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Photorefractive Properties of Cr-doped Single Crystal Strontium Barium Niobate

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Abstract

Cr-doped strontium barium niobate has shown significant reduction in the time of response compared to previously grown Ce-doped crystals, with room temperature response times as short as 0.2 sec. The experimental photorefractive two-beam coupling gain and response time of 1% and 1.6% Cr-doped SBN:60 and 1% Cr-doped SBN:75 will be presented and compared to results in Ce-doped SBN:60. The photorefractive effect in Cr-doped SBN:60 has also shown a strong temperature dependence, with gain increasing by a factor of two when the crystal was cooled from 40 to -20° C. Significant gain enhancement was also predicted and obtained by applying a DC electric field of up to 10 kV/cm.

1. Introduction

Ce-doped $\text{Sr}_{0.6}\text{Ba}_{0.4}\text{Nb}_2\text{O}_6$ (SBN:60) and $\text{Sr}_{0.75}\text{Ba}_{0.25}\text{Nb}_2\text{O}_6$ (SBN:75) have been shown to be effective media for optical processing and phase conjugation applications because of their large coupling constants, high optical quality, and relatively short response time.¹ In addition, the properties of SBN can be readily changed by varying its composition, large (~ 2 cm cube) crystals have been grown, it is more resistant to temperature changes, applied electric fields, and physical handling, and its open structure enables the addition of a variety of dopants.² The large photorefractive gain coefficients of materials like SBN and BaTiO_3 are desirable for high-efficiency devices and large optical amplification. However, another major goal is to reduce the response time of the materials for signal processing applications where speed is desired. In this paper, we present the results of Cr-doping in SBN:60 and SBN:75, which showed an almost order of magnitude decrease in the response time over Ce-doping, with a corresponding loss in gain by about a factor of 2.

2. Material Properties

SBN is a tungsten bronze ferroelectric material with a general formula of $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$, with both $x = 0.6$ and $x = 0.75$ crystals having been successfully grown. The cation ratio x in large part determines its ferroelectric and electro-optic material properties. Table I shows some of these main properties of SBN:60 and SBN:75.^{2,3}

Table I: Properties of SBN:60 and SBN:75

Material	T_C ° C	E-O Coeff (pm/V)	$n_i^3 r_{ij} / \epsilon_j$ (pm/V)	μ (cm ² /Vsec)	γ_R (cm ³ /sec)
SBN:60	75	235	5.8	0.5	5×10^{-8}
SBN:75	56	1340	5.0	0.5	5×10^{-8}
BaTiO ₃	128	1640	4.9	0.5	5×10^{-8}

Two Cr-doped SBN:60 samples, one with 1% and the other with 1.6% Cr in the flux, and one 1% Cr-doped SBN:75 sample were studied. All were grown using the Czochralski method and were poled into a single domain by

cooling through their cubic to ferroelectric phase transition temperatures with an applied electric field of 8 kV/cm along their *c*-axes.

Fig. 1 shows the absorption spectrum of the three Cr-doped SBN samples as well as that of Ce-doped SBN:60 for comparison. Ce-doped SBN:60 has a broad-band absorption level around 480 nm.¹ Cr-doped SBN has an additional absorption band centered around 650 nm, which may indicate a photoactive transition in the near infra-red.

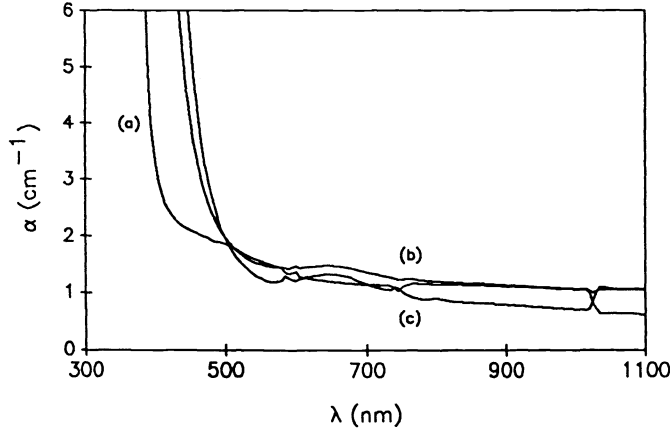


Fig. 1: Absorption spectrum for (a) Ce-doped SBN:60, (b) Cr-doped SBN:60, and (c) Cr-doped SBN:75.

