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Optical properties of $Ba_{0.5}Sr_{0.5}TiO_3$ thin films grown on MgO substrates by pulsed laser deposition

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Abstract

Barium strontium titanate ($Ba_{0.5}Sr_{0.5}TiO_3$ or BST (50/50)) thin films were grown on MgO(1 0 0) substrates using pulsed laser deposition (PLD). The surface morphology of the thin films was observed using a scanning probe microscope and the grain size was found to be around 100–150 nm. The surface roughness is below 5 nm for a 250 nm thick film. X-ray diffraction (XRD) results revealed that the BST thin films were epitaxially grown on the MgO(1 0 0) substrates. Optical transmittance and electro-optic (E-O) properties of the BST thin films were measured using a phase modulation detection method. The BST/MgO configuration was highly transparent in the visible region. The BST film exhibited a predominantly quadratic E-O behavior and the quadratic E-O coefficient was found to be 0.42×10^{-17} m^2/V^2.

Keywords: BST thin films; Electro-optic; Ellipsometer

1. Introduction

Thin films of ferroelectric materials have attracted considerable attention due to their wide range of applications such as high dielectric constant capacitors, dynamic random access memories, surface acoustic wave (SAW) devices and uncooled infrared detectors [1]. Ferroelectric thin films with large electro-optic (E-O) effects are potential materials for integrated optical devices [2,3]. The use of thin films in E-O devices can lead to a reduction in device size and an increase in interaction efficiency [1]. A large number of ferroelectric oxide thin films has already been investigated as promising candidates for E-O applications, including lanthanum modified lead zirconate titanate (PLZT), lead zirconate titanate (PZT), lead magnesium niobate-lead titanate (PMN-PT), lithium niobate ($LiNbO_3$) and strontium barium niobate (SBN) [4–7].

Barium strontium titanate [$Ba_{1-x}Sr_xTiO_3$ or BST ($1-x/x$)] is an excellent microwave material because of its unique combination of large dielectric permittivity and high dielectric tunability in the microwave regime at room temperature. Great efforts had been focused on this material in the past few years since the discovery of the promising E-O characteristics in BST thin films [8,9]. At room temperature, BST is a tetragonal ferroelectrics when the Sr content x is in the range of 0–40% and has a cubic paraelectric structure when x is in the range of 40–100%. In this paper, we report the E-O properties of (1 0 0)-oriented BST (50/50) thin films deposited on MgO(1 0 0) substrates. Other optical properties such as optical transmission and band gap were also studied.

2. Experimental

BST (50/50) thin films were deposited on MgO(1 0 0) substrates using the pulsed laser deposition (PLD) technique. Two hundred and fifty nanometers thick films were grown on MgO substrates by irradiating the target for 15 min with the laser beam of 248 nm wavelength and 25 ns pulse duration from a KrF excimer laser (Lambda Physik COMPex 205). The laser beam energy, repetition rate, oxygen pressure and substrate temperature were 300 mJ, 10 Hz, 200 mTorr and 650 °C, respectively. The as-prepared BST thin films were transparent. The crystal structures of the thin films were examined using an X-ray diffractometer (Bruker D8 Discover) equipped with Cu Kα radiation. The surface morphology of the thin films was observed using a scanning

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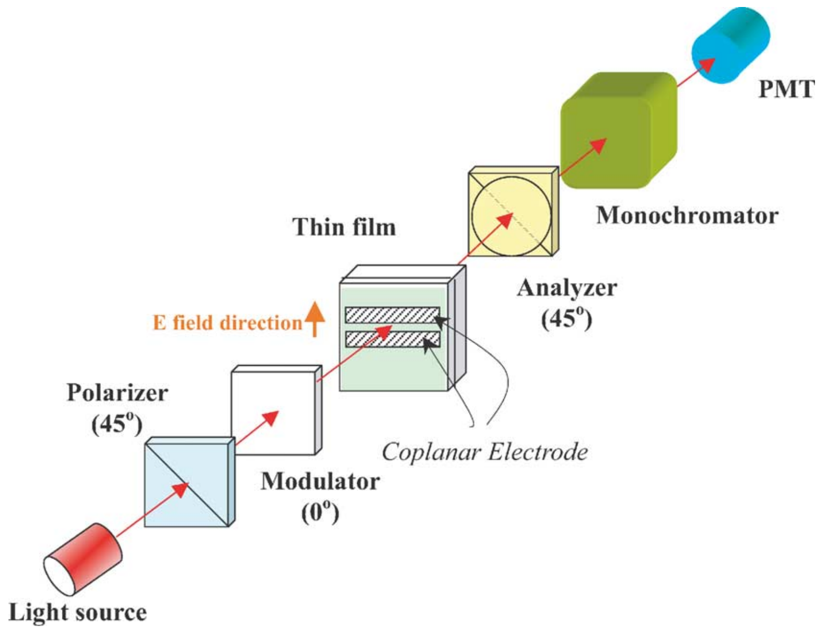

