# Thermo-optic properties of epitaxial Sr<sub>0.6</sub>Ba<sub>0.4</sub>Nb<sub>2</sub>O<sub>6</sub> waveguides and their application as optical modulator

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**Abstract:** A prism-coupler technique was introduced to determine the refractive indices and thermo-optic coefficients of epitaxial  $Sr_{0.6}Ba_{0.4}Nb_2O_6$  (SBN) waveguides, in a temperature range covering the ferroelectric-paraelectric phase transition. A strong enhancement in the TO coefficient is observed near  $T_c$ . This strong enhancement is related to the critical change of the polarization. The values of  $dn_e/dT$  are significantly larger than

 $dn_o/dT$  due to the larger quadratic electro-optic coefficient in TM polarization. In TM mode, the refractive index of SBN is increased by 1.3% as the temperature is increased to 160°C. Our results suggest that SBN waveguide is a potential candidate for thermo-optic modulators and switches.

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**OCIS codes:** (230.7390) Waveguides, planar; (130.4110) Modulators; (160.6840) Thermo-optical materials.

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#### 1. Introduction

Thermo-optic (TO) coefficient (dn / dT) in materials is defined as a change in refractive index as a function of temperature change. Materials with high TO coefficients have been used in fabrication of optical switches, optical modulators and optical cross-connectors. These devices are used to switch the optical path by controlling the temperature [1–3]. In the past several decades, optical switches and modulators have been developed using nonlinear optic effects such as two-photon absorption, electro-optic (EO) effect, and cross phase modulation to realize ultrafast switching, but the needs of high pumping power, low modulation efficiency and costly short-pulse laser systems may limit their wide applications in optical networks [4– 6]. Some other types of optical switches, such as subgroup of microelectromechanical systems (MEMS), have attracted lots of interests [7,8]. However, the control of these devices is mainly based on mechanical movement of micro-mirrors, and thus it suffers from the problem of mechanical reliability and repeatability. Thermo-optics devices, meanwhile, have advantages such as good reliability and high optical modulation efficiency. These advantages make them become of great interest for scientists and engineers.

Currently, silica and optical polymers are two major materials used to fabricate optical waveguides for TO modulators and switches [9,10]. The main advantages of silica-based TO switches are their easy fiber connection, low optical loss, and good thermal stability. However, they typically require high switching power due to low TO coefficient ( $\sim 10^{-5} \text{ K}^{-1}$ ) and exhibit long response time. The polymer-based TO materials are another type of promising TO materials with high TO coefficient ( $\sim 10^{-4} \text{ K}^{-1}$ ). They are cheap and easy fabrication in any shape, however, their bad thermal stability is the major drawback for practical applications. Comparing with these traditional TO materials, ferroelectric relaxors have been shown to possess an obvious temperature dependence of refractive index and good thermal stability [11]. These properties indicate that they are potential candidates for TO device applications. Tetragonal tungsten-bronze (TTB) Sr<sub>0.6</sub>Ba<sub>0.4</sub>Nb<sub>2</sub>O<sub>6</sub> (SBN), a typical ferroelectric relaxor, has been widely studied with regard to both scientific studies of its EO

and nonlinear properties and possible applications in the field of EO and photo-refractive devices [12,13]. Meanwhile, its optical parameters such as EO coefficient, second nonlinear coefficient and refractive index show a strong temperature dependency. Ramirez *et. al.* and Jacinto *et. al.* reported the coherent light generation and optical distortion from a Nd:SBN crystal at temperatures around its ferroelectric-paraelectric phase transition [14,15]. Goulkov *et. al.* and Zhu *et. al.* studied the temperature dependence of the linear EO coefficient and second harmonic generation in SBN single crystals [16,17]. Up to now, however, limited studies on the temperature dependence of refractive indices as well as TO properties of SBN thin films have been reported. In this paper, we employed a simple, direct and accurate prism-coupler technique to investigate the TO properties of SBN films. A three-dimensional random field Ising model (RFIM) was used to fit the data. Based on our results, we demonstrate that optical modulator and switch can be obtained by using SBN waveguide.

