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Domain effects on the electro-optic properties of thin-film barium titanate

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Abstract: On-chip electro-optic modulation is essential to realize complex on-chip optical signal processing. Recent developments in thin-film ferroelectric oxide for high-speed electro-optical modulators have gained considerable interest in understanding and correlating the material property with the electro-optic response. Particularly, the effect of thin film, domain orientation, and polling on the electro-optic response is not well understood. In this article, we present the effect of ferroelectric domains of thin-film Barium Titanate on the electro-optic response in a waveguide configuration. We also show the impact of drive electrode orientation with respect to the in-plane polarization angle in a multi-domain structure. Our theoretical findings corroborate the experimental observations in the literature, which substantiate the theoretical framework.

1. Introduction

Bandwidth and power consumption limit communication over electrical interconnects and creates a performance bottleneck for data processing. Parasitic resistance and capacitance appear while conventional electronics at high-frequency limits bandwidth expansion. Power consumption increases as we push more signals to the existing data processing systems. With the increase in demand for larger bandwidth for communication, we require a platform that is both fast and power efficient. These requirements can be met using optical interconnects that use light as the carrier. Having optical devices on-chip further adds compactness and a lower footprint. An important component of a photonic integrated circuit is an electro-optic modulator (EOM). An EOM converts electrical data signals to optical signals that can be transported thousands of kilometres with minimal loss. Furthermore, the electro-optic (EO) effect can be used to tune the phase of light to create various optical functionality [1]. Silicon-based nanophotonic devices can enable chip-scale photonic systems with different devices integrated together [2,3]. Silicon, an extensively understood material for nano-devices, has enabled many devices that function in the optical wavelength of interest. Silicon modulators with a bandwidth >50 GHz have been demonstrated. Silicon-based EOMs work under the principle of the plasma-dispersion effect. The bandwidth of such devices has an intrinsic limitation due to the slow carrier mobility [4]. The modulation comes at the cost of absorption loss. A pure phase modulator in silicon is nearly impossible. Thus, a material that could push the current bandwidth limit and achieve pure phase modulation would be an ideal solution for various applications that exploit light modulation.

Pockels effect, also called the linear EO effect, is an ideal alternative. The material's refractive index changes linearly with the applied electric field. This intrinsic material property is exhibited by only a few metal oxides and ferroelectric materials, like Lithium Niobate ($LiNbO_3$ or LNO), Barium Titanate, and Lead Zirconium Titanate. LNO-based EOM is a mature and commercial technology. However, due to the lack of integration of various photonic functionalities and a large footprint, the material platform is limited to a single device. Integrating such EO material into silicon is ideal for realizing a highly integrated photonic circuit. Single crystals of LNO-on-insulator substrates are commercially available for different electro-optic applications.

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However, the highest Pockels effect is offered by a ferroelectric perovskite Barium Titanate ($BaTiO_3$ or BTO) [4].

BTO crystals exist in their tetragonal structure at room temperature, with their Ti^+ ion at the body centre of the lattice unit cell. Each BTO unit cell has a spontaneous polarization along the c-axis of the tetragonal unit cell, described in terms of the shift of the Ti^+ ion along this axis [5,6]. The spontaneous polarization in a single crystal or a single grain in the film is usually not uniformly aligned throughout the material in the same direction. The crystal regions with uniformly oriented spontaneous polarization are called ferroelectric domains. The tetragonal structure of the crystal implies that the material can be grown with two different orientations depending on the process conditions. The so-called a-axis or c-axis orientation depends on whether the optical axis (crystalline z-axis, also called c-axis) is in-plane or out-of-plane in the BTO layer, respectively (Fig. 1).

