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Microstructural Properties of Piezoelectric 0.95($Na_{0.05}Bi_{0.05}TiO_3$)-0.05($BaTiO_3$) Thin Films on $SrTiO_3$ (001) Substrates

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Lead-free piezoelectric materials of sodium bismuth titanate (NBT)-based compounds, e.g. (1-x)($Na_{0.5}Bi_{0.5}TiO_3$)-x($BaTiO_3$) (NBT-BT) solid solutions, have been extensively studied due to their excellent dielectric and piezoelectric properties [1-3]. Nevertheless, the strong deviation of the electrical properties from the bulk has been observed in the NBT-BT films, e.g. high leakage current density [4]. It is known that the microstructure, structural defects and chemical inhomogeneity can strongly affect the physical properties of materials and thus affect the performance of the related devices. In spite of the extensive studies on physical properties and thin film growth [5], the structural properties of the NBT-BT films at the atomic scale still remain unclear.

Thin films of 0.95NBT-0.05BT were deposited on $SrTiO_3$ (001) substrates at 850°C using the high-pressure sputtering system. The mixed ambient of Ar and O_2 at the ratio of 1:1 was used with a flowing pressure of 0.2 mbar during the film deposition. Cross-sectional specimens for (scanning) transmission electron microscopy ((S)TEM) investigations were prepared by both argon ion milling method and focused ion beam (FIB) technique (FEI Dualbeam Helios NanoLab 600i). To explore the microstructural properties of 0.95NBT-0.05BT, atomic-resolution high-angle annular dark-field (HAADF) and annular-bright field (ABF) imaging were performed on a JEOL ARM200F with a probe aberration corrector, operated at 200 kV.

Figure 1(a) shows a low-magnification bright-field (BF) TEM image of 0.95NBT-0.05BT films on $SrTiO_3$ (001) substrates. The contrast difference between 0.95NBT-0.05BT and $SrTiO_3$ is visible, which allows us to determine the 0.95NBT-0.05BT/$SrTiO_3$ interface. Contrast variations can also seen in the image, which is due to the existence of planar defects in the 0.95NBT-0.05BT films. In most cases, the planar faults show the habit plane parallel to the {100} plane of 0.95NBT-0.05BT and either terminate within films or penetrate the whole films, as indicated by a red and a white arrow, respectively. A selected-area electron diffraction (SAED) pattern is inserted in Figure 1(a), which was recorded from the films and part of the $SrTiO_3$ substrate. Based on the zone axis SAED pattern, the crystallographic orientation relationship can be determined as $[010]_{NBT\text{-}BT}//[010]_{SrTiO3}$ and $(001)_{NBT\text{-}BT}//(001)_{SrTiO3}$. In addition, we noted that diffuse scattering streaks around reflection spots from the 0.95NBT-0.05BT films appear in the SAED pattern, as indicated by a pair of horizontal arrows. The presence of these diffuse streaks results from a high density of planar defects parallel to the film growth direction.

An atomic-resolution HAADF-STEM image of a typical planar fault in the 0.95NBT-0.05BT films is displayed in Figure 1(b), which is recorded along the [010] zone axis of 0.95NBT-0.05BT. The planar defect consists of edge-sharing oxygen octahedra with Bi atoms at the center of the space formed by

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octahedral network. In Figure 1(b), the structure of the planar defect has a repeat period three times as large as that of 0.95NBT-0.05BT along the film growth direction. This is consistent with the diffuse streaks appeared in the SAED pattern. It should be mentioned that this planar defect is identical to one of planar defects observed in the 0.95NBT-0.05BT films grown on Ni substrates buffered by a NiO layer [6]. The structural model of the planar defect is displayed in Figure 1(c). In fact, an ordered intergrowth of the planar defect and perovskite-type 0.95NBT-0.05BT phase has been observed in the 0.95NBT-0.05BT/$SrTiO_3$ system. Regarding the thickness of 0.95NBT-0.05BT between two adjacent planar defects and relative displacement of adjacent planar defects along the film growth direction, a number of ordered sequences characteristic of new structures could be locally obtained. The formation of the planar defect in the films occurs during the film growth since no planar defects have been observed in the 0.95NBT-0.05BT bulk materials [6]. Considering the difference in properties between thin films and bulk materials, the observed planar defect in the films can play an important role in modifying the film properties [7].

