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Ferroelectric domain structures and polarization switching characteristics of polycrystalline $BiFeO_3$ thin films on glass substrates

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ABSTRACT

We investigated ferroelectric characteristics of $BiFeO_3$ (BFO) thin films on $SrRuO_3$ (SRO)/yttria-stabilized zirconia (YSZ)/glass substrates grown by pulsed laser deposition. YSZ buffer layers were employed to grow highly crystallized BFO thin films as well as SRO bottom electrodes on glass substrates. The BFO thin films exhibited good ferroelectric properties with a remanent polarization of $2P_r = 59.6\ \mu C/cm^2$ and fast switching behavior within about 125 ns. Piezoelectric force microscopy (PFM) study revealed that the BFO thin films have much smaller mosaic ferroelectric domain patterns than epitaxial BFO thin films on Nb:$SrTiO_3$ substrates. Presumably these small domain widths which originated from smaller domain energy give rise to the faster electrical switching behavior in comparison with the epitaxial BFO thin films on Nb:$SrTiO_3$ substrates.

1. Introduction

Recently considerable attention has been paid to magnetoelectric multiferroics due to their various physical properties and potential application [1–4]. Among these materials, $BiFeO_3$ has been studied as an attractive lead-free candidate because it exhibited not only ferroelectricity with a Curie temperature of ~1103 K but also antiferromagnetic property with a Néel temperature ~643 K [5,6]. A direct coupling between ferroelectric and ferromagnetic order parameters offers the potential for memory device application and advanced spintronics [2,7].

Meanwhile, ferroelectric properties of BFO thin films have been widely investigated on the effect of the preferred orientaiton in their growth [8–10]. By controlling the texture of the perovskite bottom electrodes and using proper single crystal substrates, we can properly obtain the preferred orientation of BFO thin films [8–12]. More importantly, it is necessary for us to enable ferroelectric thin films to be deposited on Si substrates or other substrates for their practical applications. However, using single crystal substrates in the industry is not recommended due to their high cost. For this reason, glass is frequently regarded as a substitute for substrates in the devices fabrication. Yttria-stabilized zirconia (YSZ) is known to be an attractive buffer layer on glass for the subsequent deposition of perovskite oxide thin films, because it is well crystallized on glass substrates even at room temperature [13,14].

Accordingly, we fabricated BFO thin films on $SrRuO_3$(SRO)/YSZ/glass substrates by pulsed laser deposition (PLD). YSZ buffer layers were used for the deposition of SRO bottom electrodes and BFO thin films on glass substrates. Previously, we have shown the multiferroic properties of highly *c*-oriented BFO thin films on glass substrates [14]. In this work, ferroelectric domain structure and electrical switching behavior of BFO thin films on SRO/YSZ/glass substrates are further examined.

2. Experimental

We deposited well crystallized BFO thin films in the thickness of 100 nm by pulsed laser deposition (PLD) onto glass substrates. The conducting perovskite SRO electrodes and YSZ buffer layers were chosen to ensure heteroepitaxial growth. We used fused silica glass substrates. Commercially available 1 inch $Y_{0.2}Zr_{0.8}O_3$ target was first used for the YSZ buffer layers on glass substrates. A frequency tripled (355 nm, ~2 J/cm^2) Nd:yttrium aluminum garnet laser was used for the deposition, and the distance between target and substrate was 5 cm. The substrate temperature was set to 700 °C with oxygen partial pressure range of 1–4 mTorr after the base pressure reached 5×10^{-7} Torr. We also deposited SRO thin films on YSZ buffer layers for the bottom electrodes, since SRO is widely known as good electrode for BFO thin films [15]. For the optimal deposition condition of SRO thin films, we set 700 °C for the

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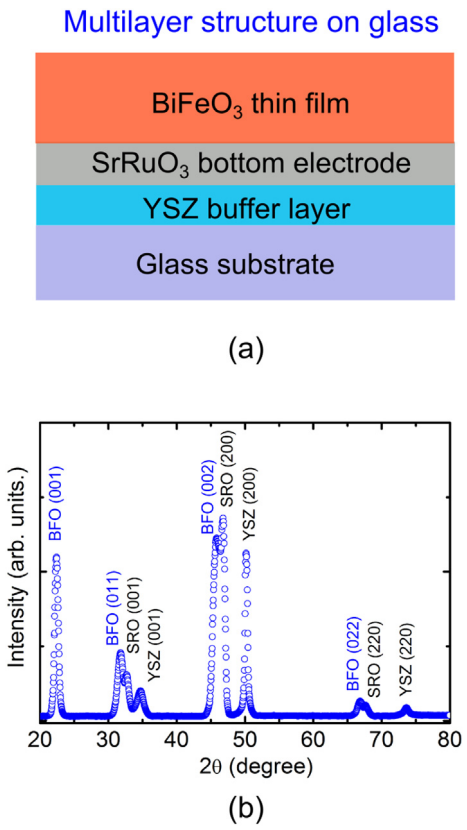

Fig. 1. (a) A schematic diagram of BFO thin film on/SRO/YSZ/glass substrate. (b) The XRD peaks of the BFO thin film on the highly *a*-oriented and (110)-oriented SRO bottom electrodes.

