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Speed of the photorefractive effect in a $BaTiO_3$ single crystal

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We present data on the speed of light-induced refractive index changes in a $BaTiO_3$ single crystal. The light-induced erasure rate of a refractive index grating is shown to depend on optical intensity as I^x where $x < 1$. The exponent x depends weakly on temperature and increases from 0.62 ± 0.02 to 0.71 ± 0.02 when the temperature is varied between 12 and 40 °C. The sublinear dependence of rate on intensity implies that higher optical intensity is required to achieve high-speed operation of $BaTiO_3$ devices than previously thought. The dark erasure rate has an anomolously strong temperature dependence; it increases by a factor of 50 over the same temperature range. We have also determined that the number density of photorefractive charge carriers is 6×10^{16} cm^{-3} in this crystal.

INTRODUCTION

$BaTiO_3$ crystals have recently been used for optical phase conjugation,[1] optical image processing,[1] interferometry,[2] and optical gyroscopes.[3] These applications all depend on the photorefractive effect, in which nonuniform illumination induces a semipermanent change in the refractive index of the crystal.[4] The rate of this refractive-index change was previously thought to increase *linearly* with the intensity of the incident light.[5,6] However, we show here that in $BaTiO_3$ the speed of erasure of a photorefractive index change depends sublinearly on the intensity I of the incident light, i.e., as I^x where $x < 1$. For a given optical intensity, the time required to write a photorefractive grating is comparable to the time required to erase the grating. This implies that high speed operation of a photorefractive device using $BaTiO_3$ requires more optical intensity than would be calculated by the linear extrapolation of low intensity rates. We also show that the photorefractive speed depends on the temperature of the $BaTiO_3$ crystal. According to the results presented, the rate of grating erasure is given by

$$\Gamma = \Gamma_{\text{dark}} + \Gamma_{\text{light}}, \tag{1a}$$

$$\Gamma_{\text{dark}} \propto f(k_g) \exp(-T_0/T), \tag{1b}$$

$$\Gamma_{\text{light}} \propto f(k_g)\left(\frac{I}{I_0}\right)^x. \tag{1c}$$

Here T is the crystal temperature and I is the erasure intensity. We take $I_0 = 1$ W/cm^2. The parameters T_0 and x are determined by experiment. The function $f(k_g)$ contains the dependence of rate on the grating spatial frequency k_g as discussed below.

EXPERIMENTAL DESIGN

We present data on the speed of light-induced refractive index changes in a photorefractive crystal of $BaTiO_3$. The experiments were performed by illuminating the $BaTiO_3$ crystal with two optical beams of the same wavelength. These "writing" beams created an intensity interference pattern where they intersected in the crystal. The spatial frequency of the interference pattern was varied by changing the crossing angle between the two writing beams. According to the present models[5,6] of the photorefractive effect, charges (of unknown origin in $BaTiO_3$) migrated in the presence of the light pattern to create a spatially varying electrostatic field, which in turn altered the refractive index of the crystal by the Pockels effect. The resulting refractive-index variation had a component with the same spatial periodicity as the intensity interference pattern, and was monitored by another optical beam, the "reading" beam, incident at the Bragg angle of the refractive index pattern. If the writing beams were removed, the refractive-index grating persisted in the dark, although it was slowly erased both by the reading beam and by thermal charge transport. The grating could be rapidly erased by flooding the crystal with light from a uniform intensity erasing beam. By measuring the grating decay rate as a function of the intensity of the erasing beam, both the dark and the light-induced erasure rates were determined.

The $BaTiO_3$ sample was mechanically and electrically poled into a single domain crystal of dimensions $4.3 \times 5.4 \times 5.6$ mm^3 with the c axis parallel to the 5.4-mm dimension. The crystal was contained in a quartz spectroscopic cuvette filled with silicone oil of refractive index $n = 1.4$. A calibrated thermistor temperature sensor and proportional feedback control was used to regulate the oil bath temperature to within ± 0.1 °C of a set temperature.

