions. Such redistribution of charges between brighter and darker regions leads to the development of internal electric field which in turn modulates the refractive index of the material via electro-optic effect. Therefore, light absorption by these impurity centers is the initial and the important step for the photorefractive process.

A model based on single carrier primary centers,<sup>2</sup> explains the PR effect in most of the cases satisfactorily. However, it fails to explain the sublinear dependence of the two-beam coupling coefficient, nonlinear dependence of photoconductivity and intensity dependence of absorption coefficients in BaTiO<sub>3</sub> crystals. Increase in absorption due to illumination of BaTiO<sub>3</sub> was first discovered in 1987 by Motes and Kim.<sup>3</sup> In 1988, Brost *et al.* explained this effect by a two-center charge transport model.<sup>4</sup>

This model is based on the assumption that two different impurity centers, each of them occurring in different valence states, may be present. The first is deep and the second is shallow (also called secondary traps) with respect to the valence band edge. The secondary centers are highly ionized at room temperature, and therefore, do not contribute much to absorption at low light intensity. It is only at high laser intensity when the photon-excitation rate of shallow traps becomes comparable to thermal excitation rate, that the absorption increases. Experimentally, the LIA change in absorption coefficient  $\Delta\alpha$  can be determined using the relation<sup>3</sup>

$$\frac{I_s(\text{with } I_p \text{ on})}{I_s(\text{with } I_p \text{ off})} = \exp(-\Delta\alpha d), \quad (1)$$

where  $d$  is the effective length of the crystal, and  $I_s$  and  $I_p$  are probe and pump beam intensities, respectively. Time evolution of the LIA in the buildup process can be described by<sup>5</sup>

$$\Delta\alpha(t) = [1 - A_0 \exp(t/\tau)]. \quad (2)$$

Here,  $A_0$  is a constant, whereas the rise time constant  $\tau$  can be written as<sup>6,7</sup>

$$\tau^{-1} = aI + \beta_2, \quad (3)$$

<sup>a)</sup>Electronic mail: brp@iiaap.ernet.in

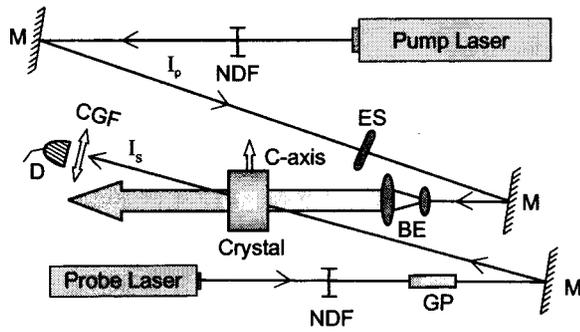


FIG. 1. Schematic of the experimental setup: M, mirrors; NDF, neutral density filter; ES, electromechanical shutter; GP, Glan laser polarizer; D, detector; BE, beam expander; CGF, color glass filter;  $I_p$ , pump beam; and  $I_s$ , probe beam.

where  $a$  is a factor that is independent of intensity and  $\beta_2$  thermal excitation rate.

The dark decay of LIA is associated with the loss of shallow and deep trap population by thermal ionization. In general, the dark-decay process of LIA can be described by a double exponential function<sup>8</sup>

$$\Delta\alpha(t) = a_0 + a_1 \exp(-t/\tau_1) + a_2 \exp(-t/\tau_2), \quad (4)$$

where  $a_0$ ,  $a_1$ , and  $a_2$  are the constants and  $\tau_1$  and  $\tau_2$  are the fast and slow decay-time constants for shallow and deep traps, respectively.

### III. EXPERIMENTS

We used  $0^\circ$  cut as-grown undoped and Rh:BaTiO<sub>3</sub> crystals that had dimensions of 6.72 mm × 6.28 mm × 5.5 mm and 6.6 mm × 5.8 mm × 4.2 mm ( $c \times a \times a$ ), respectively. Change in absorption can be induced at one wavelength and observed at different wavelengths. This helps to eliminate the possibility of beam coupling effects that can rise if both beams are coherent. An expanded pump beam  $I_p$  at one wavelength is used to illuminate the crystal as shown in the experimental schematic in Fig. 1.