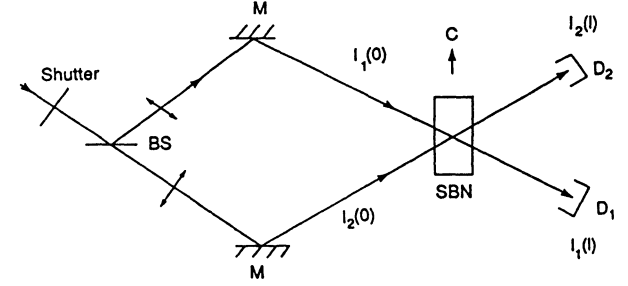


Fig. 2: Configuration of the two-beam coupling experiment used to characterize the SBN:60 crystals. The beam polarization was in the same plane as the *c*-axis. $I_1(0) + I_2(0)$ was approximately 0.25 W/cm².

3. Photorefractive Properties

The photorefractive properties of the SBN:60:Cr crystals were studied using two-beam coupling. Fig. 2 shows the experimental configuration used. Both beams were polarized in the direction of the *c*-axis, i.e. horizontally. The 514.5 nm line of an argon-ion laser with beam diameter of 0.3 cm was used. When the two beams intersect inside the crystal, energy is transferred from one beam to the other in the direction of the *c*-axis, which can be described by

$$\begin{aligned} I_1(z) &= I_1(0) \exp[-(\Gamma + \alpha)z] \\ I_2(z) &= I_2(0) \exp[(\Gamma - \alpha)z] \end{aligned} \quad (1)$$

where α is the absorption coefficient and Γ is the two-beam coupling constant, which is given by⁴⁻⁶

$$\Gamma \propto E_{sc} = iE_N \frac{E_0 + iE_d}{E_0 + i(E_d + E_N)} [1 - \exp(t/\tau)]. \quad (2)$$

The response time of the material is given by

$$\tau = t_0 \frac{E_0 + i(E_d + E_\mu)}{E_0 + i(E_d + E_N)} \quad (3)$$

where

$$t_0 = \frac{h\nu N_A}{sI_0(N_D - N_A)} \quad (4)$$

is the fundamental limit of the speed of the photorefractive effect.⁷ In the Eqns. (2) and (3), E_0 is the externally applied electric field, and the characteristic fields are given by

$$\begin{aligned} E_N &= \frac{eN_A}{\epsilon K} \left(1 - \frac{N_A}{N_D}\right) \approx \frac{eN_A}{\epsilon K} \text{ for } N_A \ll N_D \\ E_d &= \frac{k_B T K}{e} \\ E_\mu &= \frac{\gamma N_A}{\mu K}, \end{aligned} \quad (5)$$

where $K = 2\pi/\lambda_g$ is the wavenumber corresponding to the grating period, γ is the electron recombination rate, μ is the electron mobility, N_A is the trap density, N_D is the donor density, and s is the photoionization cross section.

Figs. 3 and 4 show the experimentally measured two-beam coupling constant and response time, respectively, of the Cr-doped SBN:60 and SBN:75 crystals along with Ce-doped SBN:60 for comparison as a function of the grating wavelength. By differentiating Eqn. (2), the trap density can be obtained as a function of the optimum grating wavelength for maximum Γ . The 1% Cr-doped crystal showed the fastest response time, around 0.2 sec, but had the smallest coupling constant, around 3 cm^{-1} . SBN:75 showed high gain even for a smaller E_{sc} due to its larger electro-optic coefficient.