Figure 1 shows the arrangement of the crystal and the various optical beams. The writing beams (no. 1 and no. 2), with optical wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ respectively in the crystal, created an intensity interference pattern with grating wavevector $\mathbf{k}_g = \mathbf{k}_1 - \mathbf{k}_2$, where the magnitude of the grating wavevector is

$$k_g = (2\omega/c) \sin \theta_{\text{air}}, \tag{2}$$

where ω is the optical circular frequency, c is the speed of light in vacuum, and $2\theta_{\text{air}}$ is the full angle of intersection of the writing beams in air. The c axis of the $BaTiO_3$ crystal was aligned parallel to $\mathbf{k}_g$. The reading beam (no. 3) incident on the resulting photorefractive grating at the Bragg angle, was deflected to form the signal beam (no. 4). All of these optical beams were polarized perpendicular to the plane of inci-

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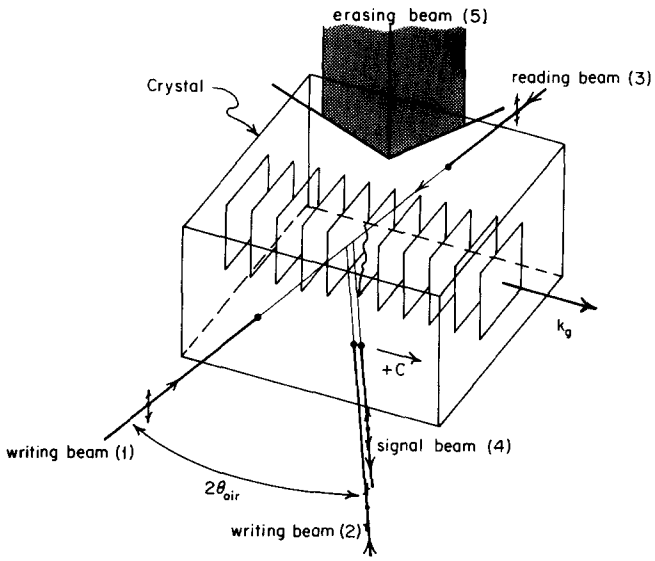

FIG. 1. The two optical writing beams (no. 1 and no. 2) create a refractive-index grating in a crystal of $BaTiO_3$. The crystal is aligned with its c axis perpendicular to the grating planes as shown. The optical reading beam no. 3 is Bragg deflected by the grating and produces the signal beam no. 4, which propagates back along writing beam no. 2 and is separated by a beamsplitter (not shown). The optical erasing beam no. 5 is incident from the top of the crystal and is uniform in intensity. All beams are from a single mode argon-ion laser at 514.5 nm unless otherwise specified.

dence so as to be ordinary rays in the crystal in order to minimize two-beam coupling. An erasing beam (no. 5) was incident from a direction perpendicular to the other four beams, so as to minimize scattering off of any refractive index grating formed by the other beams. The erasing beam was polarized parallel to the c axis of the crystal to minimize scattering into the signal beam (no. 4), (although the erasing rate was unaffected by the choice of polarization of the erasing beam) and entered the top of the crystal after first passing through the silicone oil meniscus which had been flattened by a thin plastic sheet with a hole for the beam.

To determine the light-induced erasure rate, a refractive index grating was first created by illuminating the $BaTiO_3$ crystal with the two writing beams and with the reading beam. After five seconds the writing beams were turned off and the erasing beam was turned on by mechanical shutters. The signal was then observed to decay with time. The decay curve was digitized, the process repeated, and the average of typically five decay curves was fit by the method of least squares to a simple exponential decay.The decay or erasure rate is defined as the reciprocal of the exponential decay time constant. In all cases the observed decay curves were well fit by a single exponential decay. (No evidence was seen for multiple decay time constants, although an initial fast decay with a time constant shorter than 1 ms could not have been resolved due to the finite closing time of the mechanical shutter.)

All optical beams were derived from a single-mode argon ion laser at 514.5 nm. The spot size of beams 1, 2, and 3 was 200 μm ($1/e^2$) in diameter for the light-induced erasure experiments. The spot size of the erasing beam far exceeded the size of the grating region in the crystal, so that the erasing intensity was uniform over the grating. The intensities of the various beams were: the writing beams $I_1 = I_2 = 4$ W/cm^2, the reading beam $I_3 = 10^{-4}$ W/cm^2, and the erasing beam varied over the range $I_5 = 10^{-4}$–10^2 W/cm^2. The observed decay rate was independent of the relative intensity of the writing beams. The intensity of the reading beam was sufficiently weak so as to have no measurable effect on the grating decay rate compared to the light-induced decay rate for erasing beam intensities exceeding 10^{-3} W/cm^2. To measure the dark decay rate no erasing beam was used, the writing and reading beams were increased in diameter to 3 mm ($1/e^2$) and the intensity of the reading beam was reduced to $I_3 = 2\times10^{-10}$ W/cm^2, so as to have no measureable effect on the dark decay rate. The diffraction efficiency $R = I_4/I_3$ was deliberately made less than 20% (by using a small interaction length and ordinary polarization of the beams) so that $\sqrt{R}$ was proportional to the light-induced refractive-index change.[7]

