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Fabrication and characterization of epitaxial $Sr_{0.6}Ba_{0.4}Nb_2O_6/La_{0.7}Sr_{0.3}CoO_3$ heterostructures

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Abstract

Epitaxial $Sr_{0.6}Ba_{0.4}Nb_2O_6$(SBN60)/$La_{0.7}Sr_{0.3}CoO_3$ heterostructures were fabricated on LAO(0 0 1) substrates using pulsed laser deposition (PLD). Their structural properties were investigated by X-ray diffraction. The θ–2θ scans showed single crystalline $Sr_{1-x}Ba_xNb_2O_6$ (SBN) and $La_xSr_{1-x}CoO_3$ (LSCO) layers with a $\langle 0\ 0\ 1 \rangle$ orientations perpendicular to the substrate plane. Phi scans on the (2 2 1) plane of SBN layer indicated that the films have two in-plane orientations with respect to the substrate. The SBN unit cells were rotated in the plane of the film by $\pm 18.4^\circ$ as well as $\pm 45^\circ$ with respect to the LAO substrate. This rotation was explained by considering the lattice matching between films and substrate, and minimization of electrostatic energy. Spectroellipsometry (SE) was used to characterize the depth profile, the microstructural inhomogeneities, including voids and surface roughness, refractive indices and extinction coefficients of the films.

Keywords: Epitaxial; Heterostructures; Pulsed laser deposition

1. Introduction

Ferroelectric $Sr_{1-x}Ba_xNb_2O_6$ (SBN) has tetragonal tungsten-bronze (TTB) structure and forms solid solutions in the range of $0.25 < x < 0.75$ [1]. It is one of the most important ferroelectric materials for various applications due to its significant spontaneous polarization (34 $\mu C/cm^2$) occurring along *c*-axis and its Curie temperature that can be continuously changed in the range from 60 to 250 °C by controlling the Sr/Ba ratio. SBN has remarkable linear electric-optic characteristics and can be applied in the area of photorefractive devices [2]. Furthermore, reports showed that SBN can be used as pyroelectric detector for infrared radiation with a response time less than 30 ns at room temperature due to its relatively large pyroelectric coefficient (2×10^{-2} $\mu C/cm^2$ K at 27 °C) as well as it has lower oscillatory piezoelectric signal due to mechanical oscillation than other detector [3]. SBN is relatively stable and environmental friendly as compared with lead-base ferroelectric materials. Hence, thin film forms of SBN materials have the potential to be sensor elements compatible with integrated circuits.

For ferroelectric thin films grown on Pt-coated Si wafer with Pt top contact electrodes to form the capacitor structure, the non-ideal interfaces between ferroelectric layers and electrodes lead to degradation for the performance. To solve this problem, conductive oxide electrodes have been used. Among various conductive oxides, $La_xSr_{1-x}CoO_3$ (LSCO) is a commonly used conductive oxide of perovskite structure with lattice parameter matching very well with that of $Pb_xSr_{1-x}TiO_3$ perovskite [4–7]. The pulsed laser deposition (PLD) process takes advantageous over other thin film deposition techniques by the accuracy in composition control and the simplicity in process control [8,9].

In this paper, we used LSCO as the bottom electrode for non-perovskite type ferroelectric material $Sr_{0.6}Ba_{0.4}Nb_2O_6$. We reported the epitaxial growth of SBN thins films on LSCO buffered LAO(0 0 1) single crystal substrate by PLD technique. The structural and optical properties of these SBN/LSCO heterostructure were investigated.

2. Experimental procedures

A KrF excimer laser (Lambda Physik, model Compex 2005) of 248 nm in wavelength and 20 ns in pulse width was used to

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fabricate our films. The LSCO and SBN targets were prepared by sintering pellets of ultrafine powder obtained from ball milling [10]. The distance between the substrate and the target was kept at 6 cm throughout the experiment. The SBN/LSCO/LAO films were deposited in a high vacuum chamber equipped with a rotating multi-target system. The LSCO and SBN films were deposited with a repetition rate of 10 Hz with base pressure of 2×10^{-3} Pa. The laser fluence was about 4 J/cm^2. For depositing LSCO bottom electrode, the pressure of the deposition chamber was kept at 13 Pa ambient oxygen pressure and the substrate temperature was kept at 650 °C. The as-grown LSCO films were then post annealed for 15 min at the same deposition conditions to improve its crystallinity and release thermal stress. For the deposition of SBN, the pressure of the deposition chamber was kept at 27 Pa ambient oxygen pressure and the substrate temperature was kept at 740 °C. After deposition, the as-grown SBN films, similar to LSCO, were post annealed at the deposition temperature and pressure for 15 min. The films were then cooled naturally to room temperature.

The film thickness was measured by an α-step profiler (KLA Tencor P-10). The orientation and crystalline quality of the films were measured by a four-circle X-ray diffractometer (XRD Philip PW3710) equipped with Cu Kα radiation source. Surface morphology of the films was characterized by a scanning electron microscope (SEM, Leico, Stereoscan440) and atomic force microscope (AFM Nanoscope IV). For optical analyses, spectroellipsometry (SE) measurements were carried out by a spectroscopic phase-modulated ellipsometer (Jobin Yvon UVISEL) at photon energy between 1.5 and 4 eV with an interval of 0.01 eV. An incidence angle of 70° was used throughout our ellipsometric measurements [11,12].

