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Temperature dependence of magnetoelectric coupling in $La_{0.7}Sr_{0.3}MnO_3$/$BaTiO_3$ layered heterostructure with various volume fraction

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ABSTRACT

The epitaxial $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3$ (LSMO/BTO) layered heterostructures were grown onto (001) oriented $LaAlO_3$ single-crystal substrate by pulsed laser deposition. Temperature dependence of the magnetoelectric (ME) voltage coefficient (α_E) was studied for the heterostructures over temperature range of −20–85 °C (253.15–358.15 K). The values of α_E for all of the samples exhibit a monotonous decreasing tendency with increasing temperature. Moreover, the variation in ME voltage coefficient as a function of temperature was found to be significantly dependent on the volume fraction of BTO layer. Our results showed that maximum number of the volume fraction of BTO layer can provide more stable ME voltage coefficient in wide temperature range, due to the competition of the effects of temperature influenced piezoelectricity, magnetostriction, and electrical transport property of the constituents. This finding could provide an available method for promote miniaturization of the ME heterostructure devices in view of temperature stability.

1. Introduction

The multiferroics magnetoelectric (ME) layered heterostructures have been extensively studied recently to understand the mechanisms responsible for cross coupling between ferromagnetism and ferroelectricity, which is at the center of their promise for applications in multifunctional devices, such as ME sensors, multiple-state memories, etc. The well established extrinsic ME coupling is mediated through the strain across the interface between ferromagnetic (magnetostrictive effect) and ferroelectric (piezoelectric effect) materials. [1–6] In such system, ME coupling originates from the strain/stress, i.e., the strain induced in one constituent (either by magnetostriction in ferromagnetic phase or by converse piezoelectric effect in ferroelectric phase) is transferred to the others and alters its polarization or magnetization. Additionally, the other mechanisms of the ME coupling may be mediated by free carriers at the interface between the ferromagnetic and ferroelectric oxidation. [7–9] In this case, an electric field results in the accumulation or depletion of spin-polarized carriers at the interface producing a change in the interface magnetization. Because a strong ME effect can be realized in this system at room temperature and used in a wide temperature range. So, it can be used for technological device applications, including magnetic field sensors, current sensors, actuators, transducer, tunable devices, resonators, and so on. [10–12].

The interaction between the strain-mediated ME coupling and interface-charges driven ME coupling leads to a remarkable volume fraction (ferromagnetic or ferroelectric phase) and temperature dependent ME behavior [13]. It is well known, most of devices have to undergo a wide ambient temperature range; however, most of the studies so far are performed at room temperature (around 300 K). Thus, the study of temperature dependence of ME properties is of technological importance for both understanding the physics of ME coupling and application of the ME heterostructures as devices [14–18]. On the other hand, there are some studies correlated to the effect of thickness of either ferromagnetic or ferroelectric layer on magnetoelectric, electrical, magnetic properties for bilayer films in which the magnetic layer is LSMO [19,20]. However, studies on the effect of the temperature on ME voltage coefficient by adjusting the geometric construction (volume fraction of ferromagnetic or ferroelectric phase) of ME heterostructures seem rather rare.

In this paper, the epitaxial $La_{0.7}Sr_{0.3}MnO_3/BaTiO_3$ (LSMO/BTO) layered heterostructures with different volume fraction of BTO (by adjusting the thickness of LMSO) were prepared. Variation of ME voltage coefficient was measured as a function of temperature (−20 to 85 °C) and at room temperature. The study aim is examine the influence of geometric construction (volume fraction of ferroelectric phase BTO)

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on the temperature dependent ME voltage coefficients for such heterostructures. This study would be beneficial to understanding the temperature dependent ME coupling, and the appropriate structure design for practical device applications.

2. Experimental

The LSMO and BTO films were grown in turn onto the (001) oriented $LaAlO_3$ (LAO) substrates (10 mm × 5 mm × 0.5 mm) by pulsed laser deposition technique. The stoichiometric $La_{0.7}Sr_{0.3}MnO_3$ and $BaTiO_3$ ceramic target were ablated using a KrF excimer laser at a wavelength of 248 nm with 1.5 J/cm^2 and 5 Hz. During deposition, the substrate temperature of 700 °C and a flowing oxygen pressure of 10 Pa were maintained, and the deposition distance was 12 cm. After deposition, all of the LSMO/BTO heterostructures were in-situ annealed at 800 °C about 30 min at oxygen pressure of 1000 Pa.

X-ray diffraction (Bruker D8) measurement was employed to probe the phase structure. The film thickness was measured by using a Step profiler (Surfcorder ET150). Magnetic properties were measured by a physical property measurement system (PPMS-9). The ferroelectric hystersis loops were recorded with a ferroelectric analyzer (TF2000E). The ME measurement was carried out on the all LSMO/BTO layered heterostructures by changing the constituent volume fraction. The LSMO/BTO bilayers possessed various FE phase (BTO) volume fraction (ν). Here, $\nu = \frac{d_{FE}}{(d_{FE}+d_{FM})}$, d_{FE} and d_{FE} denoted the thickness of ferroelectric (BTO) layer and ferromagnetic (LSMO) layer, respectively.

The ME voltage coefficients were measured by subjecting the sample to a stationary bias magnetic field ($H_{bias} = 2000$ Oe) and a parallel alternating magnetic field ($H_{AC} = 10$ Oe) at frequency 0.1 kHz–100 kHz. Heating and cooling of the sample were carried out by utilizing a semiconductor refrigerator. The sample was placed on the center of the sample stage with a thermocouple close beside, so the thermocouple could detect the sample temperature, which approximately reflects the real-time temperature of the sample. The transverse ME voltage coefficients were recorded with the superimposed sinusoidal and static magnetic field applied parallel to sample surface. The schematic of temperature dependence ME measurement and the cross section of LSMO/BTO layered heterostructure were presented in Fig. 1.