RESULTS

Light-induced erasure

The results of light-induced erasure experiments at crystal temperatures of 16, 24, and 40 °C are summarized in Fig. 2. The dark rate is subtracted from the measured rate to obtain the light-induced erasure rate. A log-log plot of this light-induced decay rate is linear over four decades of the erasing beam intensity, deviating only when the erasing beam intensity approaches the reading beam intensity of 10^{-4} W/cm^2. At a crystal temperature of 16 °C the slope of the plot is $x = 0.62$ [see Eq. (1)], so that the grating erasure rate is seen to be proportional to $I^{0.62}$. The results of light-induced erasure rate measurements at other temperatures are summarized in Table I, where the exponent is seen to increase with crystal temperature, although never reaching linear dependence.[8]

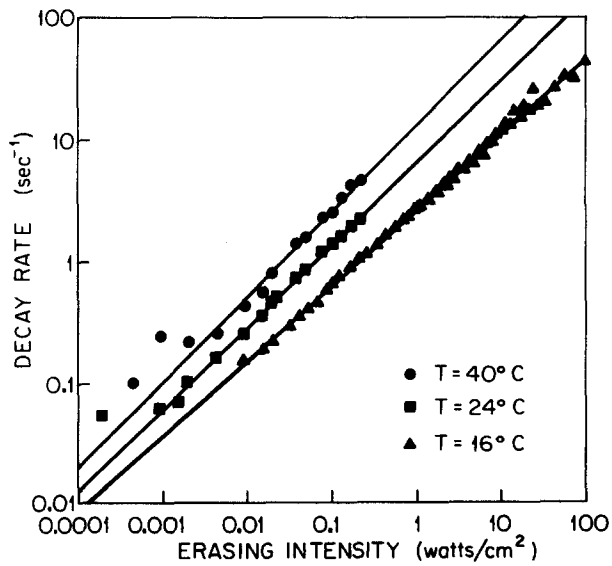

FIG. 2. Log-log plots of the light-induced decay rate Γ_{light} vs the intensity of an erasing beam for crystal temperatures of 16, 24, and 40 °C. This data was taken at a crossing angle of $2\theta_{air} = 30°$. The dark decay rate Γ_{dark} has been subtracted. The $T = 16$ °C plot is linear over four orders of magnitude, from 0.01 W/cm^2 to 100 W/cm^2, and shows a power law dependence of $I^{0.62}$.

TABLE I. The power law exponent x of the Eq. (1) at various temperatures determined from the slopes of Fig. 2.

Temperature (°C)	12°	16°	24°	40°
Exponent (x)	0.62 ± 0.02	0.62 ± 0.01	0.68 ± 0.02	0.71 ± 0.02

Present models[5,6] of the photorefractive effect do not account for the nonlinear dependence of speed on optical intensity seen here. Townsend and Lamaccia[9] observed similar nonlinear dependence of the grating writing rate on optical intensity over a much smaller range of intensities, but the model they proposed to account for it has since proved inadequate to explain other aspects of the photorefractive effect. Valley's analysis[10] of Kukhtarev's model with a linear photoconductivity[6] predicts a linear intensity dependence for $BaTiO_3$ at these intensitites. Nonlinear intensity dependence of the erasure rate can, however, be derived by using a nonlinear photoconductivity in the original equations of Kukhtarev's model.

Dark erasure

The dark decay rate had a pronounced temperature dependence. Figure 3 shows the dramatic fiftyfold increase in the measured dark decay rate as the temperature of the $BaTiO_3$ crystal was increased from 12 to 40 °C. (This temperature range was fixed at the lower limit by a sometimes destructive orthorhombic/tetragonal phase transition at ~ 5 °C, and at the upper limit by the tendency for the crystal to depole as the tetragonal/cubic transition at ~ 128 °C is approached.)

Figure 4 is a log plot of the dark decay rate versus reciprocal absolute temperature. The data can be well fit by parallel straight lines over the limited termperature range used, giving the temperature dependence of $e^{-T_0/T}$ in Eq. (1). The three different lines in Fig. 4 correspond to three different values for the crossing angle between the two writing beams. From the slope of the lines the value of T_0 in Eq. (1) is $T_0 = 11\ 800 \pm 400$ °K. This data used writing beams at 514.5 nm. Similar data using writing beams at 476.5 nm yielded the same value for the parameter T_0.

