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Pulsed laser deposition optical waveguiding Bi_3TiNbO_9 thin films on fused silica

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Abstract

The layered-perovskite ferroelectric Bi_3TiNbO_9 (BTN) optical waveguiding thin films have been prepared on fused silica substrates by pulsed laser deposition (PLD). X-ray θ–2θ scans revealed that the films are single-phase perovskite and highly (00l) textured. The wavelength dependence of the transmittance of the films was determined. We obtained an average transmittance of 75% in the wavelength range of 400–1100 nm and the band gap E_g=3.55 eV. The optical waveguiding properties of the films were characterized by using prism-coupling method. The distinct m lines of the guided transverse magnetic (TM) and transverse electric (TE) modes of the BTN films waveguide have been observed. The cross sectional morphology of the film was studied by scanning electron microscopy (SEM).

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1. Introduction

In recent years, increasing attention has been focused on the preparation dielectric optical waveguide. The planar waveguide structures have been proposed and demonstrated for many applications, including nonlinear electro-optical devices, electro-optic waveguide modulators, passive integrated optical circuits and two-dimensional optical elements [1–4]. Ferroelectric materials have attracted a lot of attention for electrical and optical applications due to their excellent electric-optical properties [5,6]. Liking $SrBi_2Ta_2O_9$, Bi_3TiNbO_9 (BTN) with a same bismuth layer perovskite structure show excellent fatigue free properties [7,8]. However, the optical properties of BTN films have rarely been reported up to now. This ferroelectric is of perovskite type and therefore should exhibit large electro-optic coefficients. An understanding of optical properties of this kind of materials is important for evaluating its potential as electro-optical active material. In this paper, BTN waveguiding thin films were fabricated on fused silica by a pulsed laser deposition (PLD) technique.

This kind of materials with Aurivillius phase can be written as the formula: $(Bi_2O_2)^{2+}(A_{m-1}B_mO_{3m+1})^{2-}$, where A represents Bi, Ba, Pb, Sr, Ca, K, Na, etc., and rare earth elements; B represents Ti, Ta, Nb, W, Mo, Fe, etc., and m=2,3,4,... represents the number of BO_6 perovskite octahedra between two Bi_2O_2 layers [9].

2. Experiments

BTN targets used in PLD were prepared by a conventional solid-state reaction technique with starting materials Bi_2O_3, TiO_2, and Nb_2O_5. Excess 20 mol% Bi_2O_3 was added to compensate the Bi evaporation. The powder was mixed by ball milling for 12 h and then preheated at 700 °C for 3 h. In order to obtain dense BTN ceramics, a little 5% polyvinyl

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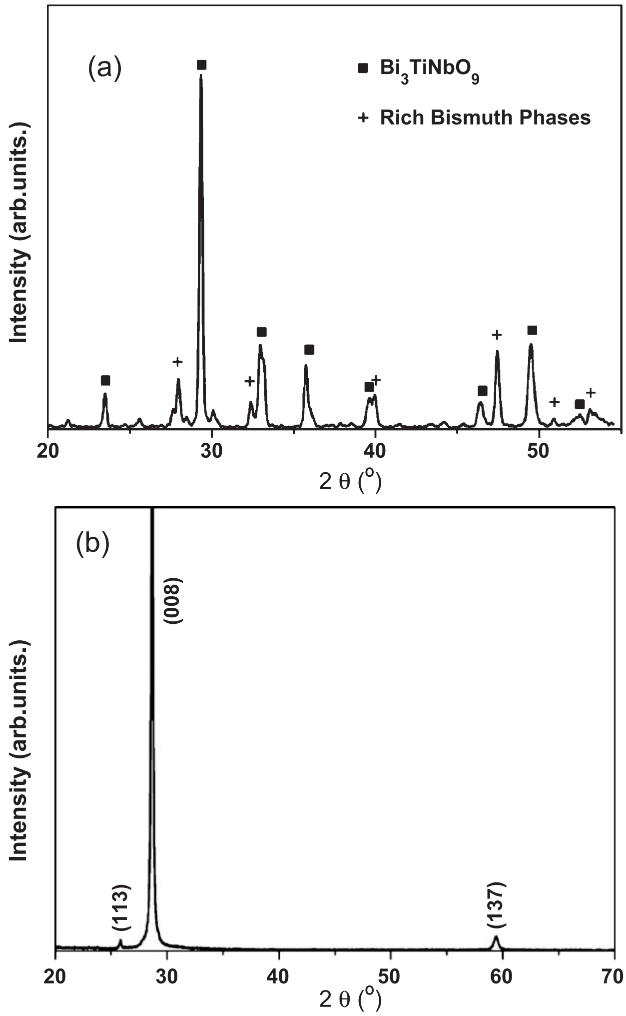