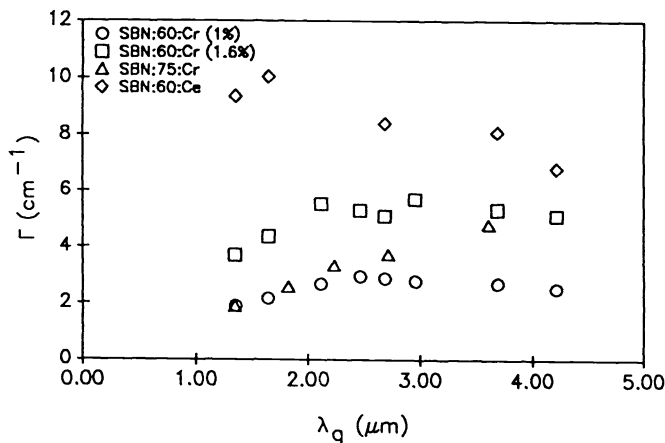


Fig. 3: Steady-state two-beam coupling constant Γ as a function of the grating period λ_g for SBN:60:Cr, doped with 1% Cr; SBN:60:Cr, doped with 1.6% Cr; SBN:75:Cr, doped with approximately 1% Cr; and SBN:60:Ce, doped with approximately 1% Ce.

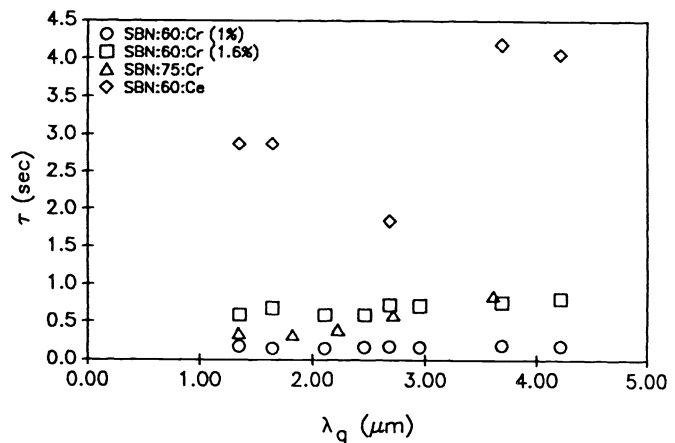


Fig. 4: Response times of the SBN:60:Cr, SBN:75:Cr, and SBN:60:Ce crystals as a function of grating period, for $\lambda = 514.5 \text{ nm}$ and $I_0 = 0.25 \text{ W/cm}^2$.

The preceding experiments were all performed using the 514.5 nm line of the argon-ion laser. These materials were found to be photorefractive at longer wavelengths as well. Figs. 5 and 6 show the effect of using the lower photon energy of the He-Ne laser for two-beam coupling measurements in a 1.6% Cr-doped SBN:60 at $\lambda_g = 2.46 \mu\text{m}$. Because of the lower absorption and fewer ionizable donors at the longer wavelength, the gain and response time results were predictably lower compared to identical measurements using the shorter wavelength sources. The absorption spectrum of Cr-doped SBN shows a broad-band absorption region in the red to near infra-red, and future investigation will determine whether these bands contribute to the photorefractive effect and whether or not these crystals are sensitive at the near infra-red wavelengths used by semiconductor lasers.

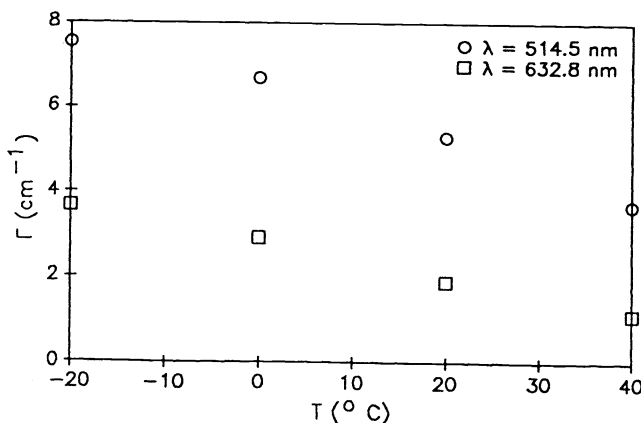


Fig. 5: Two-beam coupling constant of 1.6% Cr-doped SBN:60 at $\lambda = 514.5 \text{ nm}$ and 632.8 nm for $-20^\circ \text{C} \leq T \leq 40^\circ \text{C}$.

