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Selective orientation of $SrBi_4Ti_4O_{15}$ thin films grown on buffered Si(100) substrates

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Abstract Many efforts have been paid to uncouple the spontaneous polarizations of layer-structured bismuth-based ferroelectrics along different crystal orientations, obtaining these layered-structure films with non-*c*-axis orientations. In the paper, $SrBi_4Ti_4O_{15}$ (m=4) thin films have been deposited on Pt/MgO bilayer-buffered Si(100) substrates by pulsed-laser deposition. Selective orientation of $SrBi_4Ti_4O_{15}$ thin films mediated by different epitaxy relationships between electrode layers and MgO/Si substrates has been demonstrated. Furthermore, different hysteresis loops and remnant polarization of $SrBi_4Ti_4O_{15}$ thin films with varied orientations have been obtained.

Keywords Layer-structured · Ferroelectrics · Non-*c*-axis orientations · Thin films

1 Introduction

Thin films of layer-structured bismuth-based ferroelectrics described as $(Bi_2O_2)^{2+}(A_{m-1}B_mO_{3m+1})^{2-}$ (also known as the Aurivillius family of compounds, where A is mono-, di-, or trivalent ions or a mixture of them; B is tetra-, penta-, or hexavalent ions; m is the number of BO_6 octahedron in the pseudoperovskite layer), such as $SrBi_2Ta_2O_9$ (m=2) or $Bi_{4-x}La_xTi_3O_{12}$ (m=3), have been extensively investigated due to the discovery of their superior fatigue endurance, which is crucial for applications in ferroelectric random-access memory devices.[1, 2] However, *c*-axis oriented films are well-known to have a very small polarization component along the films' normal, because the vector of the maximum spontaneous polarization in these layered perovskite materials lies in the *a–b* plane.[3] To uncouple the spontaneous polarizations along different crystal orientations, efforts have been focused on the growth of these layered-structure films with non-*c*-axis orientations. These orientations have the spontaneous polarization vectors inclined to the *a–b* plane. In the present paper, ferroelectric $SrBi_4Ti_4O_{15}$ (m=4) thin films have been deposited on Pt/MgO bilayer-buffered Si(100) substrates by pulsed-laser deposition. Selective orientation of $SrBi_4Ti_4O_{15}$ thin films mediated by different epitaxy relationships between electrode layers and MgO/Si substrates has been demonstrated. Furthermore, different hysteresis loops and remnant polarization of $SrBi_4Ti_4O_{15}$ thin films with varied orientations have been obtained.

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2 Experimental procedure

$SrBi_4Ti_4O_{15}$/Pt/MgO/Si heterostructures were fabricated by pulsed-laser deposition. The detail of deposition conditions and epitaxial growth of Pt/MgO bilayer was described in [4]. Through ablating a stoichiometric single-phase target, $SrBi_4Ti_4O_{15}$ thin films with the same thickness of 300 nm were grown by pulsed-laser deposition at a substrate temperature of 600 °C and an O_2 partial pressure of 0.4 mTorr. A Lambda Physik KrF excimer laser (COMPex201, λ=248 nm), operating at repetition rates of 5 Hz with energy density of ×7 J/cm^2, was used for the deposition. Following the deposition, the films were first cooled slowly in an oxygen pressure of 0.4 mTorr to room temperature, and then, experienced a Rapid-Thermal-Annealing (MODEL RTP-300) process under an oxygen flow and temperature of 2.5 L/min and 580 °C, respectively. Top iridium electrode dots of 0.04 mm^2 area were also pulsed-laser deposited through mask techniques to form MIM capacitors. Crystallinity of the heterostructures was examined by X-ray diffraction (XRD; D/MAX-2550V Cu Kα) and measurements of polarization loops were performed with a standard Sawyer–Tower circuit at a frequency of 60 Hz.

3 Results and discussion

Figure 1 shows the XRD patterns for $SrBi_4Ti_4O_{15}$ thin films grown on Pt/MgO bilayer with epitaxial orientations of Pt(100)/MgO(100), Pt(111)/MgO(100) and Pt[(100)+(111)]/MgO(100), respectively. For brevity, we also denote those $SrBi_4Ti_4O_{15}$ thin films as SBT-1, SBT-2 and SBT-3, as shown in Fig. 1. It can be clearly seen that *c*-domains predominated the crystalline orientation of SBT-1, which is grown on Pt(100)/MgO(100) bilayer. This *c*-axis orientation of SBT-1 should be induced by the epitaxial orientation of Pt (100) planes. Whereas, $SrBi_4Ti_4O_{15}$ thin films exhibit apparent non-*c*-axis orientations, viz. SBT(119), on Pt(111)/MgO(100) and Pt[(100) + (111)]/MgO(100) bilayers. Especially for SBT-3, only the diffraction peak of SBT(119) was detected. Why can the co-existence of Pt(111) and Pt(100) orientations greatly suppress the crystallographic growth along *c*-axis? It is not very clear at present. But it is suggested that clamping effects of two intersecting Pt crystalline planes with two different out-of-plane orientations must play an important role in determining the out-of-plane orientations of $SrBi_4Ti_4O_{15}$ thin films. More work is presently being pursued to develop a comprehensive mechanism, which will adequately describe the phenomena.