# 2. Experiment

Epitaxial SBN thin films were deposited on single crystal MgO(100) substrates using pulsed laser deposition (PLD). The laser used for the PLD process was a KrF excimer laser (Lambda Physik COMPex205, 248 nm, 20 ns), with a repetition rate of 10 Hz. The on-target laser energy density was about 5 Jcm<sup>-2</sup>. The SBN films were deposited on MgO(100) at a substrate temperature of 740°C and under an oxygen pressure of 100 mTorr. Immediately after the deposition, as-grown films were in situ post-annealed at the growth temperature and 10 Torr oxygen ambient for 10 minutes before they were naturally cooled to room temperature to minimize oxygen deficiency. The film thickness obtained from the SEM cross-section micrograph and prism-coupler technique is 640 nm. X-ray diffraction  $\theta$ -2 $\theta$ ,  $\omega$ , and  $\phi$  scans suggested that SBN films were epitaxially grown on MgO(100) substrates. The proposed epitaxial relationships between the deposited films and the MgO substrates is out-plane (001)SBNII(001)MgO and in-plane [100]SBNII[710]MgO and [100]SBNII[220]MgO. The details of the XRD results were shown in elsewhere [18]. In this paper, a direct and accurate method based on a prism-coupler technique was employed to determine the refractive indices and the TO coefficients of the SBN waveguide films. This powerful technique has been widely used to investigate the optical waveguide properties of thin films waveguide structures. Compared with other methods, this technique is simple because only angle measurements are involved. Furthermore, this method is accurate and non-destructive. The details of the working principle of the prism-coupler have been introduced elsewhere [19]. A laser beam strikes the base of a high refractive index and is reflected onto a photo-detector. The film is brought into contact with the prism base by means of a pneumatically operated coupling head. The angle of incidence,  $\theta$ , of laser beam can be varied by means of a rotary table upon which the prism, film, coupling head, and photo-detector are mounted. At certain discrete incident angles called mode angles, photon can tunnel across the air gap to form an optical propagation waveguide mode, causing a sharp drop in the intensity of light reaching the detector. Following Ulrich and Torge, the refractive index and thickness of waveguide films can be calculated using the waveguide intrinsic equation and the angle at which the modes are excited [20]. Figure 1 shows the temperature controlled prism-coupler setup for the TO measurement. Thin films were put on a temperature controlled heater. Conductive silver paste was used as an adhesive between the heater and the sample substrate to achieve good thermal contact. The sample temperature was monitored by a thermo-couple glued on the film surface with a small amount of silver paste. A laser beam (632.8 nm) with transverse-electric (TE) or transverse-magnetic (TM) polarization was used for the measurements. By heating up the SBN thin films from room temperature to 160°C, spectra of TM and TE modes were recorded and refractive indices were calculated with an interval of 10°C.



Fig. 1. Apparatus for measuring the TO coefficient of thin films using a prism coupler technique.