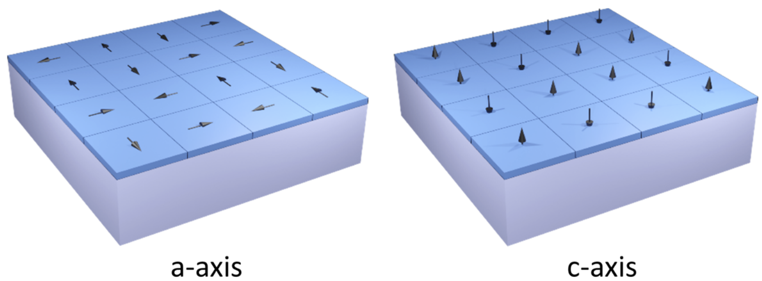

Fig. 1. Schematic view of oriented BTO thin films, a) a-axis film, where the polarization is in-plane, and b) c-axis film, where the polarization is out-of-plane

Thin-film EO modulators of BTO have been demonstrated on silicon [4,7] and other oxide substrates [8,9]. High-quality thin films grown on silicon using the molecular beam epitaxy (MBE) technique on large-scale wafers have been demonstrated for electro-optic applications [4]. However, a few practical difficulties arise when fabricating devices with BTO as the working material. BTO, being a ferroelectric material, shows hysteretic behaviour under the influence of an electric field [10,11]. Unlike a single-crystalline material or CVD deposited films, thin films deposited using physical vapour deposition such as sputtering, e-beam evaporation, or pulsed laser deposition may result in an oriented polycrystalline film [4,11]. Apart from the crystalline or polycrystalline nature, ferroelectric materials have ferroelectric domains. Spontaneous polarization can be in any direction depending on the substrate and growth conditions. Thin films grown on a substrate will get clamped to the substrate, especially near the interface. Each unit cell in the lattice will develop a local electric field during growth. The ferroelectric domains are formed during growth to balance out the forces due to the local electric field and the mechanical restriction by the substrate [5]. When an external electric field is applied to the material, the crystalline structure is slightly distorted and changes the material's refractive index [6,12,13], called the electro-optic effect. However, the electro-optic effect we are interested in here is the Pockels effect due to its high response time (in femtoseconds). Pockels effect is an intrinsic effect in a unit cell in the lattice, arising due to its asymmetry. This effect does not consider the domain nature of the whole film or the refractive index change due to a change in domain orientation. So, when a ferroelectric film is subjected to an electric field, its electro-optic response is a convolution of ferroelectric effects like domain-switching and thin-film effects like domain pinning, along with the Pockels effect [14–16]. Hence, it is essential to understand the effect of ferroelectric domains in BTO's electro-optic effect in a multi-domain thin film. We did not find a comprehensive study that incorporates various electro-optic components.

This work presents modelling and simulations of the EO effect in an a-axis BTO film. A loaded-waveguide structure with amorphous silicon (a-Si) on top of a thin-film BTO is optimized

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to obtain a single propagating TE-mode with maximum light confinement in the BTO layer. We present the effect of domains on the liner EO response, particularly the orientation, number of domains, and distribution of these oriented domains. The results presented here corroborate the experimental observations in the literature [4,17].

2. Theoretical background

Metal oxide thin films grown on a substrate often tend to be textured with a preferential rather than an entirely random orientation of the domain [18,19]. In c-axis films, the domains are mainly oriented parallel or anti-parallel to each other. In an a-axis film, the domains can be parallel or perpendicular to each other. However, since all domains are not in the same direction, their electro-optic response should be weighted when calculating the net Pockels response of the film [15]. Pockels effect, being highly directional, require a tensorial treatment. Each domain will have a different representation depending upon the direction of its spontaneous polarization. Finally, all these contributions from individual domains contribute to a net Pockels effect of a textured film. The change in the matrix elements of the impermeability tensor, $(1/n^2)_i$, due to the Pockels effect can be given as,

$$\Delta(1/n^2)_i = \sum_i r_{ij} E_j \tag{1}$$

where r_{ij} are the matrix elements of the linear electro-optic tensor and E_j are the components of the electric field applied.