A weak signal beam  $I_s$  at different wavelengths acts as a probe to study the change in absorption. The outputs of He-Ne (543 and 633 nm) and Ti:sapphire lasers (model 3900S, Spectra-Physics) tuned to 750 and 800 nm were used as the probe beams, whereas pump beams at 514 and 488 nm were obtained from an argon-ion laser (model BeamLok™ 2085, Spectra-Physics). The beam-crossing angle was made small to ensure a complete and uniform overlap of the beams inside the crystal. Transmission of the probe was monitored using computer controlled optical power meter (model, 4832-c multichannel optical power meter, Newport). In all our measurements the probe beam power was fixed at 1  $\mu$ W in the absence of pump beam illumination. The diameter of the probe beam inside the crystal was  $\sim 2$  mm. The intensity of the pump beam was varied from  $\sim 3 \mu$  to 16 W/cm<sup>2</sup> using a compensated neutral density filter (model 925B, Newport). Both beams were made o polarized to minimize beam fanning and scattering effects, which otherwise could be mistaken for absorption. An appropriate color glass filter along with a small pin hole was used to prevent the pump beam from entering the detector.

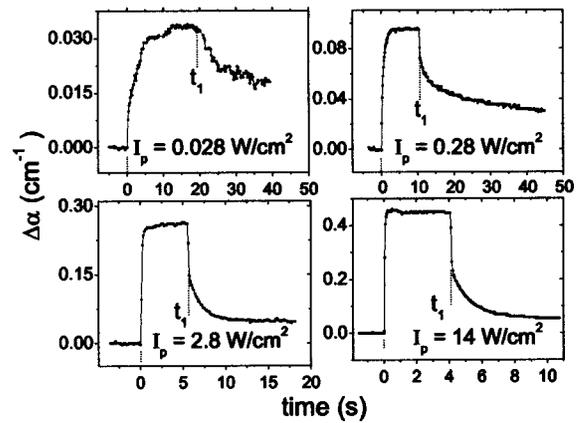


FIG. 2. LIA change for a probe beam at 633 nm in undoped BaTiO<sub>3</sub> crystal when illuminated by pump (488 nm) at different intensities.

### IV. RESULTS AND DISCUSSION

Transmission of the probe beam was monitored after switching the pump beam on at  $t_0$ . Corresponding absorption values can be calculated using Eq. (1). Figure 2 shows a typical result of LIA change and its subsequent dark decay when the pump beam was blocked at  $t_1$ . Absorption buildup curves for both crystals (i.e., the region between  $t_0$  and  $t_1$  in Fig. 2) follow a monoexponential rise as described by Eq. (2). Rise-time constant  $\tau$  was calculated by fitting the experimental data into Eq. (2). We fitted the intensity power coefficients  $x$  according to the relation  $\tau\alpha I^{-x}$  and found that the absorption rise has a sublinear intensity dependence conforming to earlier observations<sup>9</sup> that a steady state in the PR grating (formation and decay) cannot be reached until the LIA reaches the steady state. In all the measurements, the value of  $x$  was found to vary between  $\sim 0.4$  and 1.0 for Rh-doped and  $\sim 0.6$ –2.0 for undoped BaTiO<sub>3</sub> crystal.

At 633 nm probe and 514 nm pump illumination, an anomalous temporal evolution of absorption, as shown in Fig. 3, was observed in Rh:BaTiO<sub>3</sub>. After the pump beam was turned on at  $t = t_0$ , an unexpected light-induced transparency is observed for sometime, which is then followed by the usual absorption rise. However, the light-induced transpar-

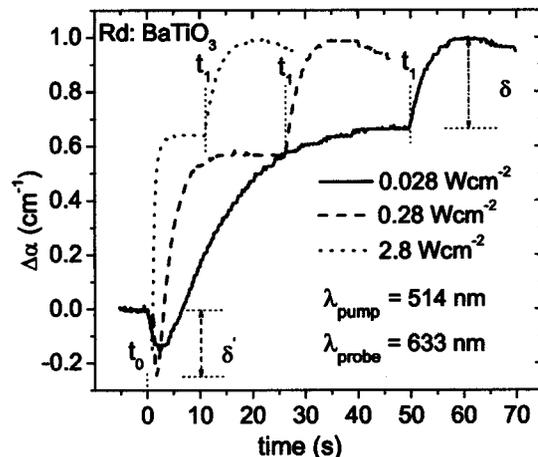


FIG. 3. Normalized temporal evolution of absorption change in Rh:BaTiO<sub>3</sub> at 633 nm probe and 514 nm pump beam at three different pump intensities.