Fig. 1. Experimental setup for measuring birefringence shifts.

probe microscope (SPM) (Digital Instruments NanoScope IV) working in tapping mode. The optical transmission of the BST thin films was measured using a transmission type ellipsometer (Jobin Yvon UVISEL) in the wavelength range of 280–850 nm.

For E-O measurements, coplanar gold electrodes with a gap of 40 μm were deposited on one surface of the film by rf magnetron sputtering. The E-O properties of the thin films were measured using a phase modulated ellipsometer (Jobin Yvon UVISEL) in transmission mode at a fixed wavelength of $\lambda = 633$ nm. The schematic diagram of the experimental setup is shown in Fig. 1.

The incident light beam was linearly polarized by a polarizer (P) and then modulated by a photoelastic modulator (M). The photoelastic modulator consisted of a rectangular fused silica block sandwiched between two piezoelectric quartz crystals oscillating at a resonant frequency of 50 kHz. After modulation, the light beam impinged normally on the surface of the thin film sample at the gap between the two electrodes and an electric field was applied in the vertical direction. The transmitted beam, then passed through an analyzer (A) and a monochromator set at 633 nm and was detected by a photomultiplier (PMT). The modulator generated a periodic phase shift $\delta(t)$ between the orthogonal amplitude components of the transmitted beam, and the detected light intensity can be described by:

$$I(t) = I_o + I_s \sin[\delta(t)] + I_c \cos[\delta(t)] \quad (1)$$

The explicit expressions for I_o, I_s and I_c can be found elsewhere [10]. For a suitable choice of optical component configuration as shown in Fig. 1, i.e. setting the polarization axes of P and A at 45° to the vertical, the ellipsometer angles Δ and Ψ are related to I_o, I_s, and I_c by:

$$I_o = K \quad (2)$$

$$I_s = K \sin(2\Psi) \sin \Delta \quad (3)$$

$$I_c = K \sin(2\Psi) \cos \Delta \quad (4)$$

where K is a constant. Therefore, we could calculate Δ and Ψ by measuring I_s and I_c. Consequently, the electric field induced birefringence can be deduced using the phase change $\delta\Delta$.

3. Results and discussion

Fig. 2(a) shows the X-ray diffraction (XRD) patterns for a BST (50/50) thin film deposited on the MgO(1 0 0) substrate. Only the (1 0 0) peaks appear in the XRD pattern, implying that the BST (50/50) thin film has a single phase with an out-of-plane lattice constant of 3.9481 Å. The epitaxial relationship was characterized by means of ϕscans which are shown in Fig. 2(b). The BST films have a four-fold symmetry, which is similar to the MgO(1 0 0) substrate. For optical applications, uniformly aligned crystal grains in an epitaxially grown film are strongly desirable.

The surface morphology and roughness of the epitaxial BST (50/50) thin film are shown in the SPM micrograph in Fig. 3. The root-mean-square (rms) roughness is below 5 nm over a 1 μm × 1 μm area for the 250 nm thick films. The grain size is estimated to range from 100 to 150 nm. The SPM image reveals that the BST thin film has a smooth and dense surface.