For the tetragonal BTO structure, which belongs to the P4mm symmetry class, the linear electro-optic tensor elements r_{13}, r_{33}, and r_{42} are non-zero. This work considers an a-axis film, where the spontaneous polarization is in-plane to the substrate. An a-axis film has the spontaneous polarization of the domains parallel or perpendicular to each other. Hence the effect of domains is more prominent in a-axis oriented films than in the c-axis. Here we consider BTO films grown on a substrate, so we expect pinning down of the crystal lattice to the substrate. Hence, we take the values of the tensor's non-zero elements for mechanically clamped BTO crystals [20], which are 8, 28, and 800 pm/V, respectively. BTO has a negative uniaxial anisotropic performance where the ordinary index, $n_o = n_x = n_y = 2.297$ is higher than the extraordinary index, $n_e = n_z = 2.268$ [4]. This leads to an index ellipsoid for a unit cell with its c-axis (or the crystalline z-axis) in the y-z plane as,

$$\left(\frac{1}{n_x^2} + r_{13}E_z\right)x^2 + \left(\frac{1}{n_y^2} + r_{23}E_z\right)y^2 + \left(\frac{1}{n_z^2} + r_{33}E_z\right)z^2 + (r_{42}E_y)2yz + (r_{51}E_x)2zx = 1 \tag{2}$$

The x-and y-axes of the crystal are interchangeable due to their symmetry in the crystal lattice. Here we consider only TE mode as the waveguide structure is optimized for the fundamental TE_{00} mode. In this simulation work, we consider a BTO layer of a-axis orientation, where the c-axis is free to rotate in the plane of the BTO layer (Fig. 2(a)). The loaded waveguide of a-Si and the electrodes for applying the field are also shown in the schematic in Fig. 2(a). The angle between the optic axis (arrow in the figures) and the direction of the electric field is referred to as ϕ (Fig. 2(b)).To analyze this model, we define a new coordinate system with the c-axis in $y' - z'$ plane and making an angle ϕ to the direction of the electric field (Fig. 2(a)). The applied electric field is mainly in the lateral direction, so the electric field perpendicular to the substrate, E_x, is zero. The index ellipsoid in the new coordinate system can be obtained by using the following transformation,

$$\begin{aligned} y &= y' cos(\phi) - z' sin(\phi) \\ z &= y' sin(\phi) + z' cos(\phi) \end{aligned} \tag{3}$$

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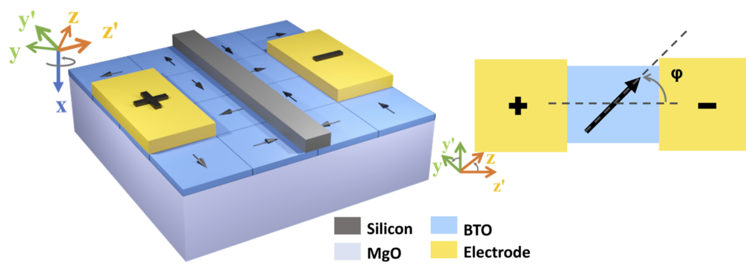

Fig. 2. a) Schematic view of the simulated model of a-axis BTO films with loaded waveguide and electrical contacts. b) Schematic view of the angle ϕ (top view).

Accordingly, the TE mode in the waveguide sees the refractive index of the material along its z' coordinate. The electric field is also applied along the z' axis. So, by operating in Eq. (1), the refractive index and effective EO coefficient can be derived for TE polarization,

$$\begin{aligned} n_{z'}(\phi) &= \frac{n_e n_0}{\sqrt{(n_e^2 sin^2(\phi) + n_o^2 cos^2(\phi))}} \\ r_{z'}(\phi) &= r_{33}cos^3(\phi) + (r_{13} + 2r_{51})sin^2(\phi)cos(\phi) \end{aligned} \tag{4}$$

A detailed derivation is presented in Supplement 1.