### 3. Results and discussion

Figure 2 shows the changes of refractive index as well as the birefringence as a function of temperature measured by the prism-coupler technique. SBN is a uniaxial crystal of tetragonal structure with c axis as its optical polar axis. Our SBN films are c-axis oriented and epitaxially grown on MgO(100) substrates. Therefore, the measured  $n_{TE}$  and  $n_{TM}$  represent the ordinary refractive index  $n_{e}$  and the extraordinary refractive index  $n_{e}$  of the SBN films, respectively. In general, the refractive indices of both TM and TE polarizations ( $n_e$  and  $n_a$ ) increase with temperature. For temperatures well below the ferroelectric-paraelectric phase transition, the refractive indices increase slowly with temperature. As the temperature is close to the phase transition temperature, the refractive indices increase rapidly. Finally, as the temperature is well above the phase transition temperature, they increase slowly again. The birefringence decreases from 0.017 to -0.002 gradually since the direction of the dipole moments randomizes gradually with the increase of the temperature, giving a disordered state of smaller net polarization. Through differentiating the temperature dependent curves of refractive index, TO coefficients at different temperatures for TM and TE modes were obtained, as shown in Fig. 3. For temperature near Curie temperature  $(T_c)$ , a strong enhancement in the TO coefficient is observed. SBN is a typical ferroelectric relaxor and most of its related properties are temperature dependent. For most applications it is favorable to work as close to the transition temperature as possible without exceeding it, since most physical effects such as self-frequency doubling, light induced optical-distortion and EO coefficient reach their maxima at the ferroelectric-paraelectric transition [13–15]. As shown in Fig. 3, TO coefficient shows similar behavior as the above mentioned physical properties, that is, it reaches a maximum at  $T_{\rm c}$ . We believe that this enhancement is related to the critical change of the polarization near  $T_{\rm c}$ .



Fig. 2. Experimental and theoretic temperature dependence of refractive index and birefringence of epitaxial SBN films.



Fig. 3. Temperature dependence of TO coefficient: (a)  $dn_{TM} / dT$  and (b)  $dn_{TE} / dT$  of epitaxial SBN films.

In order to explain the experimental results, we correlate the changes in refractive index at different temperature n(T) with the quadratic EO effect which occurs in all crystals and relates a change in the refractive index to the square of a polarization P [21].

$$\Delta(n^{-2})_{mn} = \sum g_{mnop} P_0 P_p \tag{1}$$

where g is the quadratic EO coefficient. Relationship between polarization and refractive index can be described using the following equation:

$$\Delta n_i = n_i - n_i^0 = -(n_i^0)^3 g_{i3} P_3^2 / 2 \tag{2}$$

where  $n_i^0$  is the refractive index at P = 0, i = 1 or 3 where 1 stands the direction perpendicular to the polarization, 3 stands the direction along the polarization. From Eq. (2), the refractive index depends on polarization P as well as quadratic EO coefficient g. From literature, both  $g_{33}$  and  $g_{13}$  depend on temperature. In additional, the value of  $g_{33}$  (~0.10 m<sup>4</sup>/C<sup>2</sup>) of SBN is about 4 times of that of  $g_{13}$  (~0.026 m<sup>4</sup>/C<sup>2</sup>) [22,23]. This explains why the longitude TO coefficient  $dn_e / dT$  is about 3.5 times of that of  $dn_o / dT$  in our measured temperature range as observed in Fig. 3.

Three-dimensional RFIM was employed to calculate the temperature dependence of the polarization [24]. SBN, being a typical relaxor ferroelectrics, is polar with glassy or domain

state properties at low temperatures, and becomes nonpolar paraelectrics at high temperatures. Based on the RFIM, predictions based on critical exponents are possible. According to thermodynamic theories, a phase transition can be described by a constant set of critical exponents, which are well determined and unchangeable in a single system. This critical behavior is an inherent property caused by the interaction of the system's constituent parts. In this letter, the behavior of the order parameter P, the spontaneous polarization, over temperature T is of special interest. Below the phase transition temperature, it is described by:

$$P(T) = P_0 (1 - \frac{T}{T_c})^{\beta}$$
(3)

with a critical exponent  $\beta$ . In the RFIM,  $\beta$  is 0.125 for fully poled SBN [24,25]. Using Eqs. (2) and (3), the calculated temperature dependence of the refractive index can be obtained, as shown in Fig. 2. Below  $T_c$ , the experimental data is well consistent with that of the calculated data, indicating that the temperature assistant prism-coupler technique is an accurate, effective and reliable method to measure the temperature dependence of refractive index and TO coefficient of waveguide materials. But since the RFIM model cannot be used to calculate the polarization above  $T_c$ , the experimental data is deviated from the theoretical data in this region.

