Photorefractive dark conductivity in Cr-doped strontium barium niobate

Koichi Sayano, George A. Rakuljic, Aharon Agranat, and Amnon Yariv

Department of Applied Physics, California Institute of Technology, Pasadena, California 91125

Ratnakar R. Neurgaonkar

Rockwell International Science Center, Thousand Oaks, California 91360

Received August 15, 1988; accepted February 22, 1989

We present the theoretical and experimental results of photorefractive two-beam coupling in Cr-doped strontium barium niobate:60, using thermal excitation (i.e., dark conductivity) to model the experimentally observed temperature-dependent behavior of the two-beam coupling constant and response time.

Certain strontium barium niobate (SBN) crystals have previously been observed to exhibit increased photorefractive gain as the crystal temperature is lowered below room temperature and away from its ferroelectric phase transition near 345 K. This effect was reported for a doubly Ce- and Ca-doped $Sr_{0.6}Ba_{0.4}Nb_2O_6$ crystal over the temperature range of 243–313 K and in Cr-doped GaAs from 280 to 400 K.[1,2] This was also recently observed in a Cr-doped $Sr_{0.6}Ba_{0.4}Nb_2O_6$(SBN:60:Cr) sample. In this Letter we present the experimental results of the temperature-dependent behavior of the photorefractive properties of SBN:60:Cr and compare them with a theoretical model obtained by incorporating dark-conductivity effects in the Kukhtarev equations for photorefractive two-beam coupling.

The single-crystal SBN:60:Cr sample was grown at Rockwell International Corporation using the Czochralski method and was polished to an optical-quality cube approximately 6 mm on each side. The crystal appeared pale green in color, which we attribute to the Cr^{3+} oxidation state of the dopant. The photorefractive two-beam coupling constant was obtained by measuring the beam intensities before and after the beams passed through and interacted inside the crystal of length l. The 514.5-nm line of the Ar-ion laser was used, with the input-beam intensity ratio $I_1(0)/I_2(0) = 10$ and $I_1(0) = 0.15\ \mathrm{W/cm^2}$. Both beams were horizontally polarized and oriented so that the grating wave vector was parallel to the c axis. The grating period was approximately 1 μm. Given these values of $I_1(0)$, $I_2(0)$, $I_1(l)$, and $I_2(l)$, the coupling constant is given by

$$\Gamma = \frac{1}{l}\ln\left[\frac{I_2(l)}{I_1(0)}\frac{I_1(l)}{I_2(0)}\right], \tag{1}$$

if we assume that the effective interaction length is equal to the physical length of the crystal. The time dependence of the two-beam coupling constant $\Gamma(t)$ was used to obtain the characteristic rise time τ of the material, if an exponential fit of the form $\Gamma = [\Gamma(t \to \infty)][1 - \exp(-t/\tau)]$ is assumed.

The experimental steady-state value of Γ as a function of temperature is shown in Fig. 1. The photorefractive coupling constant increased from 0.9 $\mathrm{cm^{-1}}$ at 313 K to 5.7 $\mathrm{cm^{-1}}$ at 233 K. Beam fanning was observed, although it was not so pronounced as in some SBN:60:Ce samples with a larger Γ. Limitations of the cooling chamber prevented us from going to lower temperatures. However, based on previous theory the crystal should have a positive temperature dependence $d\Gamma/dT > 0$ since the dielectric constant increases sharply near the phase-transition temperature and $\Gamma \propto T/(T + B/\epsilon)$, where $B = e^2N_A/k_Bk^2 = \text{const.}$ Therefore we expect to find a competing phenomenon with a negative temperature dependence near the phase transition to explain the observed negative temperature dependence of Γ over this temperature range.

Kukhtarev *et al.*[3–5] described a functional dependence of Γ and τ for photoexcited states. Inclusion of the effect from thermally excited sites, i.e., dark con-