3. Simulation results

The cross-section of the loaded-waveguide structure for BTO grown on MgO substrate is represented in Fig. 4(a), with a lateral electric field. The thickness of the BTO layer and the top a-Si is 100 nm each. The waveguide width was optimized for single TE-mode propagation, with the maximum field in the BTO layer. A waveguide width of 450 nm is found to be optimal. The waveguide dispersion curve and the curve for the confinement factor of the fundamental TE mode as a function of waveguide width is provided in Fig. 3. The electrodes are kept 2 μm far from the waveguide on either side to avoid the electrode metal absorbing the optical field. Hence, the electrode gap is 4.45 μm. The various parameters used in the simulation models are tabulated in Table 1. We use COMSOL Multiphysics solver, considering material anisotropy in the cross-section presented in Fig. 4(a).

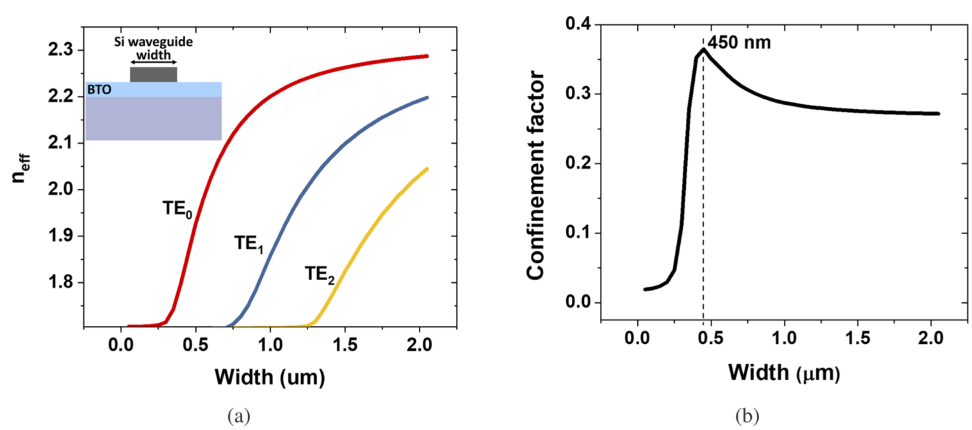

Fig. 3. a) Waveguide dispersion. b) Confinement factor of the fundamental TE mode as a function of waveguide width.

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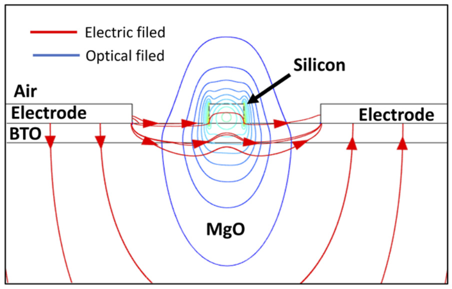
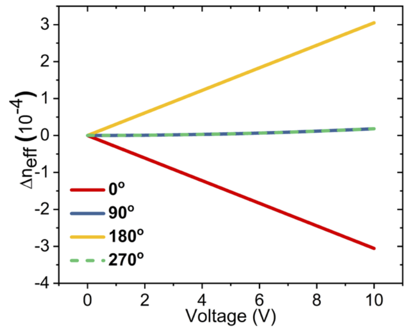


(a)



(b)

Fig. 4. a) Results of EO simulations of the loaded-waveguide structure showing the optical and electrical fields. b) Change in n_{eff} as a function of voltage for a single domain with different orientations.