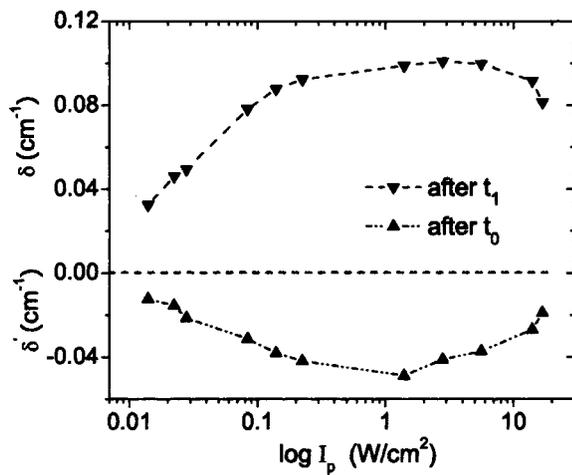


FIG. 4. Plot of anomalous absorption variations versus  $I_p$  for Rh:BaTiO<sub>3</sub> crystal.

ency reported in Refs. 11 and 13 has no resemblance to our observations. Contrary to the behavior shown in Fig. 2, a further absorption increase was noticed even after the pump beam was put off at  $t = t_1$ . For clarity, we have denoted the magnitude of these additional absorption changes by  $\delta$  and  $\delta'$  in the Fig. 3.

The magnitude of these variations as a function of pump intensity is plotted in Fig. 4. We observe a linear change in  $\delta$  up to  $\sim 0.23$  W/cm<sup>2</sup>, beyond which it is almost constant and falls at higher intensities. These anomalous temporal changes in absorption seen only at the 633 nm probe and 514 nm pump in our Rh:BaTiO<sub>3</sub> sample has not been reported so far.

The physical mechanism responsible for this particular behavior is not very clear. But, we speculate that immediately after the pump beam is turned on, a gradual rise in temperature causes an increase in the thermal excitation rate of the secondary traps. Therefore, the number density of available filled traps that can absorb probe photons is also reduced. This results in decreases in absorption as shown in the Fig. 3. The decline in absorption continues for sometime until a thermal equilibrium is attained within the crystal. Similarly, an increase in absorption after blocking the pump beam is a consequence of the drop in the thermal excitation rate of the secondary traps. Temperature-dependent absorption studies<sup>3,10</sup> in the past have shown that the rise in temperature can cause a decrease in absorption, and vice versa. However, we must add that there is no strong evidence against the possibility of additional PR centers or some other crystal defects which could possibly lead to similar results.

Measurements of the steady-state LIA changes at different pump and probe wavelengths for both crystals have also been carried out. Here we summarize only the main results.

- For both the crystals, the magnitude of the absorption change caused by the 488 nm pump was found to be more than the 514 nm pump.
- Unlike undoped BaTiO<sub>3</sub>  $\Delta\alpha$  was found to increase with increases in probe wavelengths for Rh:BaTiO<sub>3</sub>, indicating the different origins of secondary centers in both crystals.