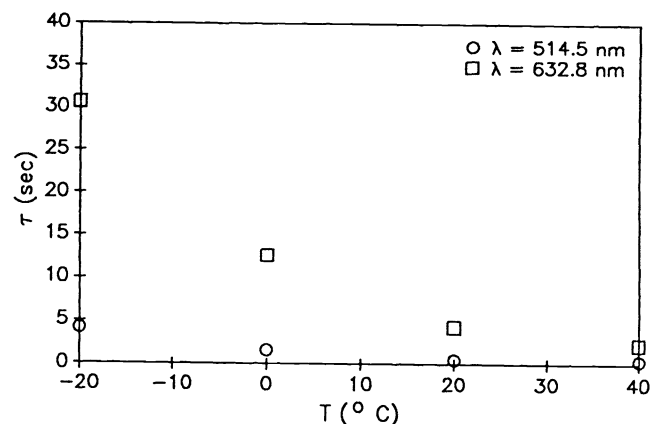


Fig. 6: Response time of 1.6% Cr-doped SBN:60 at $\lambda = 514.5 \text{ nm}$ and 632.8 nm for $-20^\circ \text{C} \leq T \leq 40^\circ \text{C}$.

4. Enhancement of Gain

Various methods are available for increasing the photorefractive gain of SBN. These include optimization of the grating period, lowering the temperature, and increasing the doping. Cooling Cr-doped SBN:60 has been found to increase gain, but results in a considerable increase of the response time, as shown in Figs. 5 and 6. This increase in Γ for lower temperatures can be attributed to decreased leakage of separated charges across the grating due to thermal excitation of trapped carriers.⁸ Increasing the doping is not too fruitful since E_{sc} tends to the smaller of E_d or E_N (see Eqn. (2)). In addition, there exists the practical problem of obtaining high optical quality crystals with large dopant concentrations.

Experimental results have shown that the application of an external DC field on Cr-doped SBN:60 results in a marked improvement in the photorefractive two-beam coupling constant. An external field tends to drive the excited electrons into their traps half a grating period away, resulting in a larger space charge field. In Eqn. (2), for $E_0 = 0$, the limiting field E_{sc} is the smaller of E_d and E_N . For large E_0 , the space charge field approaches E_N , which can be increased by increasing the trap density N_A .

Fig. 7 shows the experimental results of applying a DC field of up to 10 kV/cm to the 1% and 1.6% Cr-doped SBN:60 samples, where increases by more than a factor of two were realized. Since the two-beam coupling intensity gain is exponential, any increase in Γ results in a significant improvement in beam amplification and energy coupling in devices utilizing this effect. It would be possible to use thinner crystals of SBN in experiments and applications, or realize larger signal gain.

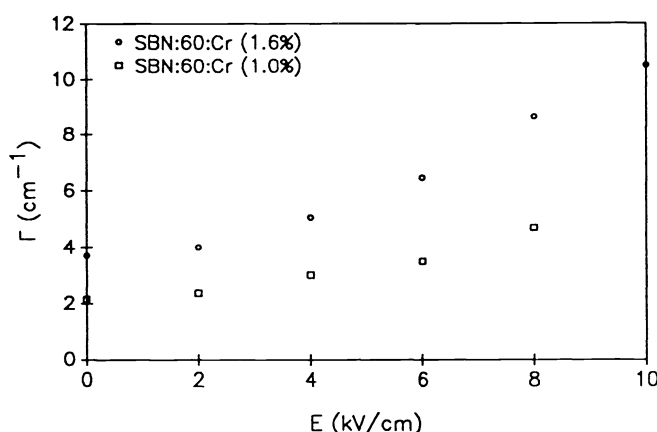


Fig. 7: Measured two-beam coupling constant of 1% and 1.6% Cr-doped SBN:60 with an applied electric field of $0 \leq E_0 \leq 10$ kV/cm.

5. Conclusions

Cr-doped SBN:60 has shown significant advantages in having a faster response over Ce-doped SBN. With the reduced response time, Cr-doped crystals had significantly lower photorefractive gain coefficients that previously grown Ce-doped ones. However, enhancement of the gain was possible through the application of an external DC electric field, resulting in increases in gain Γ by over a factor of two.

6. Acknowledgements

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