Table 1. List of simulation parameters and values

Parameters	Values	Parameters	Values
BTO thickness	100 nm	Wavelength	1550 nm
n_o (BTO)	2.297	Waveguide thickness	100 nm
n_e (BTO)	2.268	Waveguide width	450 nm
$r_{13} = r_{23}$	8 pm/V	n (Si)	3.45
r_{33}	28 pm/V	Electrode gap from waveguide	2 μm
$r_{42} = r_{51}$	800 pm/V		

The effective index n_{eff} of the fundamental TE (transverse electric) mode is monitored as a function of the applied voltage to the electrodes. Though the following results show the effects of applied voltage, one should keep in mind that the film's properties change as per the electric field it sees. For the configuration used in this simulation, with a voltage of 10 V, the film experiences 22 kV/cm of the electric field. So, we need to scale the voltage based on the gap between the electrodes varies. The change in the effective index, Δn_{eff}, due to applied voltage depends on the orientation of the domain (Fig. 4(b)). The index change depends on the direction of the c-axis of the domain with respect to the applied field direction, denoted by the angle ϕ (Fig. 2). Between an orientation angle of 0^o and 90^o, the effective Pockels coefficient is shown in Fig. 5, as per equation (4). This effective coefficient is maximum at an angle of 54^o. It is interesting to note that this angle is not 45^o [14]. We would expect 45^o since that is when we have an electric field in both the c- and a-axes.

When the BTO layer is considered to have two domains, the relative orientation of the two domains determines the overall change in the refractive index, as shown in Fig. 6. The relative domain effect is studied with one domain fixed while the other is varied with respect to the electric field. We consider only those domains with an orientation of 0^o, 90^o, 180^o, and 270^o since an a-axis BTO film will only have these domain alignments. The waveguide mode index, n_{eff}, can increase, decrease, or remain unchanged with the applied voltage based on the domain alignment. Figure 6 shows the EO response and corresponding schematic representation of the orientation of the two domains considered.

When either domain is 0^o, we observe a negative shift, except when the other domain is 180^o. Furthermore, when either domain is 180^o, we observe a positive shift (Fig. 6; the yellow trace in Fig. 6(b), 6(c), and 6(d), except when the other domain is 0^o (Fig. 6(a)). When both 0^o and

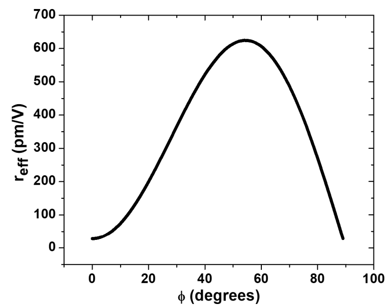


Fig. 5. Change in effective Pockels effect $r_{z'}$ as a function of angle between in-plane c-axis and applied electric field.

180^o domains are present, the linear electro-optic effect from the two domains negates each other resulting in a null change in the refractive index; yellow trace in Fig. 6(a), red trace in Fig. 6(c). Similarly, if both domains have identical orientations, they aid each other resulting in a significant shift. When the other domain is 90^o or 270^o, we observe a minor change; green and blue trace in 6(b) and 6(d). The 90^o or 270^o domains do not contribute along these directions.

However, a BTO film can have multiple domains with different orientations. Figure 7(b) shows a model where the BTO layer is constructed with multiple domains of size 100 nm each between the electrodes. 100 nm is chosen to show the multi-domain effect of the film. The effect of domain size is discussed later on in the Simulation Results section. These domains are randomly distributed, so we have zero net polarization using a random number denotation. For an arbitrary random distribution (RD1), the resultant n_{eff} increases with voltage (Fig. 7(a)). Figure 7(b) shows the schematic view of the model with multiple domain orientations. Figure 7(b) is the top view of the schematic showing the domains' relative orientations. However, with domains poled, the n_{eff} shift exhibits the characteristic reduction in n_{eff} with voltage (RD1 poled in Fig. 7(a)). Poling all the domains along the direction of the electric field will create the effect of a single-domain film of BTO. Here we refer to poling of only the 180^o domains since 90^o or 270^o domains require a higher electrical field to pole in a thin film [5]. Figure 7(c) shows the domain poling. When the 180^o domains are poled along the electric field, the n_{eff} decreases with voltage. This decrease is not as significant as in the case of a poled single domain (Fig. 4(b)). We also find that despite net-zero polarization, a different combination of the oriented domains (RD2 unpoled in Fig. 7(a)) decreases n_{eff} with voltage, and the change improves with poling (RD2 poled in Fig. 7(a)). These results indicate that the adjacent domain orientations determine the net electro-optic response of the devices. Poling of just the parallel domains is not enough to give the effect of a single-domain BTO film. We have to pole the entire film in one direction, along the direction of the applied electric field.