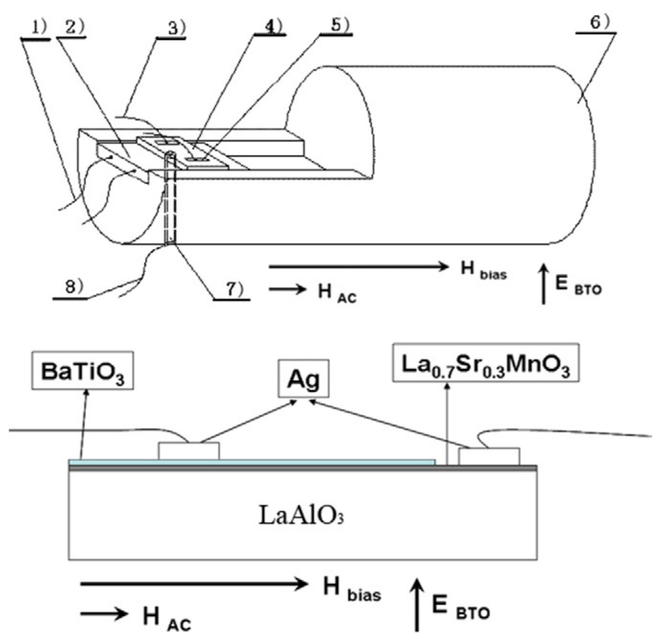

Fig. 1. schematic of temperature dependence ME measurement and the cross section of the LSMO/BTO layered heterostructure. 1). Lead wire of semiconductor refrigerator 2). semiconductor refrigerator (TEC1-12706) 3). Lead wire of LSMO/BTO heterostructure 4). LSMO/BTO heterostructure 5). Ag electrode on the top of LSMO/BTO heterostructure 6). Sample stage (PE) 7). Thermal couple meter (HT-1008F) 8). lead wire of thermal couple.

3. Results and discussion

Fig. 2 presents the XRD patterns of the LSMO/BTO layered heterostructures. Here, the thickness of BTO layer is always 100 nm for the all samples, and the thicknesses of LSMO layer are 100 nm, 20 nm, and 10 nm, and the corresponding values of ν (volume fraction of BTO) are 0.5, 0.83, and 0.91, respectively. As it is shown in the figure, only (00*l*) diffraction peaks of the LSMO and BTO films can be observed. No other peaks are detected in these patterns, which indicate that high quality LSMO and BTO films are grown epitaxially on LAO substrate. On the other hand, with decrease of ν, the position of (00*l*) peaks for LSMO film shifts to high angle, and the corresponding out-of-plane (along the film growth direction) lattice constant of LSMO decreases. As we all known, the bulk lattice parameters of BTO (a-axis), LSMO (pc-pseudocubic), and LAO (pc-pseudocubic) are 0.3995 nm, 0.3874 nm, and 0.3793 nm, respectively, the LSMO films deposited on LAO substrate and the BTO film deposited on LSMO layer will suffer a large biaxial compressive stress due to the lattice mismatch. The compressive stress makes the in-plane lattice constant of the LSMO film becomes lower, while the out-of-plane lattice constant becomes larger than that of stress-free LSMO, the smaller in-plane lattice constant, the larger out-of-plane lattice constant. As a result, with decrease of ν (increasing of LSMO thickness), the out-of-plane lattice constant of LSMO film will decrease due to the strain relaxation. That is the reason of the volume fraction dependence for LSMO (00*l*) peak.

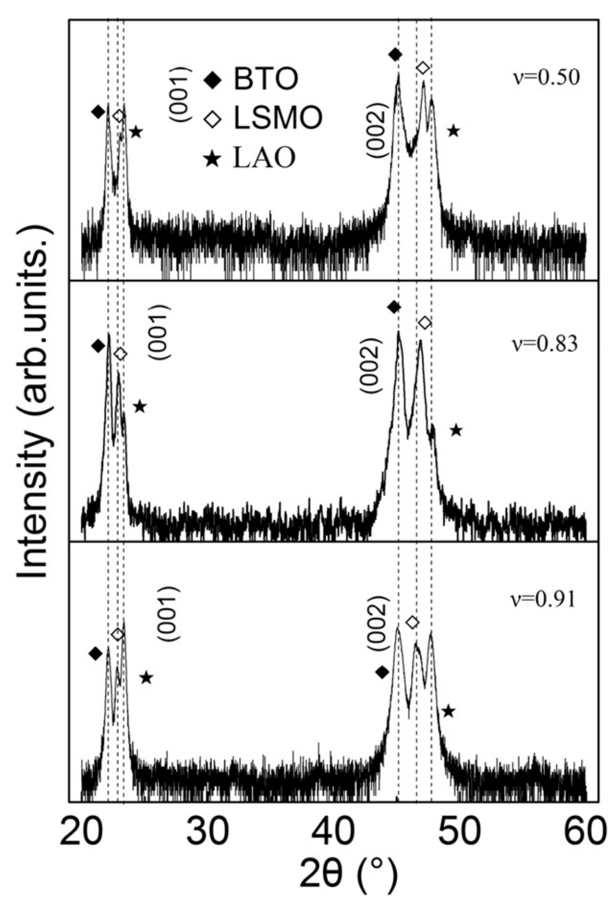

Fig. 2. The XRD patterns of LSMO/BTO layered heterostructures with various volume fraction of BTO.