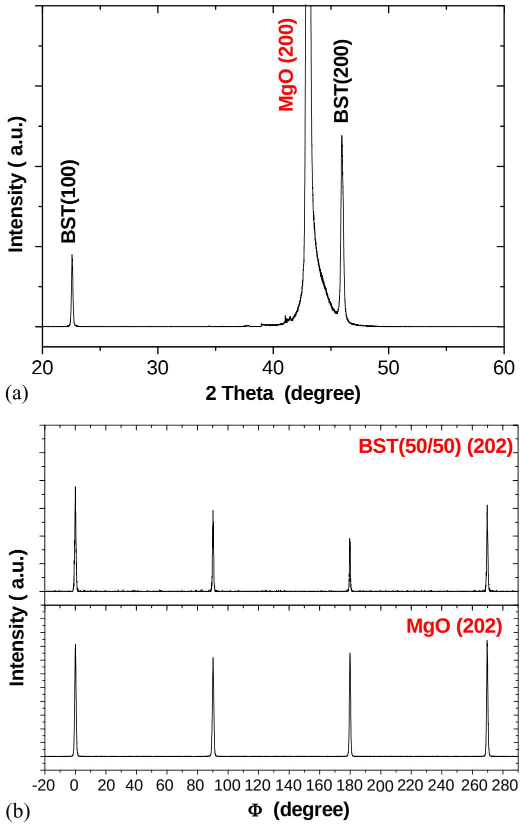

Fig. 2. XRD pattern of (1 0 0)-oriented BST ($x = 0.5$) thin films. (a) $\theta/2\theta$ scan; (b) ϕ scan of the (2 0 2) reflections of the BST (50/50) thin films.

The optical transmission of the BST (50/50) thin film (250 nm thick) in the wavelength range of 280–850 nm is shown in Fig. 4. The films are highly transparent in the visible region with a transmittance between 60 and 90%. The transparency of the film drops sharply in the UV region and the absorption edge is located at 328 nm. The optical band gap energy E_{gap} of a thin film can be deduced from the spectral dependence of the absorption constant $\alpha(\upsilon)$ by applying the Tauc relation [11]:

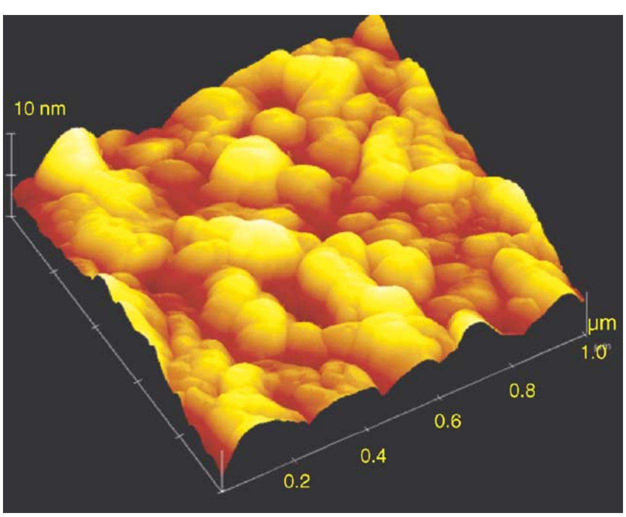

Fig. 3. SPM image of (1 0 0)-oriented BST (50/50) thin film grown on MgO(1 0 0) substrate (250 nm thick).

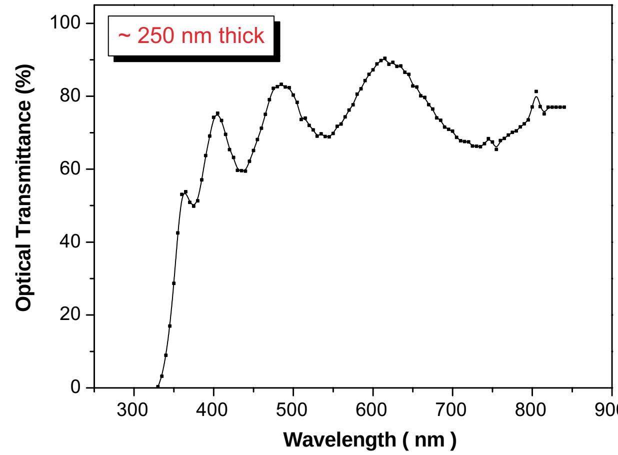

Fig. 4. Optical transmission spectra of (1 0 0)-oriented a BST (50/50) thin films grown on MgO(1 0 0) substrate.

$$(\alpha h\upsilon)^2 = \text{constant}(h\upsilon - E_{gap}) \quad (5)$$

where υ is the frequency and h is the Planck's constant.

The absorption constant $\alpha(\upsilon)$ is determined from the transmittance spectrum using the relation [11]:

$$\alpha(\upsilon) = \frac{\ln(1/T(\upsilon))}{d} \quad (6)$$

where $T(\upsilon)$ is the transmittance at frequency υ and d the film thickness. The optical band gap energy is then obtained from Eq. (5) by extrapolating the linear portion of the plot of $(\alpha h\upsilon)^2$ versus $h\upsilon$ to $(\alpha h\upsilon)^2 = 0$. The optical band gap energy is found to be 3.65 eV for the BST (50/50) thin film.