Since the effective Pockels coefficient is maximum at 54^o (Fig. 5), we expect the change in the refractive index to be larger in this orientation for the same applied field. The simulation result in Fig. 8 confirms this hypothesis. Here we have the same model where the BTO layer is constructed with multiple domains of width 100 nm each between the electrodes separated by a gap of 4.45 μm. These domains are all randomly distributed in four perpendicular orientations, using a random number denotation as used earlier. All the domains are tilted at an angle of 54^o with respect to the applied electric field. We consider two random orientation sets; RD1 and RD2. For RD1 unpoled distribution, the resultant n_{eff} decreases. However, when the domains are poled, the n_{eff} shift increases with voltage (Fig. 7(a)). Figure 7(b) shows the schematic of the domains named in Fig. 8(a). When domains are partially poled, as shown in Fig. 7(b) configuration-1, we observe a positive change in n_{eff} and further poling as shown in Fig. 7(a) configuration-2, the observed shift is larger for the same voltage. We confirm the observation

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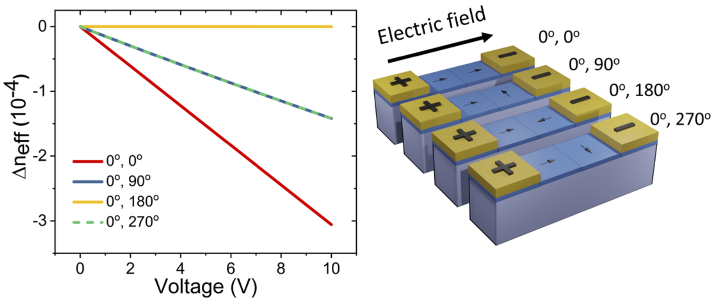
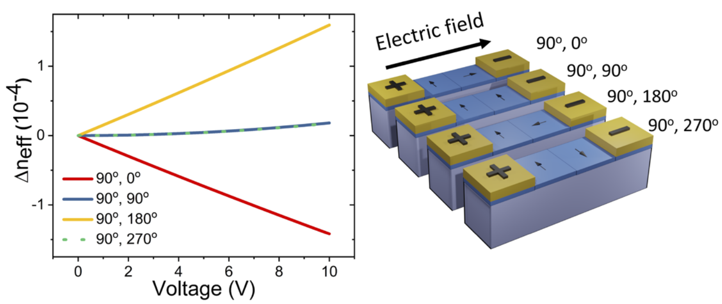
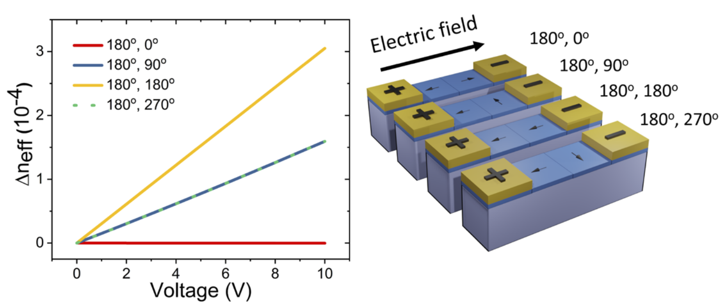