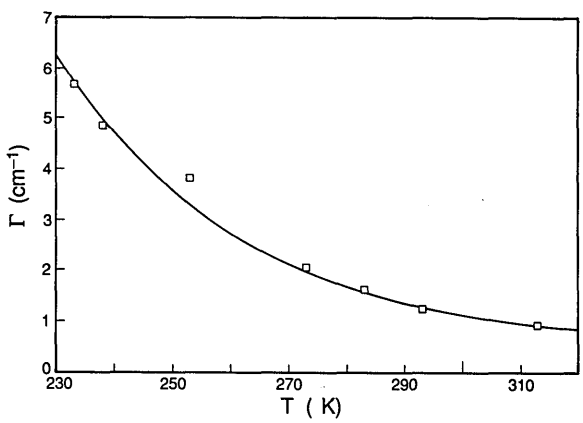

Fig. 1. Experimental values of the steady-state photorefractive two-beam coupling constant Γ of the SBN:60:Cr crystal as a function of temperature. For the experiments $\lambda = 514.5$ nm, $\lambda_g = 1\ \mu$m, $I_1(0)/I_2(0) = 10$, and $I_1(0) = 0.150\ \mathrm{W/cm^2}$. The solid curve represents the theoretical value of Γ versus the temperature for some typical material constants.

0146-9592/89/090459-03$2.00/0

ductivity, results in the thermal excitation and photoexcitation competing for the ionizable donor sites. The resulting equation included an added term in the Kukhtarev rate equation, which results in

$$\frac{\partial N^+}{\partial t} = \left(\frac{sI_0}{h\nu} + \beta\right)N - \gamma n N^+, \tag{2}$$

where β accounts for the thermally excited sites, N^+ and N are the number densities of ionized donors and un-ionized donors, respectively, s is the photoionization cross section of N, n is the number density of electrons, and γ is the two-body recombination rate. The other equations for the band-transport model remain unchanged. Assuming that the SBN crystal can be modeled as a doped semiconductor, we can write the dark excitation term as

$$\beta \propto \exp(-E_\beta/kT). \tag{3}$$

For the stationary grating, the one-carrier/one-species case, the modified theory yields

$$\Gamma = Re\left(\frac{i\omega}{2cl} r_{\text{eff}} n_0{}^3 E_{\text{sc}}\right), \tag{4}$$

where the relevant dark-conductivity temperature dependence is contained in the space-charge electric field E_{sc} and its dependence on the characteristic time t_0. For an applied electric field E_0 and an optical modulation index m, the space-charge field is given by

$$E_{\text{sc}} = iE_N m \frac{(E_0 + iE_d)(1 - e^{-t/\tau})}{[E_0 + i(E_d + E_N)]\left(1 + \frac{h\nu}{sI_0}\beta\right)}, \tag{5}$$

and the response time is given by

$$\tau = t_0 \frac{E_0 + i(E_d + E_\mu)}{E_0 + i(E_d + E_N)}, \tag{6a}$$

where

$$t_0 = \frac{h\nu N_A}{sI_0\left(1 + \frac{h\nu}{sI_0}\beta\right)(N_0 - N_A)}, \tag{6b}$$

and the characteristic fields are

$$E_N = \frac{eN_A}{\epsilon k}\left(1 + \frac{N_A}{N_0 - N_A}\right)^{-1}, \tag{7a}$$

$$E_d = \frac{k_B T k}{e}, \tag{7b}$$

$$E_\mu = \frac{\gamma N_A}{\mu k}. \tag{7c}$$

In these equations N_0 is the number density of impurities, which in this case is dominated by Cr; N_A is the number density of nonphotoactive, compensatory ions; and μ is the mobility. Since β decreases as the temperature decreases, E_{sc} and, consequently, the coupling constant increase through the dark-conductivity factor $(1 + h\nu\beta/sI_0)^{-1}$ as the crystal is cooled. For certain values of the material parameters N_A and N_0, Γ will first tend to increase with temperature, reach a point where the dark-conductivity factor dominates, and then start to decrease after reaching a peak at some optimum temperature T_0.

However, cooling of the crystal also increases its response time since t_0, and therefore the time constant τ, increases. From the equations for τ we find that the crystal becomes faster at higher temperatures even for $\beta = 0$; the presence of the dark-conductivity term further accentuates this effect. This suggests that heating of the crystal will result in a faster response owing to the higher electron conductivity, but as can be seen from the predicted and experimental results this improvement in response time is obtained at a cost of lower photorefractive gain.