Using the phase modulation detection method, the field induced birefringence of the BST (50/50) thin film was measured as a function of external electric field at room temperature and is shown in Fig. 5. In Fig. 5, a quadratic E-O behavior is clearly observed. Fig. 6 shows the plots of birefringence shifts versus E^2 in the BST (50/50) thin film. The

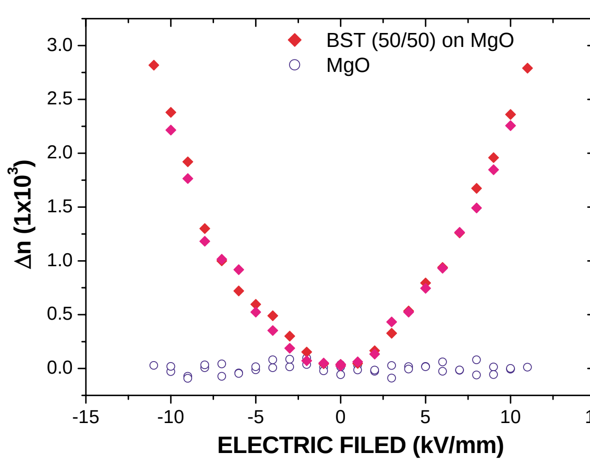

Fig. 5. Electro-optic responses of a (1 0 0)-oriented BST (50/50) thin film grown on a MgO(1 0 0) substrate.

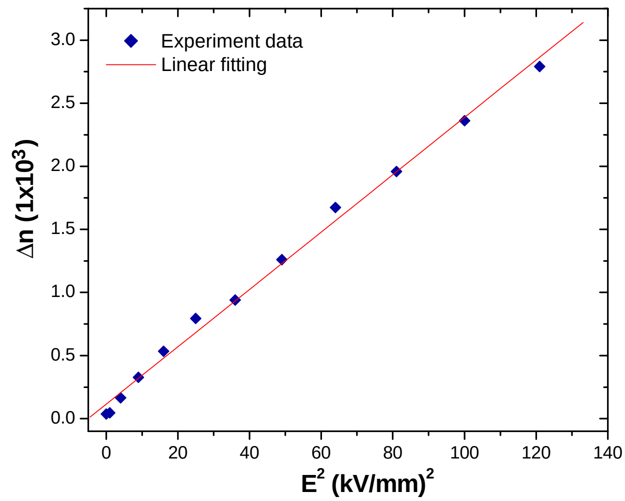

Fig. 6. Electro-optic response vs. E^2 of a (1 0 0)-oriented BST (50/50) thin film grown on a MgO(1 0 0) substrate.

experimental data exhibits good linearity with the E^2. The quadratic electro-optic coefficient R is defined by the equation [12]:

$$\Delta n = -\frac{1}{2}n^3RE^2 \tag{7}$$

where Δn is the birefringence, n the refractive index (n = 2.2 for BST) and E the external electric field. Using Eq. (3) and Fig. 5, the quadratic E-O coefficient R was calculated to be $0.42 \times 10^{-17}\,m^2/V^2$ for BST (50/50) thin film. This value is comparable to the E-O coefficients of ferroelectric relaxor PLZT thin films [4]. A similar electrode pattern has been deposited on a bare MgO substrate and upon application of external dc field, $\Delta n = 0$ was observed (Fig. 5).

4. Conclusions

BST (50/50) thin film has been epitaxially grown on a MgO(1 0 0) substrate using pulsed laser deposition. SPM images reveal that the BST (50/50) thin film has a smooth and dense surface and has a grain size ranging from 100 to 150 nm. The BST (50/50) film is highly transparent in the visible light region. The optical band gap of the BST (50/50) thin film has been calculated by applying the Tauc relation. The E-O property of the BST thin film has been characterized using a phase modulation detection method. A quadratic dominated E-O behavior has been observed. The quadratic E-O coefficient of BST (50/50) thin film has been found to be $0.42 \times 10^{-17}\,m^2/V^2$. This value is comparable to those of PLZT thin films, indicating that BST is a promising candidate for E-O device applications.

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