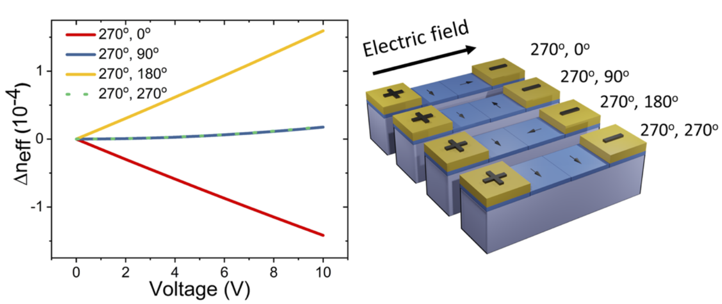
(a) 0^o-ϕ



(b) 90^o-ϕ



(c) 180^o-ϕ



(d) 270^o-ϕ

Fig. 6. Change in n_{eff} as a function of voltage for two domains across the electrodes, with the schematic image of the relative orientations.

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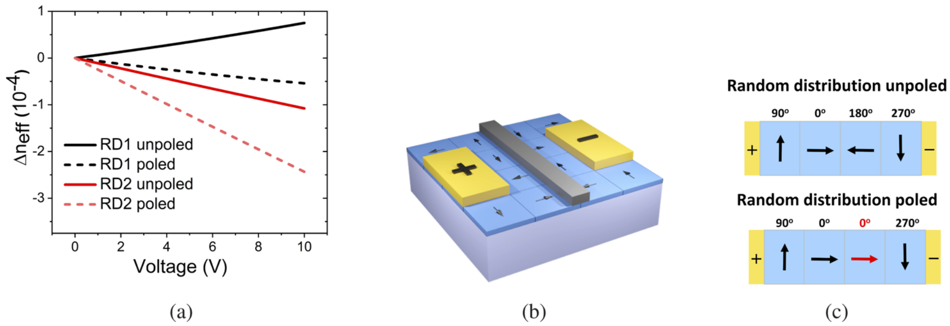

Fig. 7. Change in n_{eff} as a function of applied voltage across electrodes with multiple domains with random orientations (RD). b) Schematic of the domains. c) Top view of the relative orientations.

with a second random set RD2 and observe a positive n_{eff} slope despite starting with an unpoled combination that results in a negative slope. The change in refractive index upon application of an electric field decreased when the domains were oriented parallel or perpendicular to the electric field but increased when they were at an angle. This change in the trend of Δn_{eff} can be attributed to a non-zero electric field in both c- and a-axes. The simulation presented here is supported by the experimental observation presented by S. Abel et al. [17]. The work reports the response of a micro-ring resonator (MRR) based EO modulator as a function of an external electric field applied for two configurations. The first configuration is when the electric field is along the 90^o and 180^o domains, and the second is when the field is 45^o to the domains. S. Abel et al. reports a decrease in n_{eff} of the optical mode in the MRR, indicated by a blue shift of its resonance frequency for the former case and an increase in n_{eff} for the latter case. One can also observe a more significant shift in the second case than in the first [17]. This observation agrees with our simulation studies that showed higher r_{eff} when the domains are at an angle to the electric field applied (Fig. 5). These results in experimental devices ascertain the importance of domain orientations in an epitaxially grown a-axis film of BTO on a substrate.