Fig. 1. X-ray θ–2θ scans of (a) BTN BTN targets and (b) thin films deposition on fused silica substrates at the substrate temperature of 600 °C.

alcohol solution prepared by immersion in a water bath at 90 °C was added to the mixed powders in the ratio of 1:95 (polyvinyl alcohol solution to the mixed powders). The screened uniform mixtures of the powder were pressed under pressure of 10 MPa to form pellets of diameter 2 cm. Finally; the pellets were sintered at 1000 °C for 2 h in a conventional box furnace. Thus we acquired the dense yellowish pellets.

The PLD experiments were performed using a KrF excimer pulsed laser of 248 nm wavelength, 30 ns pulse width and a 5 Hz frequency. The films were deposited on fused silica substrates of dimensions 10×5 mm. The deposition temperature, ambient pressure, and laser fluence were optimized at 600 °C, 200 mTorr, and 2.0 J/cm^2, respectively. In order to get a uniform thin film of BTN, the target and the substrate holder were rotated during the deposition procedure. After deposition, all the films were annealed in situ for 30 min with 0.5 atm O_2 pressure. The microstructure and the morphology of the as grown films were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. The optical transmittance spectra of the BTN thin films were measured in the wavelength range of 200–1100 nm using UV/VIS spectrometer. The optical waveguiding properties of the film were investigated by the prism-coupler method. In this method, a He–Ne (632.8 nm) laser beam was coupled into the films using a high refractive index rutile (TiO_2) prism, which was pressed against the thin film surface. The refractive index of the prism is 2.45.

3. Results and discussion

Fig. 1(a) and (b) show the XRD diffraction spectra of BTN targets and the as-deposited BTN films obtained using Cu Kα radiation, respectively. Because of the exceed 20% mol of bismuth ion in the mixed powder, some rich bismuth phases were observed in BTN targets, as show in Fig. 1(a). But for the BTN thin films, there is not any diffraction peak from rich bismuth phases. All the peaks belong to BTN orthorhombic phase. It means that excess 20% mol bismuth ions in starting materials can compensate the Bi evaporation during preparing BTN targets and thin films. Even though fused silica has no direct relation in lattice parameter with the Aurivillius BTN unit cell, a preferential polar axis (00l) orientation texture is observed. This result could be attributed to the intrinsic properties of BTN films, such as differences in surface energies for different planes of the unit cell, and strong interaction between the BiO_6 octahedrons [7]. The relative intensity between (00l) and other peaks have been estimated by measuring the ratio of the (*hkl*) planes to other planes [8]. For the BTN films, The relative intensity of BTN (008) is 0.96. The as-grown BTN thin films fabricated on fused silica substrates are highly *c*-axis oriented texture. The BTN films have a polycrystalline structure.

Optical transmission measurements in the UV/VIS wavelength range have been realized to determine the

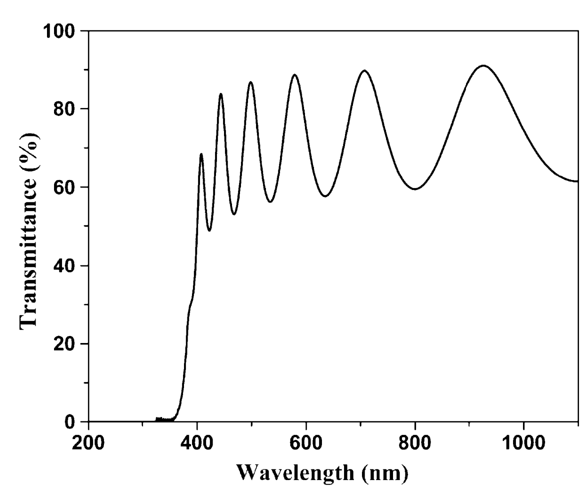

Fig. 2. The wavelength dependence of the transmittance of the 450-nm-thick BTN thin films.

dispersion of the refractive index. Fig. 2 shows the wavelength dependence of the optical transmittance of a BTN thin film. The well oscillating optical transmittance demonstrates that the films have a flat surface and a uniform thickness. The transmittance decreases to zero at approximately 352 nm. For wavelengths longer than 352 nm, the films are transparent. A transmission coefficient value of 75% is measured for wavelengths ranging from 500 to 1100 nm. A band gap energy E_g of 3.55 eV is directly derived from the $\alpha^2(hv)$ plot first time.