We investigated the presence of a significant dark-excitation factor by measuring Γ for low total intensity I_0. If dark excitation were present, it would follow that Γ should be intensity dependent at a low total intensity and constant at a high intensity, as observed by Ewbank *et al.* in Ce-doped SBN, which showed a much lower dark conductivity than did Cr-doped SBN:60.[6] From Eqs. (5) and (7) above, this functional dependence is of the form

$$\Gamma \propto \frac{1}{1 + K/I_0}, \tag{8}$$

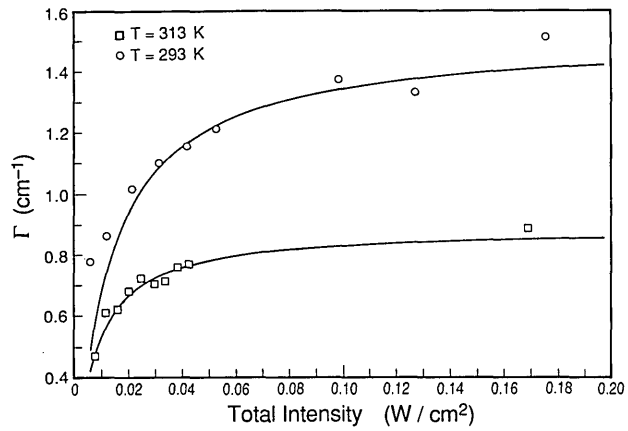

Fig. 2. Γ as a function of the total intensity for 293 and 313 K, with the best fit shown by the solid curves.

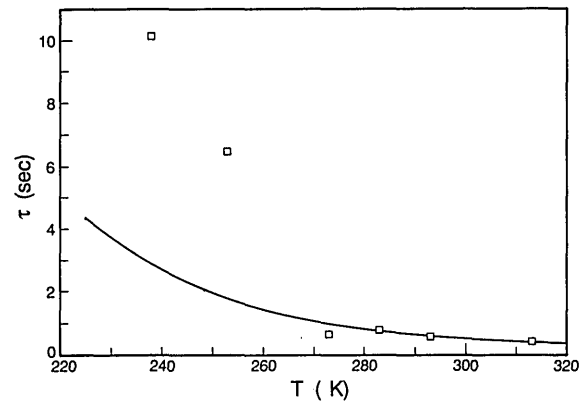

Fig. 3. Response time τ of the SBN:60:Cr crystal as defined by the rise time for Γ to reach $\Gamma_{\text{steady state}}(1 - e^{-1})$. The data are for $\lambda = 514.5$ nm, $\lambda_g = 1$ μm, $I_1(0)/I_2(0) = 10$, and $I_1(0) = 0.150$ W/cm^2.

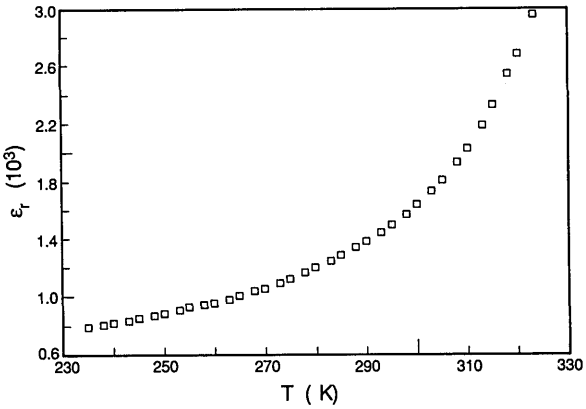

Fig. 4. Experimentally measured relative dielectric constant ϵ_r for the SBN:60:Cr sample obtained by measuring the low-frequency capacitance of the crystal.

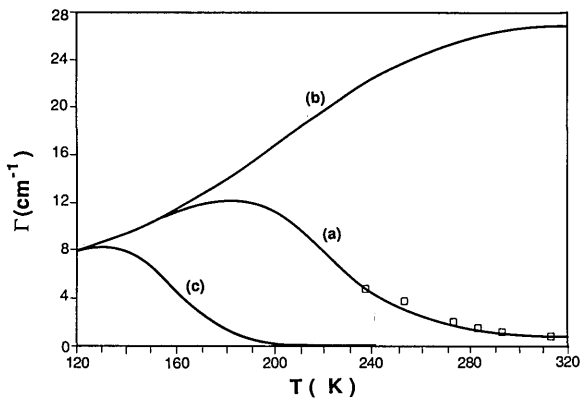

Fig. 5. Theoretical value of Γ from T = 120 K to room temperature showing the peak at some optimum temperature T_0 for curve (a), the best fit to the experimentally obtained points, with $h\nu\beta/sI_0 = 2 \times 10^5$; curve (b), low dark conductivity, with $h\nu\beta/sI_0 = 3 \times 10^3$; and curve (c), high dark conductivity, with $h\nu\beta/sI_0 = 2 \times 10^7$.

where $K = h\nu\beta/s$ is constant. Figure 2 shows the dependence of Γ on the total intensity at T = 293 and 313 K. The solid curves represent the best fit based on relation (8) above.

