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We report on the growth of epitaxial bilayers of the La\textsubscript{2/3}Sr\textsubscript{1/3}MnO\textsubscript{3} (LSMO) half-metallic ferromagnet and the BiFeO\textsubscript{3} (BFO) multiferroic, on SrTiO\textsubscript{3}(001) by pulsed laser deposition. The growth mode of both layers is two dimensional, which results in unit-cell smooth surfaces. We show that both materials keep their properties inside the heterostructures, i.e., the LSMO layer (11 nm thick) is ferromagnetic with a Curie temperature of \(\sim 330\) K, while the BFO films shows ferroelectricity down to very low thicknesses (5 nm). Conductive-tip atomic force microscope mappings of BFO/LSMO bilayers for different BFO thicknesses reveal a high and homogeneous resistive state for the BFO film that can thus be used as a ferroelectric tunnel barrier in tunnel junctions based on a half-metal. © 2006 American Institute of Physics. [DOI: 10.1063/1.2170432]
Reflections of the BFO film were collected at an acceleration voltage of 25 kV. The sample was finally cooled to room temperature in BFO film/STO/LSMO/LSMO//STO bilayers with LSMO=11 nm and BFO=5 nm and 70 nm. S, L, and B label peaks of STO, LSMO, and BFO, respectively. (d) X-ray diffraction spectra of BFO/LSMO/STO bilayers with t_{LSMO}=11 nm and t_{BFO}=5 nm and 70 nm. S, L, and B label peaks of STO, LSMO, and BFO, respectively. (e) RSM of the (103) reflections of the BFO (70 nm)/LSMO/STO bilayer; r.l.u. is for reciprocal space units.

$P_{dep}T_{dep}$ phase stability diagram for BFO films and found that optimal conditions are around $T_{dep}=580 \degree C$ and $P_{dep}=6 \times 10^{-3}$ mbar. In the present samples, the LSMO layer (11 nm thick for all samples) was grown first, and the BFO film (t_{BFO}=1–70 nm) afterward. To limit a possible deoxygenation of the manganite, after the growth of the LSMO film, the pressure was kept at the LSMO deposition pressure while decreasing $T_{dep}$ to 580 $\degree C$. Then, the pressure was rapidly decreased to $6 \times 10^{-3}$ mbar, to grow the BFO film. The sample was finally cooled to room temperature in 300 mbar of oxygen.

Reflection high-energy electron diffraction (RHEED) patterns were collected (at an acceleration voltage of 25 kV) before growth, and after depositing each layer. Images for the [100] direction are shown in Figs. 1(a)–1(c), and indicate a two-dimensional growth for the LSMO and the BFO layer (up to at least t_{BFO}=35 nm). An azimuthal analysis revealed an in-plane epitaxy for both layers.

X-ray diffraction θ-2θ spectra (in the 15°–115° 2θ range) were collected for all samples and only showed peaks corresponding to (00l) reflections (pseudo-cubic indexation) of STO, LSMO, and BFO. Figure 1(d) shows the spectra for t_{BFO}=5 nm and 70 nm. From the angular position of the (003) reflections of BFO, we calculated the out-of-plane parameter $c_{BFO}=4.10$ Å that was found to vary only slightly with thickness. A reciprocal space map (RSM) of the (103) reflections [see Fig. 1(e)] for a 70 nm film shows that both the LSMO and BFO layers are very heavily strained on the STO substrate. Note that no splitting of the BFO (103) peak is detected, suggesting a tetragonal rather than monoclinic or rhombohedral symmetry for the BFO layer, in contrast to the results of Xu et al. or Qi et al., respectively.

Figure 2(a) shows a magnetization hysteresis cycle of a BFO(5 nm)/LSMO bilayer measured at 10 K with the field applied in-plane along [100] after zero-field cooling, measured in a superconducting quantum interference device (SQUID). The saturation magnetization ($M_s$) for t_{BFO}=5 nm is about 580 emu cm$^{-3}$, close to the magnetization of bulk LSMO (590 emu cm$^{-3}$). Even for larger BFO thickness values, the contribution of the BFO layer to the magnetization was not visible, as expected from the very small magnetization values obtained for optimized BFO single films grown in the same conditions. We measured the evolution of the magnetization with the temperature in order to check the quality of the LSMO layer [see Fig. 2(b)]. The Curie temperature ($T_C$) is 330 K, somewhat smaller than the bulk $T_C=370$ K, but in good agreement with the $T_C$ of thin single films of similar thickness.

The BFO/LSMO bilayers were imaged with a conductive-tip atomic force microscope (CTAFM). In these type of experiments, the morphology of the sample surface and the resistance between the bottom electrode and the tip are measured simultaneously, and coupled height-resistance maps are recorded. Examples of such maps 3 x 3 μm$^2$ are shown in Figs. 3(a) and 3(b) for a BFO(5 nm)/LSMO bilayer. The left image reveals a very flat surface (in agreement with the two-dimensional growth mode inferred from the RHEED patterns) with terraces separated by ~4 Å high steps, i.e., a perovskite unit cell. The corresponding resistance map [Fig. 3(b)] shows a high resistance level (average value ~1 GΩ), with a remarkable homogeneity. Similar coupled maps were collected for samples with different BFO thicknesses. Identical unit-cell steps were observed up to t_{BFO}=20 nm. Resistance maps could be collected for t_{BFO}=1, 2, and 5 nm samples, without saturating the capability of the CTAFM electronics. The average resistance of the maps is plotted as a function of the BFO thickness in Fig. 3(c). The data at t_{BFO}=0 correspond to a single LSMO film. An exponential increase of the resistance with t_{BFO} is obtained, which indicates that transport occurs through the BFO film by tunneling. This observation, together with the flatness and the homogeneity of the BFO on the LSMO buffer, qualifies BFO very thin films as potential barriers in tunnel junctions.

Piezoelectric atomic force microscopy (PFM) was used to probe the ferroelectricity of the BFO films in BFO/LSMO bilayers, for several values of t_{BFO}. An alternatively positive and negative voltage was applied between the conductive tip of the AFM and the bottom electrode (LSMO) to pole the BFO layer into “up” and “down” stripes. PFM was then used to detect the domain structure. Figure 4(a) shows the topography, and Figs. 4(b) and 4(c) the PFM images of the 5 nm film after writing. We observe a clear contrast in Fig. 4(b) that reveals the presence of up and down ferroelectric domains in this film. Note that this pattern is not observed in FIG. 2. (a) Magnetic hysteresis loops of the BFO/LSMO/STO bilayer with $t_{BFO}=5$ nm measured by SQUID at 10 K. (b) Temperature dependence of the magnetization in a field of 1kOe, normalized to the magnetization at 10 K ($M_{10 K}$) for the same sample.
films give virtually no signal, as expected for a weak ferromagnet. Through a combined CTFM and PFM study on these BFO/LSMO heterostructures, we have shown that BFO layers as thin as 5 nm are ferroelectric and can be used as tunnel barriers. This opens the way for the realization of several types of devices, such as ferroelectric tunnel junctions\textsuperscript{15} or magnetic tunnel junctions\textsuperscript{4} with ferroelectric tunnel barriers. In this latter type of structure, a control of the ferroelectric polarization by an external magnetic field can be envisaged, via the magnetoelectric effect.

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