DISCUSSION

If the hopping model of Ref. 5 is modified to include a dark decay rate, then the dark decay data of Fig. 5 can be used to determine a charge carrier number density. This model shows that both the dark and the light-induced decay rates are then proportional to the factor $f(k_g)$ of Eq. (1) given by

$$f(k_g) = (1 + k_g^2/k_0^2), \tag{3}$$

where the grating wavevector k_g is given by Eq. (2) and k_0 is defined by[5]

$$k_0^2 = Ne^2/(\epsilon\epsilon_0 kT). \tag{4}$$

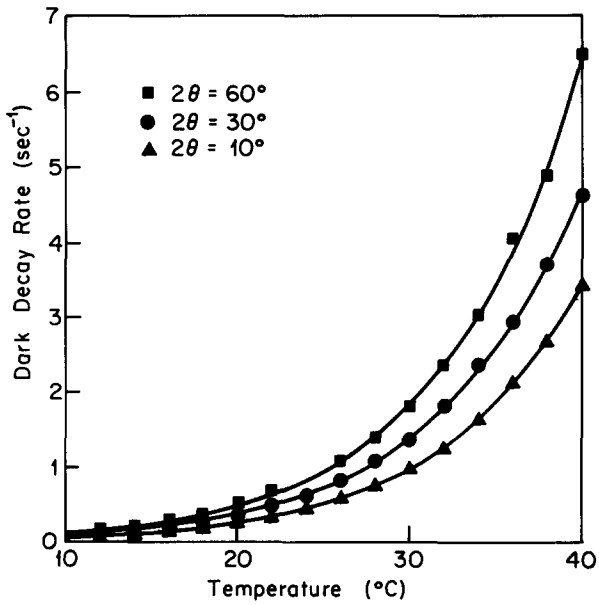

FIG. 3. Measured dark decay rate vs temperature. Note the fiftyfold increase over a small (~ 30 °C) temperature range around room temperature. The three curves correspond to data taken at three different crossing angles of the writing beams. The solid lines are theoretical fits according to Eq. (1b).

Here N is the number density of photorefractive charges available for charge migration, e is the charge of a charge carrier, ϵ is the dc dielectric constant (along the direction of the grating wavevector), ϵ_0 is the permitivity of free space, and kT is the thermal energy of the lattice.

By determining k_0 the number density of charges can be obtained. From Eq. (3) k_0 can be found from a plot of the dark decay rate vs k_g^2, as is shown in Fig. 5. The ratio of the intercept to the slope of Fig. 5 yields

$$k_0 = 1.4\omega/c = 1.7 \times 10^5\ \text{cm}^{-1}.$$

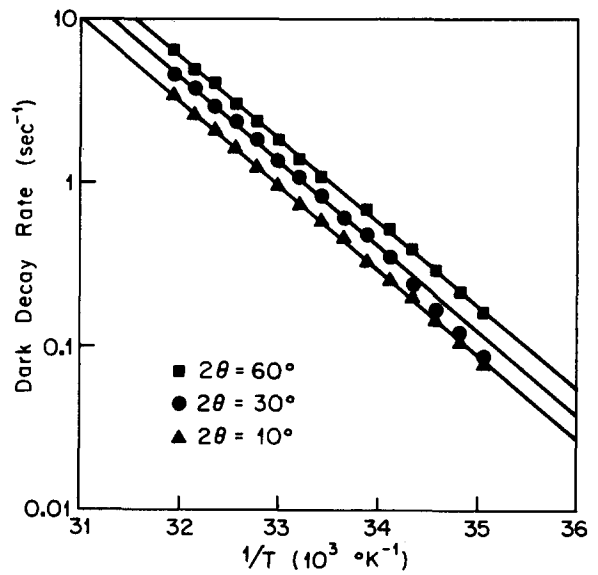

FIG. 4. Log of the dark decay rate vs reciprocal absolute temperature. Three different crossing angles yield straight line plots with the same slope, indicating the $e^{-T_0/T}$ dependence of Eq. (1), with $T_0 = 11\ 800 \pm 400$ °K. The solid lines are least squares fits to the data.