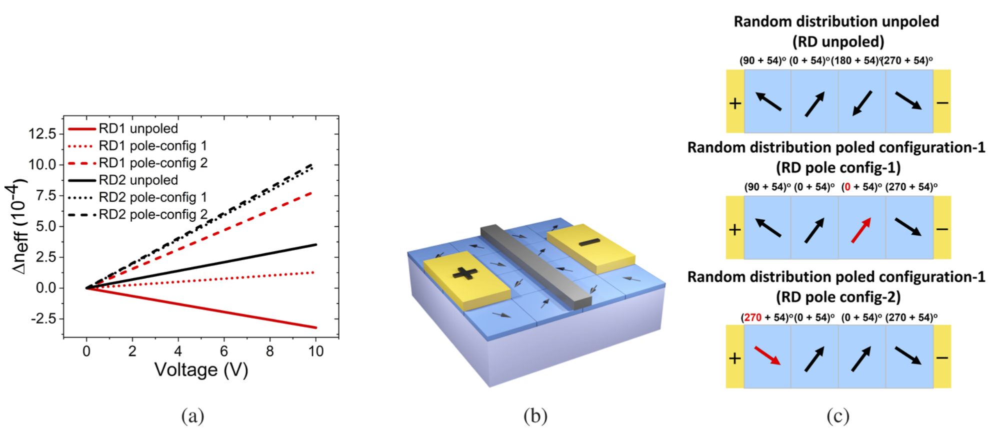

Fig. 8. a) Change in n_{eff} as a function of voltage for the models with multiple domains of BTO with random distributions (RD) and their poled versions. b) Schematic of the domains and (c) their orientation, in top view.

Additionally, to study the effect of domain size on the electro-optic effect, we made a model where the entire BTO film has its c-axis perpendicular to the electric field, except for one domain

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beneath the Si waveguide. The rest of the film is considered perpendicularly oriented without any contribution to the n_{eff}. This domain is oriented parallel to the electric field, and its size varies. Figure 9 shows the n_{eff} changes with applied voltage for different domain sizes. As the oriented domain grows, we see that the reduction in n_{eff} increases as the size increases. The trend in Δn_{eff} moves towards a single domain film oriented along the electric field. The $1/e^2$ width of the optical field intensity of the fundamental TE mode of the waveguide structure is approximately 500 nm from the centre of the waveguide. When the size of the domain is smaller than the optical field, the change in n_{eff} is small than a single crystal BTO film. However, once the domain size is bigger than the optical field (1000 nm), the response is identical to a single crystal-single domain configuration. The domains that interact with the electric and optical field alone determine the overall character of the device.

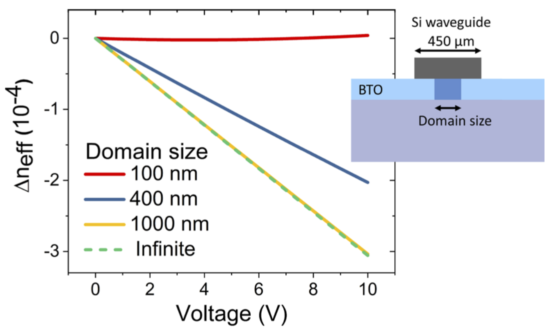

Fig. 9. Change in n_{eff} as a function of voltage for different domain sizes of a poled domain.

All these results indicate that ferroelectric domains play an essential role in the electro-optic characteristics of individual devices on a-axis BTO thin films. Controlling the size of the domains, and their relative orientations, say during the growth of the film or by poling the film, we can have different optical properties of the film for the same voltage. In other words, we can make use of this domain structure of the film to have different states of the materials that can be used in applications like photonic neuromorphic and quantum computing networks. Recently, work on using BTO as a non-volatile optical phase shifter exploits the multi-domain structure of textured BTO thin films [21]. They report achieving analog and non-volatile optical phase tuning by manipulating ferroelectric domains in BTO with controlled electric fields. An extensive analysis of the domain structure and orientation in electro-optically active ferroelectrics is crucial to fully utilize them for innovative applications.

4. Conclusion

We have demonstrated that the relative orientations of adjacent ferroelectric domains, along with the net polarization, determine the electro-optic response of a device made on a-axis BTO thin films. The response of a fabricated device will intrinsically depend on the distribution of domains in the device area, leading to device-to-device variability of electro-optic response. Efficient polling could reduce the variability; however, measuring the initial multi-domain distribution is essential for efficient polling. Applying the electric field at an off-axis angle to the domains will help probe the material's largest Pockels coefficient. Domains interacting with electric and optic fields alone respond in an electro-optic measurement. Hence, changing the domain size can also help to get the effect of a single domain film, which gives the best response. However, having multiple domains can also be utilized for specific applications. We can manipulate these domains, size, and relative orientations to achieve multi-state material ideal for non-volatile optical device components.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.

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