Figure 2 shows ferroelectric hysteresis loops recorded from SBT-1 [(11*l*)+(00*l*)—oriented], SBT-2 [(119)+(00*l*)—oriented] and SBT-3 [(119)—oriented], respectively. Evidently, the ferroelectric anisotropy of $SrBi_4Ti_4O_{15}$ thin films is demonstrated: the measured remnant polarization ($2P_r$) for SBT-1, SBT-2 and SBT-3 are 3.6, 7.4 and 11.8 $\mu C/cm^2$, respectively. Although the values are relatively small for practical applications, an effective way to enhance the ferroelectric properties of layer structured bismuth based ferroelectric thin films is provided. It is expected that, by optimizing the growth techniques and fabrication conditions, non-*c*-axis oriented $SrBi_4Ti_4O_{15}$ thin films with higher remnant polarization could be deposited onto Pt/MgO bilayer-buffered Si substrates.

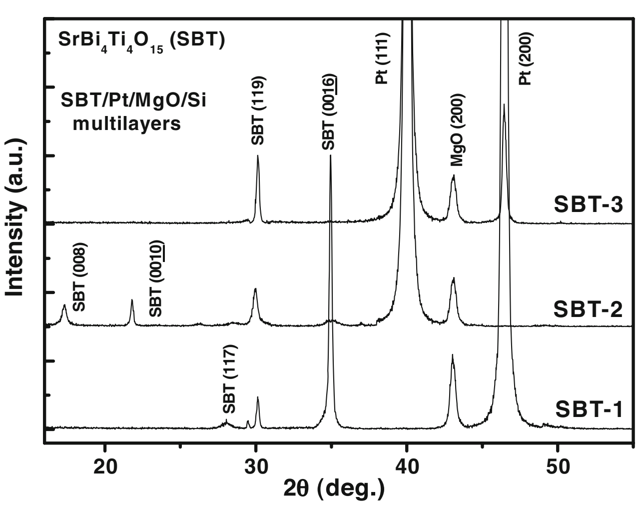

Fig. 1 Selective orientation of $SrBi_4Ti_4O_{15}$ thin films mediated by different epitaxy relationships between electrode layers and MgO/Si substrates

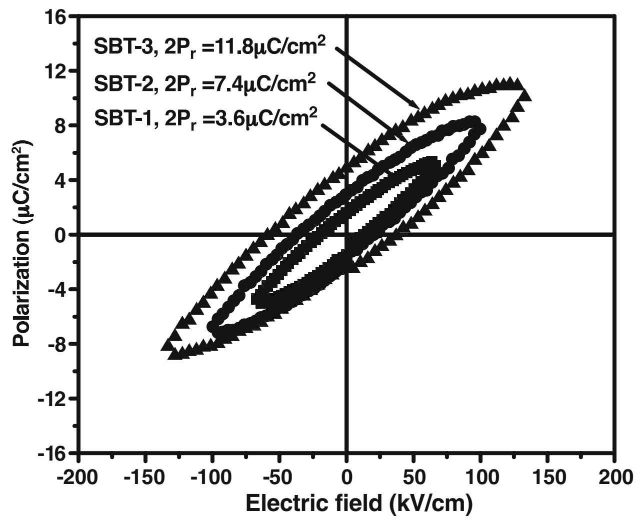

Fig. 2 Hysteresis loops of the $SrBi_4Ti_4O_{15}$ thin films with selective out-of-plane orientations

4 Conclusion

Ferroelectric $SrBi_4Ti_4O_{15}$ (m=4) thin films have been deposited on Pt/MgO bilayer-buffered Si(100) substrates by pulsed-laser deposition. Selective orientation of $SrBi_4Ti_4O_{15}$ thin films mediated by different epitaxy relationships between electrode layers and MgO/Si substrates has been obtained. By measurements of ferroelectric hysteresis loops, the ferroelectric anisotropy of $SrBi_4Ti_4O_{15}$ thin films is demonstrated.

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