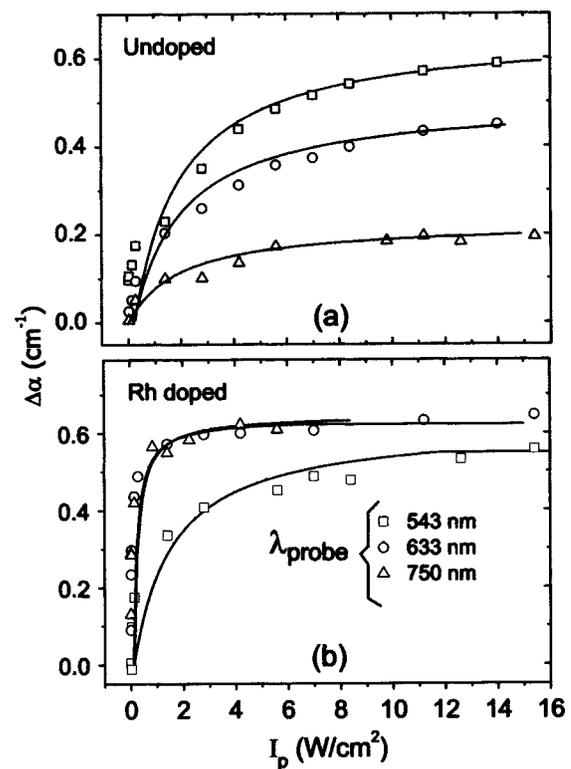


FIG. 5. Steady-state LIA change  $\Delta\alpha$  vs pump beam intensity ( $\lambda_{\text{pump}} = 488$  nm) at different probe wavelengths. Solid lines are theoretical curves based on the two-center model.

- The thermal excitation rates  $\beta_2$  calculated using Eq. (3) were found to be 0.1–0.3 and 0.5–0.6 s<sup>-1</sup> for Rh-doped and undoped BaTiO<sub>3</sub> crystals respectively. These values are considerably smaller than the results reported in Refs. 4 and 6.
- As per the theory of LIA described in Ref. 6 and the fact that thermal ionization rate  $\beta_2$  for undoped crystal is relatively higher than Rh:BaTiO<sub>3</sub>, the pump beam intensity at which the absorption saturates is higher (see Fig. 5) for undoped BaTiO<sub>3</sub> than the Rh-doped crystal.

Absorption decay studies were carried out by monitoring the transmission of the probe beam after switching off the pump beam at  $t = t_1$ . Shown in Fig. 6 is the normalized dark

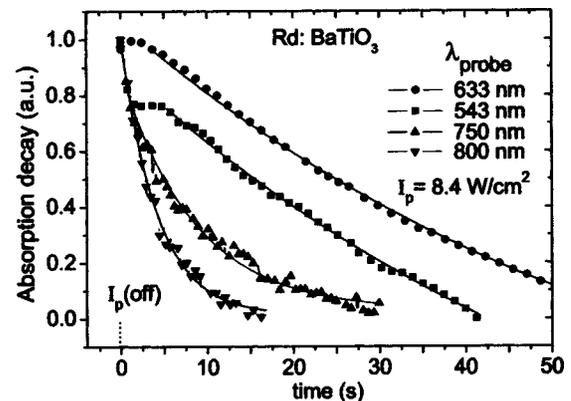


FIG. 6. Typical dark-decay process of  $\Delta\alpha$  observed in Rh:BaTiO<sub>3</sub> for different  $\lambda_{\text{probe}}$  after blocking the pump beam (488 nm).

TABLE I. Decay-time constants for undoped BaTiO<sub>3</sub> at 488 nm (514 nm) pump.

$\lambda_{\text{probe}}$	$\tau_1$ (s)	$\tau_2$ (s)
543 nm	0.1(0.12)	1.25(1.3)
633 nm	0.13(0.12)	1.5(1.4)
750 nm	0.57(0.53)	...

decay of absorption for Rh:BaTiO<sub>3</sub>. The symbols represent the experimental data, whereas the solid curves are fits obtained using Eq. (4). We note that the dark decay at the 543 nm probe and 488 nm pump occurs in two separate steps. It shows an initial fast relaxation having two time constants  $\tau_1 \sim 50$  ms and  $\tau_2 \sim 0.7$  s, followed by a slow relaxation with longer time constant  $\sim 60$  s. At the 633 nm probe wavelength a slow mono-exponential decay with  $\tau_2 \sim 62$  s alone was observed. Decay at wavelengths 750 and 800 nm was again found to be monoexponential with  $\tau_2 \sim 7.25(6.25)$  s and  $\tau_2 \sim 5.82(5.42)$  s, respectively, at 488 nm (514 nm) pump. The values reported in Refs. 12 and 13 ( $\sim 6-10$  s) and Ref. 14 ( $\sim 7$  s) are in close agreement with our measurements. For undoped BaTiO<sub>3</sub> crystal we observed a double exponential decay at 543 and 633 nm probe wavelengths, whereas the decay at 750 and 800 nm was monoexponential. The results are listed in Table I. The reported values of two decay constants in Ref. 15 closely match our results.