Finally, let's briefly discuss the potential application of TO modulators and switches based on the SBN waveguide. The existence of propagation modes is clearly demonstrated in Fig. 4 (a) and (b), where three TM and three TE modes are observed. All of the excited mode-lines are sharp and distinguishable, indicating that a good confinement of light propagation is achieved. Figure 4(c) and (d) display the digital camera captured pictures of excited TM and TE light mode-lines, respectively. From the photographs, it is noticed that both TE and TM mode-lines are sharp and distinguishable, indicating that a good confinement of light propagation is achieved in our films. In order to reduce required switching power and increase switching efficiency in TO modulators and switches, TO materials are required to have a relatively large TO coefficient. Table 1 shows the TO coefficients of some traditional TO materials such as silicon and some polymers which are commonly used in TO waveguide applications [26-29]. Comparing with these materials, epitaxial SBN thin films have larger  $\overline{TO}$  coefficients. Figures 4(a) and (b) display the TE and TM mode spectra at 20°C and 130°C. It is clearly seen that the reflected intensity at a fixed angle of incidence is strongly modulated by changing the temperature of the films. Figure 5 shows the temperature dependency of mode angle as well as the reflected intensity of  $TM_0$ . Obviously, at certain angle of incidence, i.e. mode angle, photons can tunnel across the air gap to form an optical propagation waveguide mode, causing a sharp drop in the reflected intensity of light reaching the detector. This is a full transmission state. This angle of incidence increased with temperature. Thus, if we fixed the angle of incidence at the room temperature mode angle of  $-1.74^{\circ}$  and then increased the temperature, the reflected optical intensity changed accordingly. Below 60°C, the reflected intensity kept negligibly small and increased slowly. This means that most of the photons were excited to form the waveguide mode. The received power increased rapidly by heating up to higher temperature (around 130°C) and then increased slowly again above 130°C. Nearly 80 percent of photons were switched from SBN film waveguide to detector through changing the temperature by a temperature range of 70°C. The switch from full transmission state to total reflection state had thus been achieved through the tuning of the temperature of the SBN film waveguide. Optical loss, which critically affects the performance of film devices, is a key parameter for waveguides. By measuring the scattered light intensity as a function of propagation distance, the propagation loss at  $TE_0$  mode of the SBN films was found to be 1.02 dB/cm.



Fig. 4. The mode (a) TM and (b) TE spectra and their enlarged figure at room temperature and 130 $^{\circ}$ C; Digital camera captured photographs of excited (c) TM mode-lines and (d)TE mode-lines.



Fig. 5. Reflected optical intensity in TM mode and TM<sub>0</sub> mode angle vs temperature.

Table 1	. ТО	coefficients	of	some	commonly	v used	то	materials.
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Materials	Silica	BPDA/PDA	LiTaO <sub>3</sub>	BaTiO <sub>3</sub>	SBN
		(Polymer)			
$dn_{TE} / dT_{(10^{-6} \text{ K}^{-1})}$	~10	-101	10	~0	100
$dn_{TM} / dT_{(10^{-6} \text{ K}^{-1})}$		-60	100	140	300
References	Ref 25	Ref 26	Ref 27	Ref 28	Present work

# 4. Conclusion

We reported the investigation of temperature dependence of refractive index and TO coefficient in epitxial SBN film waveguides through a simple, direct and accurate prismcoupler technique. Both refractive index and TO coefficient showed a strong temperature dependency and a strong enhancement in the TO coefficient in the phase transition range from ferroelectric to paraelectric. The experimental data matched well with the theoretical data using three dimension random field Ising model. The laser intensity can be modulated accurately and effectively in our experiments. Optical switch from full transmission state to total reflection state is achieved through TO effect, suggesting that ferroelectric SBN film waveguides are potential candidate in highly efficient TO optical modulator and switch applications.

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