substrate temperature with oxygen partial pressure of 100 mTorr. Subsequently, 1 inch $Bi_{1.2}FeO_3$ pellets were used for the deposition of BFO thin films, in which we increased Bi concentration just in case of Bi volatilization. After the base pressure reached 5×10^{-7} Torr, the substrate temperature was kept to 800 °C with oxygen partial pressure of 100 mTorr. We cooled down to room temperature in oxygen ambient at 300 Torr after the BFO thin films deposition. For the measurement of ferroelectric polarization hysteresis loops, we made BFO thin film capacitors with thickness of 100 nm. Radio frequency (RF) magnetron sputtering was used to fabricate Pt top electrodes which were circular shapes with diameter of 100 um and thickness of 100 nm. These capacitor samples (Pt/BFO/SRO/YSZ/glass) were annealed at 400 °C for 5 min before measuring ferroelectric hysteresis loops. For comparison, epitaxial BFO thin films were also grown on Nb-doped $SrTiO_3$ substrates in the same experimental procedure. The structure of BFO thin film on glass was investigated by X-ray diffraction (XRD, CuK radiation 1.542 Å). To obtain ferroelectric hysteresis loops of BFO thin films, we used a RT66A test system (Radiant Technologies, Inc.) The surface morphology and ferroelectric domain structure were investigated by atomic force microscopy (AFM) and piezoelectric force microscopy (PFM) respectively.

3. Results and discussion

Fig. 1a illustrates a schematic diagram of our sample configuration which describes sequential deposition from upper layer to lower layer, *i.e.* BFO thin film, SRO bottom electrode, and YSZ buffer layer on glass substrate (BFO/SRO/YSZ/glass). We first investigated the crystal structure of the multilayer thin films on glass substrate by XRD measurement as shown Fig. 1b. The YSZ thin film for the buffer layer shows the highest intensity of (100) peak with low *full width at halt-maximum* (FWHM) of 0.7°, which indicates highly *a*-oriented growth on glass substrate. Here, the lattice constant of YSZ with cubic crystal structure was about 3.634 Å. As stated earlier, we could obtain good crystallinity of YSZ buffer layer on glass substrate from temperature of 700 °C. SRO bottom electrode was fabricated on highly *a*-oriented and (110)-oriented YSZ buffer layer prior to the deposition of BFO thin film. Compared to YSZ buffer layer, XRD peaks of the SRO bottom electrode exhibited higher intensity with FWHM below 1.2°, demonstrating good crystallinity. On the highly *a*-oriented and (110)-oriented YSZ buffer layer, each SRO bottom electrode presents the same orientation with corresponding YSZ layer. The lattice misfit between SRO electrode film and YSZ layer was about 7% due to compressive strain. Although not shown here, the only (110)-oriented SRO thin film could be obtained when we deposited SRO bottom electrode on glass without the YSZ buffer layer. It is worth noting that the highly *a*-oriented YSZ buffer layer plays a key role in the highly *a*-oriented growth of the SRO bottom electrode. For such reason, the well-crystallized YSZ buffer layer on glass substrate can improve the crystallinity of BFO thin film as well as SRO bottom electrode. The XRD peaks of BFO thin film were obtained on highly *a*-oriented and (110)-oriented SRO bottom electrodes. We could obtain a highly *c*-oriented BFO thin film on the highly *a*-oriented SRO bottom electrode. Notably, (011)-oriented BFO thin films were also deposited on highly *a*-oriented and (110)-oriented SRO bottom electrodes. The XRD peaks of BFO thin film also showed low FWHMs below 1° with high intensity, which implies a good crystallinity. Generally, the crystal structure of BFO (bulk) at room temperature is classified as a rhombohedral belonging to the space group R3c [16,17]. However, the polycrystalline BFO thin film in this study had the mixed crystal

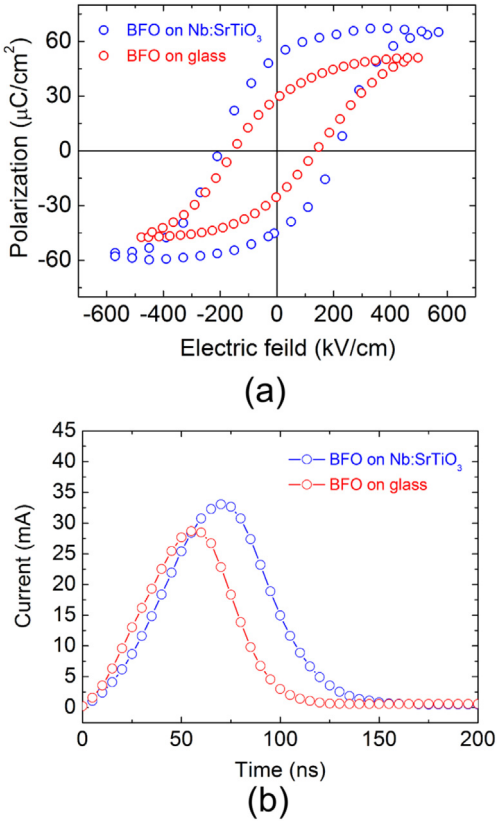

Fig. 2. (a) The ferroelectric polarization hysteresis loops (*P-E*) of the BFO on glass and epitaxial BFO on Nb:$SrTiO_3$ at a measurement frequency of 10 kHz. (b) The switching current as a function of time for BFO thin films.