3. Results and discussions

The XRD θ–2θ profiles of the SBN/LSO/LAO films are shown in Fig. 1(a). The pattern only shows SBN(0 0 1) reflection indicating a *c*-axis preferred orientation growth. The out-of-plane orientation of the SBN(0 0 2) peaks was studied by ω-scan. In Fig. 1(b) the full-width-at-half-maximum (FWHM) of the rocking curve was 0.35°. It was comparable with the FWHM of LAO(0 0 2) which was 0.14° as shown in Fig. 1(c). This indicates that SBN films were grown normal to the substrate with highly orientated.

The XRD Φ-scans of SBN(2 2 1), LSCO(1 1 1) and LAO(1 1 1) as shown in Fig. 2 were performed to confirm the epitaxial growth of the films. Two sets of in-plane orientation rotations of ±18.4 and ±45° from the substrate ⟨1 0 0⟩ direction are observed. The ±18.4° in-plane rotation have been reported previously in SBN films deposited on MgO [13–15]. SBN is a layered material with atoms concentrated in *c*-planes. Similar to those SBN films grown on MgO substrates, our SBN films have preferential growth direction by shifting its lattice ±18.4° with respect to the substrate (0 0 1) plane to minimize the electrostatic repulsion [13]. The epitaxial relationship between the SBN and LSCO/LAO can be described as SBN(0 0 1)//LSCO(3 1 0)//LAO(3 1 0). The lattice constant of SBN and LSCO are 12.45 and 3.90 Å, respectively. In this configuration, a single SBN lattice cell is grown on top of several LSCO lattice cells as shown in the inset of Fig. 2. As a result, the lattice mismatch between SBN and LSCO is about 1%. Hence, this epitaxial relationship is preferable due to the domain matching and electrostatic minimization.

However, the ±45° in-plane rotation has not been reported previously. We proposed that the 45° twisting of SBN unit cell can be described by an epitaxial relationship of SBN(0 0 1)//LSCO(2 2 0)//LAO(2 2 0). With the domain matching shown in the inset of Fig. 2, the lattice mismatch between SBN and LSCO is estimated to be about 11%. This configuration is less favor as compared to the previous configuration due to a larger mismatch. This less favor growth is confirmed by a weaker signal observed in Fig. 2. More detailed will be gained by further studies using transmission electron microscopy (TEM).

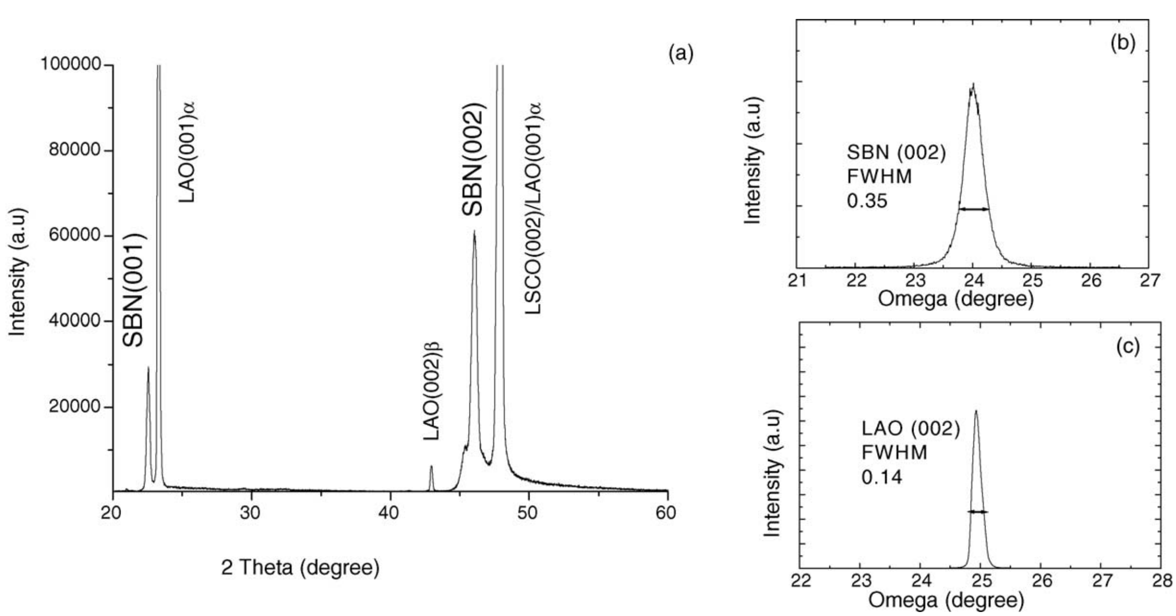

Fig. 1. (a) X-ray diffraction θ–2θ scan patterns of SBN thin film grown on LSCO/LAO. The rocking curves of (b) SBN(0 0 2) and (c) LAO(0 0 2) peaks.