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Temporal behavior of the intensity-dependent absorption in photorefractive $BaTiO_3$

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We report measurements of response time for the intensity-dependent absorption in a single $BaTiO_3$ crystal under conditions in which no photorefractive effect can be observed. The speed of response is found to increase with increasing intensity but in a sublinear fashion similar to that observed for photorefractive decay. This experiment provides unambiguous evidence for the intensity-dependent absorption in $BaTiO_3$ crystals.

Since the observation of an anomalous behavior of $BaTiO_3$ in a two-beam coupling experiment by Klein and Valley[1] and the observation of asymmetric coupling gain coefficients with respect to the sense of the c axis,[2] we have reported that the anomalous behavior and the nonsymmetric behavior of the coupling gain may be due to the intensity-dependent absorption coefficient.[3] Models[4,5] have been proposed to explain the anomalous behavior, but the exact source of the anomaly is not well understood at present. We have shown that the intensity-dependent absorption is consistent with the presence of secondary centers.[6] These are intermediate-level traps that are highly ionized at room temperature but can compete for free carriers generated by photoexcitation of the deep traps. The secondary centers then become populated and are available for additional absorption and photorefraction. Our mathematical analysis shows that this model predicts both an intensity-dependent absorption and an intensity-dependent photorefractive effect (PRE). It is the purpose of this Letter to present additional experimental evidence that clearly demonstrates the existence of intensity-dependent absorption in $BaTiO_3$.

The experimental setup that we used to measure the response of the intensity-dependent absorption is shown in Fig. 1. An unpoled $BaTiO_3$ crystal that measured 4 mm × 5.5 mm × 6.5 mm was used. An argon-ion laser operating at $\lambda = 514$ nm was used to pump the crystal and to induce the absorption. This pump beam, controlled by an electromechanical shutter with 3-msec response time, completely illuminated the crystal. A low-power He–Cd laser with a wavelength of 442 nm and intensity less than 1.4 mW/cm^2 was used to observe the induced absorption. Both the argon-ion and He–Cd beams had vertical polarization. The crystal c axis was along the signal-beam propagation direction.

The purpose for using an unpoled crystal and two different lasers was to eliminate the possibility of observing a PRE. Unpoled crystals usually do not exhibit a PRE because the pole domains have random orientation. The PRE requires that the crystal have uniform polarization. Since the PRE is a slow process it also requires a stationary or slowly varying intensity interference pattern, depending on the intensity of the writing beams. At the intensity levels that we used the PRE should not respond any faster than about 5 Hz. The beat frequency or interference-pattern oscillation rate created by the two lasers used in this ex-

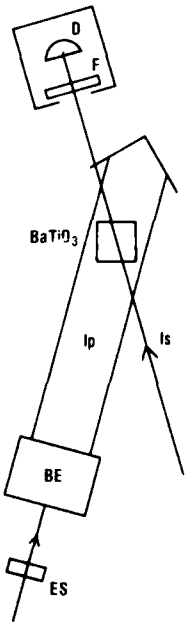

Fig. 1. Experimental setup for measuring the time response of the intensity-dependent absorption. ES, electromechanical shutter; BE, beam expander; I_p, pump beam; I_s, signal beam; F, 440-nm filter; D, detector.

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periment was 9.5×10^{13} Hz. Since this is well above 5 Hz, the interference pattern cannot produce a PRE.

Care was taken to ensure that no scattered light from the pump beam entered the detector. This scattered light can completely mask out the reduction in transmitted signal-beam intensity caused by the induced absorption. To eliminate the scattered light two steps were taken. First, a 440-nm bandpass filter with 30% transmission at 442 nm and low transmission at the argon wavelengths was placed between the detector and the crystal. Second, a lightproof shield was placed around the detector–filter arrangement. Several tests were run with the signal beam (I_s) off to verify that scattered light was not contributing to the detector output.

Figure 2 shows the results of I_s measurements while the pump beam (I_p) was turned on and then off. The data were taken by a digital storage oscilloscope, which, along with the electronic shutter, was controlled by a computer. Two curves are shown for two different pump-beam intensities. Each curve represents an average of 10–30 runs. When the pump beam is turned on, we see a decrease in the transmitted signal beam due to an increase in absorption. Absorption time response and magnitude measurements were taken from these curves.