## V. CONCLUSION

In summary, we have presented experimental studies on the temporal and steady-state behavior of light-induced absorption in Rh and undoped BaTiO<sub>3</sub> crystals. The buildup and dark-decay rate of absorption at different wavelengths and intensities were obtained. An anomalous absorption change in Rh:BaTiO<sub>3</sub> seen only at 514(633) nm pump-(probe) may be attributed to light-induced thermal effects.

However, this case will be further examined separately by performing temperature-dependent absorption studies in the near future. Finally, we want to emphasize that LIA can greatly influence the dynamics of holographic grating recording in photorefractive crystals. Therefore, it is of utmost importance to incorporate the effect of LIA before using the crystal in any device application.

## ACKNOWLEDGMENTS

The authors would like to acknowledge Dr. S. Chatterjee for useful discussions and valuable suggestions. This work was carried out at the CREST (Center for Research and Education in Science and Technology) campus of the Indian Institute of Astrophysics, Bangalore.

- <sup>1</sup>*Topics in Applied Physics: Photorefractive Materials and Their Applications I and II*, edited by P. Günter and J.-P. Huignard (Springer, Berlin, 1989), Vols. 61 and 62.
- <sup>2</sup>N. V. Kukhtarev, V. B. Markov, S. G. Odulov, M. S. Soskin, and V. L. Vinetskii, *Ferroelectrics* **22**, 949 (1979).
- <sup>3</sup>A. Motes and J. J. Kim, *J. Opt. Soc. Am. B* **4**, 1379 (1987).
- <sup>4</sup>G. A. Brost, R. A. Motes, and J. R. Rotgé, *J. Opt. Soc. Am. B* **5**, 1879 (1988).
- <sup>5</sup>L. Holtmann, M. Unland, E. Krätzig, and G. Godefroy, *Appl. Phys. A: Solids Surf.* **51**, 13 (1990).
- <sup>6</sup>K. Buse and T. Bierwirth, *J. Opt. Soc. Am. B* **12**, 629 (1995).
- <sup>7</sup>L. Holtmann, *Phys. Status Solidi A* **113**, K89 (1989).
- <sup>8</sup>Z. G. Zhang, M. Sprenger, H. J. Eicheler, Aimin Yin, Panming Fu, D. Z. Shen, X. Y. Ma, and J. Y. Chen, *Proc. SPIE* **3554**, 216 (1998).
- <sup>9</sup>D. Mahgerefteh and J. Feinberg, *Phys. Rev. Lett.* **64**, 2195 (1990).
- <sup>10</sup>Y. Li, D. Trung, B. Zhang, Y. Segawa, and T. Itoh, *Proc. SPIE* **3554**, 33 (1998).
- <sup>11</sup>J. Y. Chang, C. R. Chinjen, S. H. Daun, C. Y. Haung, R. H. Tsou, J. N. Cheng, and C. C. Sun, *Opt. Commun.* **153**, 106 (1998).
- <sup>12</sup>G. A. Brost and R. A. Motes, *Opt. Lett.* **15**, 538 (1990).
- <sup>13</sup>M. Kaczmarek, G. W. Ross, R. W. Eason, M. J. Damzen, R. Ramos-Garcia, and M. H. Garrett, *Opt. Commun.* **126**, 175 (1996).
- <sup>14</sup>L. Corner, R. Ramos-Garcia, A. Petris, and M. J. Damzen, *Opt. Commun.* **143**, 165 (1997).
- <sup>15</sup>H. Song, S. X. Dou, M. Chi, H. Gao, Y. Zhu, and P. Ye, *J. Opt. Soc. Am. B* **15**, 1329 (1998).