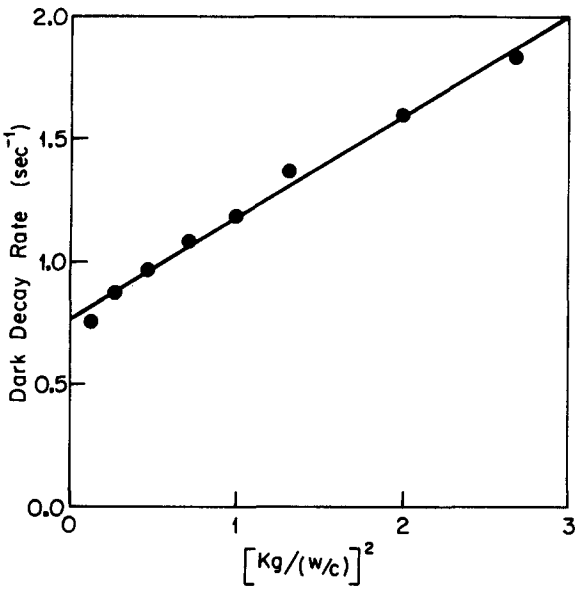

FIG. 5. Plot of the dark decay rate vs spatial frequency squared (k_g^2) of the refractive index grating at a crystal temperature of 24 °C. The intercept of this plot yields the dark conductivity $\sigma = 10^{-11}\,(\Omega\,\mathrm{cm})^{-1}$. The ratio of the intercept to the slope yields the value of $k_0 = 1.4\,(\omega/c)$, and hence a value for the number density of charge carriers of $N = 6\times 10^{16}\,\mathrm{cm}^{-3}$ in this $BaTiO_3$ crystal.

The dc dielectric constant measured with a capacitance bridge was $\epsilon \sim 250$. Taking the dc dielectric constant to be $\epsilon = 150$, assuming singly charged carriers $e = 1.6\times 10^{-19}$ C and $T = 24$ °C, Eq. (4) gives a charge carrier number density of $N = 6\times 10^{16}\,\mathrm{cm}^{-3}$.

The dark decay rate data shown in Fig. 5 were obtained with writing beams at 514.5 nm. Experiments using steady-state two-beam coupling at 514.5 nm and at 457.9 nm gave the same value for the charge carrier number density while coupling measurements at 1090 nm gave a density a factor of five smaller.[11] The sign of the charge carriers was found to be positive. The number density at 514.5 nm is a factor of three larger than that measured previously[5] in another $BaTiO_3$ crystal, and explains why the present crystal works so well for self-pumped phase conjugation.[12]

In the limit $k_g \to 0$, the dark erasure rate Γ_{dark} approaches the dielectric relaxation rate $\Gamma_{\mathrm{diel}} = \sigma/\epsilon\epsilon_0$. From the intercept of Fig. 5, $\Gamma_{\mathrm{dark}} = 0.56\ \mathrm{sec}^{-1}$ (as $k_g \to 0$) which gives a dark conductivity of $\sigma = 10^{-11}\,(\Omega\,\mathrm{cm})^{-1}$. The dark conductivity was also measured directly using silver print electrodes on the crystal faces, and gave a steady-state dark conductivity of $\sim 6\times 10^{-12}\,(\Omega\,\mathrm{cm})^{-1}$, which is in reasonable agreement with the results above. A similar attempt to measure the dc photoconductivity directly proved unsuccessful due to a photoinduced voltage that interfered with the measurement.

In conclusion, we have shown that the measured dark erasure rate of a photorefractive grating in a $BaTiO_3$ crystal is strongly temperature dependent. The temperature dependence is of the form $e^{-T_0/T}$, with $T_0 = 11\,800 \pm 400$ °K. A previous measurement[9] of the dark erasure rate in a similar $BaTiO_3$ crystal showed the same temperature dependence, but with $T_0 = 8000$ °K.

We have also shown that the measured speed of erasure (or creation) of a photorefractive grating increases less than linearly with the optical intensity I. This explains the results of Lam *et al.*[13] who observed a 20 ns response time with a $BaTiO_3$ crystal illuminated by a pulsed laser with an optical intensity of 23 MW/cm^2. This reponse time was an order of magnitude slower than predicted by a *linear* extrapolation of the low intensity rate. However their data is consistent with a *sublinear* optical intensity dependence of $I^{0.8}$. Subsequent measurements at low optical intensity on the same $BaTiO_3$ crystal have since confirmed this intensity dependence.[8] We have shown here that the sublinear intensity response can also vary with temperature, and becomes more linear with increasing temperature. Kratzig measured the photoconductivity in several $BaTiO_3$ crystals, and also observed a power law sublinear intensity dependence, with exponents ranging from 0.6 to 0.9, depending on the sample.[14] Therefore, it may be possible to prepare $BaTiO_3$ crystals with response times nearly linear in optical intensity, well into the nano-second regime.

ACKNOWLEDGMENTS

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