The refractive indices have been examined by using the prism-coupling method [10,11]. This technique has been widely used as a powerful tool to investigate the main optical measurement of thin films. In particular, the optical constants and the film thickness can be characterized accurately at a point of sample surface from waveguide mode information. Coupling curves for a BTN thin film fabricated on fused silica substrate are shown in Fig. 3. The narrow coupling width is indicative of a smooth film with uniform thickness. From the angular position of transverse electric (TE) and transverse magnetic (TM) guided modes, we deduced the corresponding effective indices. These values are then used to compute the thin films parameters, such as refractive index and thickness [11]. Typical refractive indices measured for all films are 2.35 and 2.39 in the TE and TM modes, respectively. The thickness value was determined to be 420 nm. Fig 4 shows the cross sectional morphology of the as-deposited BTN films. The interface of the substrate and the BTN film is quite sharp, indicating that there is no obvious interdiffusion between the fused silica and the BTN film. The film's thickness is about 450 nm, which is in good agreement with the prism-coupling study.

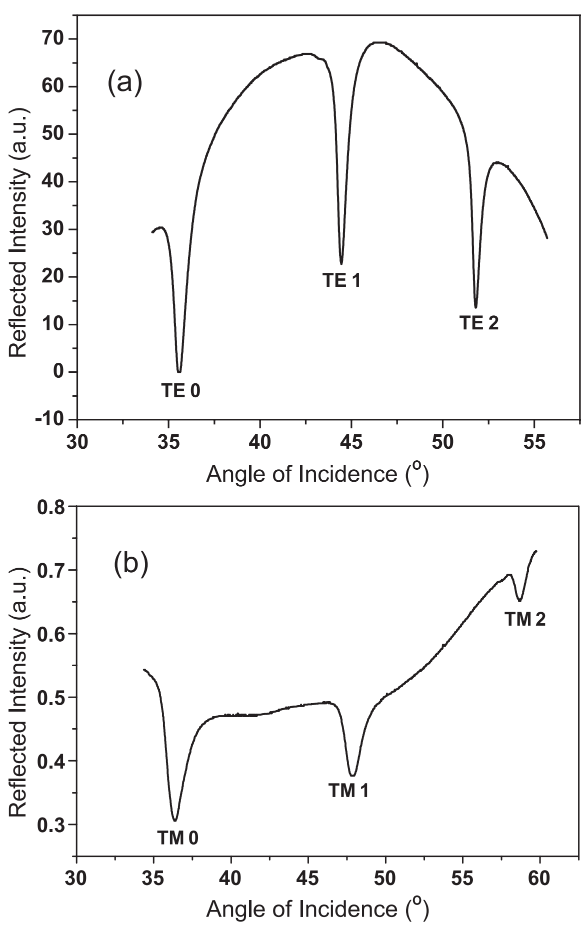

Fig. 3. (a) TE and (b) TM guided mode spectrum for a 450-nm-thick BTN thin film deposited by PLD on fused silica substrate.

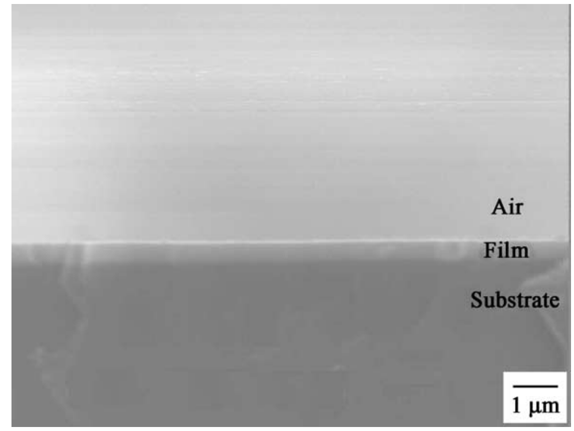

Fig. 4. SEM cross sectional morphology of a 450-nm-thick BTN thin film onto fused silica substrate.

4. Conclusion

In summary, the high c-axis orientation BTN polycrystalline films, where the degree of the relative intensity is 96%, have been prepared on fused silica substrates by pulsed laser deposition. We have investigated the structural and optical properties of the obtained planar waveguides using XRD, UV/VIS spectrometer and the prism-coupling technique. The high quality of BTN films was confirmed by the XRD. The as-grown films have an optical transmittance of 75% in the range of 400–1100 nm and the band gap is E_g=3.55 eV. In a BTN film three TE and three TM modes were observed. All of the m lines of the excited modes are sharp and clear. The refractive indices of TE and TM modes are 2.35 and 2.39, respectively. The favorable optical waveguiding performance achieved in the BTN film deposited on used fused silica implies the possibility of using the waveguiding film for integrated optics and optically active devices.

Acknowledgments

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