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Combining half-metals and multiferroics into epitaxial heterostructures for spintronics

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We report on the growth of epitaxial bilayers of the La_{2/3}Sr_{1/3}MnO_3 (LSMO) half-metallic ferromagnet and the BiFeO_3 (BFO) multiferroic, on SrTiO_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 \( T_C = 330 \) 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]

To a great extent, today’s research in spintronics focuses on the development of materials and devices concepts. In many cases, the former are requisites to the latter. Recently, several families of materials have been developed in this sense, a typical example being that of diluted magnetic semiconductors. As spintronics effects rely on the spin polarization of the electrical current, materials with large, ideally total, spin-polarization have also been extensively investigated. With these so-called half-metals, a considerable increase in the tunnel magnetoresistance of magnetic tunnel junctions has indeed been achieved, at least at low temperatures.

Another emerging family of magnetic materials are multiferroics. In many compounds, several ferroic orders (e.g., magnetic and electric) coexist, with some coupling between them (the magnetoelectric effect). Most are ferroelectric and antiferromagnetic, a notable exception being BiMnO_3 that is ferromagnetic. A prototypical multiferroic that has attracted a lot of attention lately is BiFeO_3 (BFO). It is a ferroelectric and weakly ferromagnetic rhombohedral perovskite with order temperatures far above room temperature \( T_C = 1043 \) K. BFO thus crystallizes in the same structure as several known half-metallic ferromagnets (such as La_{2/3}Sr_{1/3}MnO_3, La_{2/3}Ca_{1/3}MnO_3, or Sr_{2}FeMoO_6), which makes it possible to combine it with these materials in multifunctional epitaxial heterostructures. Several promising types of devices can be imagined from this combination, as discussed, for instance, by Binek et al. In particular, one can think of using very thin layers of BiFeO_3 as multiferroic tunnel barriers. If ferroelectric, these layers should have the same functionalities as those of recently developed and modeled ferroelectric tunnel barriers, combined with a magnetic ordering and a possible magnetoelectric coupling.

In this letter, we describe the growth and properties of bilayers of the La_{2/3}Sr_{1/3}MnO_3 (LSMO) half-metal combined with BFO and epitaxially grown onto SrTiO_3(001) substrates by pulsed laser deposition. We study the morphological, structural, electrical, and magnetic properties of BFO/LSMO bilayers. We show that the magnetic properties of LSMO are preserved and that the BFO layers are insulating and ferroelectric, down to thicknesses of 5 nm. BFO ultrathin layers thus fulfill some important criteria for being used as ferroelectric tunnel barriers.

The samples used in this study have been grown by pulsed laser deposition using a Nd:yttrium–aluminium–garnet (YAG) laser, at a frequency of 2.5 Hz. The LSMO target was stoichiometric while for BFO, a target with nominal composition Bi_{1.15}FeO_3 was used, in order to compensate for the high volatility of Bi.

The optimal growth conditions for LSMO were previously determined to be a deposition temperature \( T_{dep} = 720 \) °C and an oxygen pressure of \( P_{dep} = 0.41 \) mbar. with a postannealing at 300 mbar of O_2. We recently determined the...
The sample was finally cooled to room temperature in 062502-2 Bea was rapidly decreased to 6
and B label peaks of STO, LSMO, and BFO, respectively. Compared to 10^{-6} mbar for applied in-plane along
deoxygenation of the manganite, after the growth of the
result of Xu the LSMO and BFO layers are very heavily strained on the
parameter c_{BFO}=20 nm. A reciprocal space map
FIG. 1. RHEED patterns in the [100] direction of the sample with t_{BFO} =20 nm (a) of the STO substrate before deposition, (b) after deposition of the LSMO layer, and (c) after deposition of the BFO. The pattern is more diffuse in (b) because the pressure of measurement was 6 \times 10^{-3} mbar compared to 10^{-6} mbar for (a) and (c). (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 °C and P_{dep}=6 \times 10^{-3} mbar.\textsuperscript{19} 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 °C. Then, the pressure was rapidly decreased to 6 \times 10^{-3} mbar, to grow the BFO layer. 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.\textsuperscript{20} or Qi et al.\textsuperscript{21} 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}).\textsuperscript{22} 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.\textsuperscript{19} 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, somehow smaller than the bulk T_{C}=370 K,\textsuperscript{22} but in good agreement with the T_{C} of thin single films\textsuperscript{23} 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.\textsuperscript{24} Examples of such maps 3 \times 3 μm² 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 the 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 buffers, 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
films give virtually no signal, as expected for a weak ferromagnet. Through a combined CTAFM 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 with or without magnetic tunnel junctions 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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