Photorefractive gain and response time of Cr-doped strontium barium niobate

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We present experimental results on the photorefractive two-beam coupling constant and response time of two Cr-doped strontium barium niobate crystals with different dopant concentrations. Both showed significantly faster response times over Ce-doped SBN:60, but with corresponding decreases in their coupling constants.

Cerium-doped strontium barium niobate (SBN) has been shown to be an effective photorefractive material for optical processing applications because of its large coupling constant, high optical quality, and relatively short (∼1 s) response time. In addition, an important advantage over BaTiO$_3$ is the fact that its photorefractive properties can be varied by changing its composition, large crystals can be grown, and it is more resistant to temperature changes, applied electric fields, and physical handling. The large coupling constant of SBN makes possible efficient devices, while faster speed of response is desirable for real-time applications. By employing chromium doping rather than cerium, we have found that response time was reduced by almost an order of magnitude, however at the cost of a decrease of the photorefractive coupling constant by about a factor of 2.

SBN belongs to a class of tungsten bronze ferroelectrics with a general formula of Sr$_x$Ba$_{1-x}$Nb$_2$O$_6$ that are usually grown by pulling from a solid solution of alkaline earth niobates. The cation ratio $x$ determines its ferroelectric and electro-optic material properties. It has a nonzero electro-optic tensor arising from its 4mm point symmetry group. Undoped SBN is transparent, and since the SBN unit cell contains 10 NbO$_6$ octahedra with only five alkaline earth cations to fill ten interstitial sites, SBN can accommodate a wide range of dopants.

Two Cr-doped SBN:60 samples, one doped with 1% and the other with 1.6% Cr in the flux, were studied. Both were grown at the Rockwell International Corporation using the Czochralski method. The crystals were polished into a single domain by heating them above their Curie temperatures and cooling them slowly back to room temperature with an applied electric field of 5–8 kV/cm along their $c$ axes. In SBN:60, the dominant electro-optic coefficient $r_{33}$ is 420 pm/V after poling. Our polished samples were cut and 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 dominant impurity.

Figure 1 compares the absorption spectrum of Cr-doped as well as Ce-doped SBN:60. The addition of Ce to undoped SBN:60 has been found to shift the onset of the band-edge absorption from 400 to 430 nm and create a broad-band absorption level around 480 nm. The onset of the Cr-edge absorption in the Cr-doped SBN:60's is near 550 nm, and closer examination reveals that the material has an additional absorption band centered around 650 nm, similar to that observed in a double-doped SBN:60 with Ce and Ca in the 9-fold coordinated sites. Future investigation will determine whether this absorption band contributes significantly to the photorefractive effect.

The photorefractive properties of the SBN:60:Cr crystals were studied using two-beam coupling. Figure 2 shows the experimental configuration. 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. By measuring the incident and transmitted beam intensities $I_i(t), I_s(t), I_f(t)$, and $I_f(t)$ of the two beams, the two-beam coupling constant $\Gamma$ can be obtained by

\[
\Gamma = \frac{1}{l} \ln \left( \frac{I_f(t)}{I_i(t) I_f(t)} \right).
\]

$I_i(t)$ and $I_f(t)$ were measured simultaneously after the shutter was opened to obtain $\Gamma(t)$. The time constant was obtained by determining the time required for the real time $\Gamma(t)$ to rise to 1 - $e^{-1}$ of its steady-state value. This is the beam intensity ratio $I_i(t)/I_f(t)$ was 0.01, resulting in a modulation index $m \approx 0.1$. The total intensity $I_o$ was approximately 0.25 W/cm$^2$.

Using the band transport model, $\Gamma$ as a function of ma-

![Absorption spectrum for SBN:60 doped with approximately 1% Ce and SBN:60 doped with 1% Cr.](image-url)
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. $I_0(0) = E_0(0)$ was approximately 0.25 W/cm$^2$.

The dielectric constant is given by:

$$
\Gamma \propto \frac{E_n E_d}{E_n + E_d} \left[ 1 - \exp \left( -\frac{t}{\tau} \right) \right]
$$

and the time constant $\tau$ by

$$
\tau = \frac{t_0 (E_d + E_n)}{(E_d + E_n)}
$$

where

$$
t_0 = \frac{h v N_D}{s D (N_D - N_A)}
$$

and the characteristic electric fields are given by

$$
E_n = \frac{e N_A}{ek} \left( 1 - \frac{N_A}{N_D} \right) \approx \frac{e N_A}{ek} \quad \text{for} \quad N_A \ll N_D,
$$

$$
E_d = \frac{v}{e} \frac{N_A}{N_D} \gamma N_A / \mu k.
$$

In the above equations, $N_D$ is the donor density, $N_A$ is the trap density, $r$ is the effective electro-optic coefficient, $k = 2 \pi / \lambda_0$ is the grating wave number, $\gamma$ is the electron recombination rate, $\mu$ is the electron mobility, and $s$ is the photoionization cross section. The trap density $N_A$ can be determined from the relation

$$
N_A = \frac{(2 \pi)^2 k n T e / e^2}{\lambda_{0,\text{max}}^2},
$$

where $\lambda_{0,\text{max}}$ is the grating wavelength at which maximum beam coupling occurs. The dielectric constant was obtained by measuring the low-frequency impedance of the crystal.

The steady-state two-beam coupling constants of the two SBN:60:Cr samples are shown together with that of a SBN:60:Ce crystal in Fig. 3 for several values of the grating period ranging from 1.35 to 4.21 $\mu$m. The 1.6% Cr-doped SBN:60 had $\Gamma = 5.7$ cm$^{-1}$ at $\lambda_{0,\text{max}} = 2.4$ $\mu$m as opposed to 3 $\mu$m at 2.5 $\mu$m for the 1% Cr-doped sample. In comparison, Ce-doped SBN:60 had $\Gamma = 10$ cm$^{-1}$ at $\lambda_{0,\text{max}} = 1.6$ $\mu$m. The lines through the points represent the theoretical prediction in the coupling constant as a function of grating period using the experimentally obtained values of $N_A, N_D, \gamma$, and $\Gamma$. For both crystals, the absorption $\alpha = 1.77$ cm$^{-1}$ at 514.5 nm.

Figure 4 shows the effect of dopant concentration on response time. Both Cr-doped SBNs had faster response times than the Ce-doped SBN, with time constants of approximately 0.6 and 0.2 s for the 1.6% Cr and 1% Cr samples, respectively. On the other hand, Ce-doped SBN had a time constant of about 3 s. This result suggests that SBN:60:Cr has a lower recombination rate $\gamma$ and/or larger photoionization cross section $s$ than SBN:60:Ce. In addition, SBN:60:Cr has been observed to have a significantly lower dark conductivity than SBN:60:Ce, which also tends to decrease its coupling constant while speeding up its response time by introducing a net leakage current across the separated charges.

In summary, we have grown optical quality SBN:60:Cr crystals with two different Cr concentrations and shown them to be photorefractive. The coupling constants and response times were 5.7 cm$^{-1}$ and 0.6 s for 1.6% Cr and 3.0 cm$^{-1}$ and 0.2 s for 1% Cr SBN:60 at 0.27 W/cm$^2$ and $T = 20 \degree C$. This is in contrast to $\Gamma = 10$ cm$^{-1}$ and $\tau = 3.0$ s for SBN:60:Ce under the same conditions. Although Cr doping has resulted in decreased photorefractive gain, these materials will still be useful for applications such as signal processing where fast response is desirable but large gain is not, due to its inherent problem of losses due to beam "fanning."

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