## Interfacial Structure in Multiferroic BiFeO<sub>3</sub> Thin Films

H.J. Chang\*, A.Y. Borisevich\*, N. Balke\*\*, S.V. Kalinin\*\*, R. Ramesh\*\*\*, M. Huijben\*\*\* and S.J. Pennycook\*

- \* Materials Science and Technology Division, Oak Ridge National Laboratory, PO Box 2008, Oak Ridge, TN 37831
- \*\* Center for Nanophase Materials Science, Oak Ridge National Laboratory, PO Box 2008, Oak Ridge, TN 37831
- \*\*\* Department of Physics, University of California, Berkeley, CA 94720
- \*\*\*\* Science & Technology, University of Twente, Enschede, Netherlands

The physical properties of epitaxial complex oxide ultrathin films are significantly affected or even dominated by the interface behavior. Strain distribution or chemical inhomogeneities thus need to be investigated at the atomic scale to accurately predict macroscopic and mesoscopic electrical and magnetic properties. Aberration-corrected Z-contrast scanning transmission electron microscopy (STEM) combined with electron energy-loss spectroscopy (EELS), which allows sub-Å probe analysis on the interface structure and chemistry, is a powerful tool for interface studies [1]. In this work, we will address the characterization of the interfaces at the atomic level in BiFeO<sub>3</sub> (BFO) thin film with a distorted perovskite structure.

BFO is a multiferroic material, i.e. it can show simultaneous ferroelectric and magnetic ordering. The possibility of controlling the coupling of these properties makes this material attractive from both fundamental and applied point of view. BFO is a room-temperature G-type antiferromagnet ( $T_N \sim 650~\text{K}$ ) and a ferroelectric ( $T_C \sim 1,103~\text{K}$ ) with a large spontaneous ferroelectric polarization ( $\sim 90~\text{µC/cm}^{-2}$ ) along the pseudocubic <111> directions [2, 3]. The crystal structure of BFO has been reported as a rhombohedrally distorted perovskite lattice, with a=b=3.935 Å and c/a=1.016 [4]. The strain and electric fields are accommodated through the formation of complex domain structures with 71°, 109 ° and 180 ° domain walls

Epitaxial BFO films (~50 nm thick) were grown using molecular-beam epitaxy on single-crystal SrTiO<sub>3</sub> substrates. A thin 5 nm layer of epitaxial (La,Sr)MnO<sub>3</sub> (LSMO) was used as a bottom electrode for electrical-contact purposes and heteroepitaxial growth. STEM images and EEL spectra were collected using a VG Microscopes HB603U operated at 300 kV and a VG Microscopes HB501UX operated at 100 kV, both equipped with Nion aberration correctors and Gatan Enfina® spectrometers.

Z-contrast images (Fig. 1(a)) taken along the [110] direction show that ~4 BFO atomic layers nearest to the interface with LSMO have uneven atomic distances along the c-direction. The extended dark contrast at the interface in the simultaneously acquired BF image (Fig. 1(b)) may also indicate that the layer is highly strained. Z-contrast images obtained along [010] direction confirm the presence of dislocations at both BFO/LSMO and LSMO/STO interfaces. Domain walls were also detected in the film (Fig.2).

EEL spectra of the interfaces were studied in both core-loss and low-loss regions. Changes in the oxygen fine structure and changes in the dielectric function in the vicinity of the interface will be discussed, as well as dependence of the observed phenomena on film thickness. [5]

## References

[1] M. Varela et al., Annu. Rev. Mat. Res., 35 (2005) 539.

- [2] F. Kubel, H. Schmid, Acta Crystallogr. B 46 (1990) 698.
- [3] D. Lebeugle et al. Appl. Phys. Lett. 91, (2007) 022907.
- [4] J. Wang et al. Science 1719 (2003) 299.
- [5] Research sponsored by the Office of Basic Energy Sciences, Division of Materials Sciences and Engineering, by Laboratory Directed Research and Development (LDRD) funding from Oak Ridge National Laboratory, and by appointment (H.J.C.) to the ORNL Postdoctoral Research Program administered jointly by ORNL and ORISE.

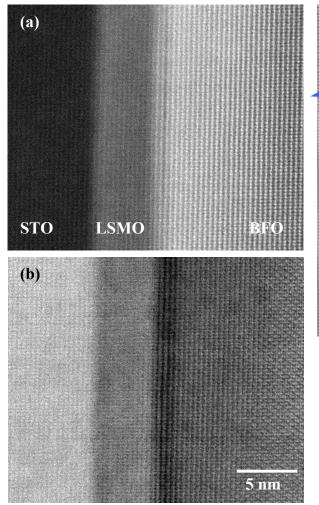


FIG. 1. (a) Z-contrast image and (b) bright field image of SrTiO<sub>3</sub>/(La,Sr)MnO<sub>3</sub>/BiFeO<sub>3</sub> obtained simultaneously along the [110] direction.

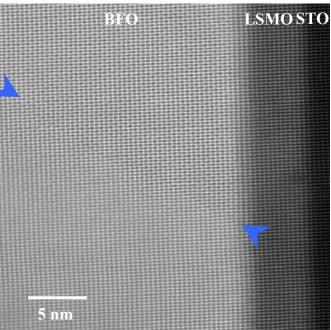


FIG. 2. Z-contrast image of SrTiO<sub>3</sub>/(La,Sr)MnO<sub>3</sub>/BiFeO<sub>3</sub> along the [110] direction, showing a domain wall between the blue arrows.