References:

[1] H Wang *et al*, *Mater. Des.* **31** (2010) p. 4403.
[2] Y Guo *et al*, *Chem. Mater.* **23** (2011) p. 219.
[3] Y Kitanaka *et al*, *Phys. Rev. B* **89** (2014) p. 104104.
[4] MM Hejazi, E Taghaddos and A Safari, *J. Mater. Sci.* **48** (2013) p. 3511.
[5] SK Acharya *et al*, *J. Alloy Compd.* **603** (2014) p. 248.
[6] SB Mi *et al*, submitted for publication.
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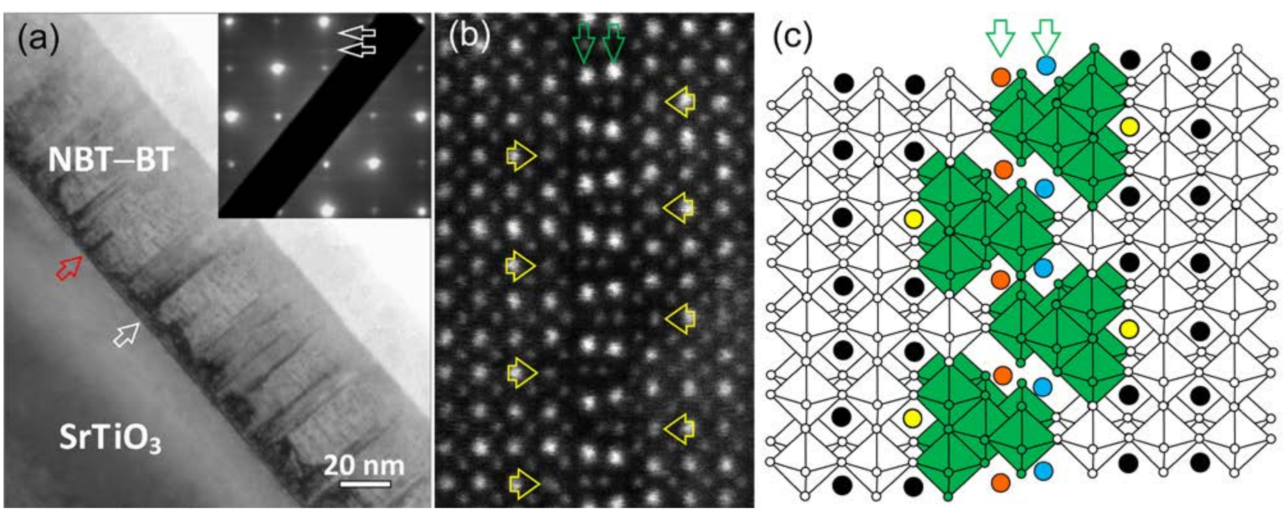

Figure 1. (a) A low-magnification BF-TEM image of 0.95NBT-0.05BT thin films on $SrTiO_3$ (001) substrates. The inset is the corresponding SAED pattern. (b) An atomic-resolution HAADF image of a typical planar defect projected along the [010] zone axis of 0.95NBT-0.05BT. Vertical green arrows denote the Bi-rich double planes, and horizontal yellow arrows mark the Bi-deficient Na/Bi(Ba) columns. (c) The model of the planar defects. The corner-sharing oxygen octahedra in the normal regions are displayed in white, while those with edge sharing ones in the fault area in green. Bi-atom columns in the defects with different height levels are in red and blue, respectively. The Na/Bi (Ba) columns in the normal regions are presented in black, and the Bi-deficient columns in yellow.