The response time τ of the crystal was determined by fitting an exponential curve $\Gamma = [\Gamma(t \to \infty)][1 - \exp(-t/\tau)]$ through the experimental points for Γ. As suggested by the theory, it is a decreasing function of time over the temperature range that we used, as shown in Fig. 3, ranging from 10.2 sec at 238 K to 0.44 sec at 313 K.

Using some typical values for the quantities in the relations for Γ and τ, we obtained a theoretical fit of the data shown in Figs. 1 and 3. For $E_0 = 0$, the functional form of Γ and τ in the steady state in terms of the temperature-dependent quantities are

$$\Gamma = \Gamma_0\left(\frac{T}{T + B/\epsilon}\right)\left[\frac{1}{1 + A\exp(-E_\beta/k_BT)}\right], \quad (9a)$$

$$\tau = \tau_0\left(\frac{T + C/\mu}{T + B/\epsilon}\right)\left[\frac{1}{1 + A\exp(-E_\beta/k_BT)}\right]. \quad (9b)$$

The values of ϵ were obtained by measuring the low-frequency capacitance of the crystal as a function of temperature and are shown in Fig. 4 as a function of temperature. The theoretical curves were obtained with $N_0 = 10^{25}\ m^{-3}$, $N_A = 3 \times 10^{23}\ m^{-3}$, $\gamma = 1.2 \times 10^{-12}\ m^3/sec$, $s = 1.6 \times 10^{-22}\ m^2$, and $\lambda = 514.5$ nm, which are based on their corresponding values obtained previously for $BaTiO_3$.[7,8] We also assumed that $E_\beta/k_B = 0.224$ eV/K, $A = (h\nu/sI_0)\beta = 2 \times 10^5$, $\mu = 1.5 \times 10^{-4}\ cm^2/V$ sec, $E_g = 0.138$ eV/K, and $\Gamma_0 = 77\ cm^{-1}$ to obtain the fits shown in the graphs. In Fig. 3 the experimental response time increases below 270 K much faster than suggested by theory; we attribute this to our assumption that μ is constant, while it actually would decrease with decreasing temperature.

Futhermore, by extrapolating the values obtained experimentally for $\epsilon(T)$, we were able to predict the temperature T_0 for optimum coupling as shown in Fig. 5. It can be inferred from the figure that for $T < T_0$ the original Kukhtarev's equations hold, while for $T > T_0$ the additional factor in our new model becomes significant. Also, as the dark conductivity decreases, the T_0 increases, so for low dark-conductivity crystals the original Kukhtarev's equations still hold at room temperature.

In conclusion, we have presented a theoretical model that explains the behavior of the photorefractive response time and gain as a function of temperature by incorporating a dark-conductivity term in the Kukhtarev rate equations. The presence of significant dark conductivity in our SBN:60:Cr sample was determined experimentally by measuring Γ as a function of I_0, the total intensity, for a small I_0, and these data were found to fit the theoretically predicted curve closely. The response time of the crystal was measured as a function of temperature and was found to decrease with increasing temperature, as expected from this theory.

This research was supported by grants from the U.S. Air Force Office of Scientific Research and the U.S. Army Research Office. Koichi Sayano also acknowledges partial support from a U.S. Air Force Weapons Laboratory Graduate Fellowship.

References

1. G. A. Rakuljic, K. Sayano, A. Agranat, A. Yariv, and R. R. Neurgaonkar, Appl. Phys. Lett. **53,** 1465 (1988).
2. L. J. Cheng and A. Partovi, Appl. Phys. Lett. **49,** 1456 (1986).
3. N. V. Kukhtarev, V. B. Markov, and S. G. Odulov, Opt. Commun. **23,** 338 (1977).
4. N. V. Kukhtarev, V. B. Markov, S. G. Odulov, M. S. Soskin, and V. L. Vinetski, Ferroelectrics **22,** 949 (1979).
5. N. V. Kukhtarev, Sov. Tech. Phys. Lett. **2,** 438 (1976).
6. M. D. Ewbank, R. R. Neurgaonkar, W. K. Cory, and J. Feinberg, J. Appl. Phys. **62,** 374 (1987).
7. G. C. Valley and M. B. Klein, Opt. Eng. **22,** 704 (1983).
8. J. Feinberg, D. Heiman, A. R. Tanguay, Jr., and R. W. Hellwarth, J. Appl. Phys. **51,** 1297 (1980).