Figure 3 shows the absorption rise time τ_R decreasing and the induced absorption α_I increasing as the pump-beam intensity is increased. The experimental data for α_I were curve fitted by using third-order regression. The resulting curve is similar to our other reported experimental and theoretical results.[3,6]

The experimental data for τ_R were curve fitted by using first-order regression. The resulting curve can be described by $\tau_R = 18 I_p^{-0.41}$, which shows that the absorption rise has a sublinear intensity dependence. This could cause the photorefractive rise time to have a sublinear intensity dependence, since a steady state in the photorefractive grating cannot be reached until the absorption has reached steady state. Other researchers have suggested that the rise and decay times of the photorefractive gratings are comparable.[7] If this assumption is correct, the sublinear response of the intensity-dependent absorption may explain the observed sublinear dependence of the photorefractive

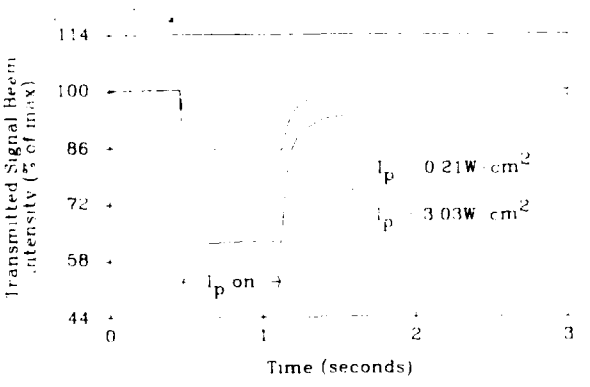

Fig. 2. Signal-beam intensity versus time as the pump beam is turned on and then off. Induced absorption in the signal beam caused by the pump beam is shown.

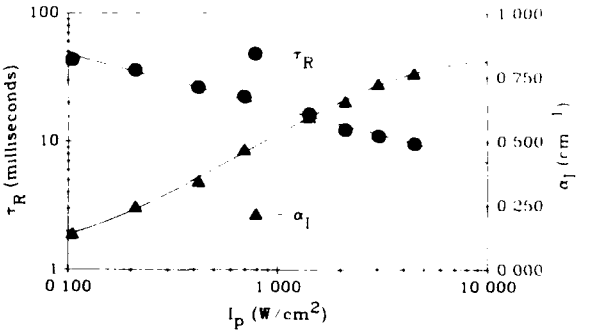

Fig. 3. Rise time and magnitude of the intensity-dependent absorption versus pump-beam intensity.

decay on intensity.[7] This is the subject of further research.

The slope of the measured τ_R versus I_p curve was dependent on where the signal beam intersected the crystal. It was always less than 1, with a typical value of -0.5. For another crystal that we studied, the typical value was -0.6.

In Fig. 2 we also notice the appearance of two decay times. There are two possible explanations for this. First, it is possible that there exist two different secondary trap centers, both contributing to the intensity-dependent absorption, one more shallow than the other. The second and more probable explanation is that the decay times are due to the nonuniform intensity distribution of the signal beam in the crystal. The low-intensity regions will produce a slower time response than the higher-intensity regions and give the appearance of multiple time constants.

In this crystal we did not study the effects of changing the relative polarization of the two beams with respect to each other or to the crystal. However, our experiments with poled crystals and uncorrelated beams have shown a slightly slower response of the induced absorption when one beam has ordinary polarization and the other has extraordinary polarization.

No beam fanning was observed in the crystal during the experiment. Beam fanning in the pump beam will build gratings that may scatter the signal beam even though it is not the same frequency. If this happens, the result can be misinterpreted as an intensity-dependent absorption. Beam fanning in the signal beam produces a phase-conjugate reflection and reduces the transmitted intensity until the pump beam comes on. The pump beam partially washes out the grating produced by the fanning and allows the signal beam to increase. We have observed this effect in poled crystals with uncorrelated beams. It gives the appearance of an absorption with an inverse dependence on intensity. Also, no self-defocusing of the beams was observed. Fanning and self-defocusing effects have photorefractive origins, and the probability of their existence is greatly reduced by the use of an unpoled crystal.

The magnitude of the induced absorption is much smaller at longer wavelengths. This has been ob

served before.[3] Also, the rise and decay times of the absorption increase as the wavelength is increased. However, the change that we observed in the rise time was small and inclusive.

In summary, we have measured the time response of the intensity-dependent absorption in $BaTiO_3$. We observed that this absorption exists in an unpoled crystal and can be induced by light of one wavelength and measured at another wavelength. This demonstrates that it is not a PRE. We also found that the speed of response for the intensity-dependent absorption increases with intensity in a sublinear way similar to that observed in photorefractive decay. We believe that these observations can be explained by a multiple-trap model. With futher research these observations may lead to a better understanding of the PRE in $BaTiO_3$.

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