Study of exchange bias effect in bilayers based on $La_{2/3}Ca_{1/3}MnO_3$ and $BiFeO_3$

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Abstract.
$BiFeO_3$ (BFO) is an interesting material due to the coexistence of magnetic and ferroelectric order at room temperature. Moreover, mixed manganese perovskite oxides like ferromagnetic $La_{2/3}Ca_{1/3}MnO_3$ (LCMO-F) present remarkable properties, like colossal magnetoresistance and non-volatile memory. Bilayers of BFO/LCMO-F and LCMO-F/BFO were grown on (100) $SrTiO_3$ (STO) substrates. LCMO-F films were deposited by DC sputtering and BFO by RF magnetron sputtering technique at high-oxygen pressures. Samples were structurally characterized by x-ray diffractometry. Electrical properties were evaluated by resistivity measurements as function of temperature, showing how the transition temperature of manganite is affected by the presence of the BFO layer. Magnetic characterizations were carried out by magnetization measurements as function of temperature and magnetic field. BFO/LCMO-F/STO bilayer showed a higher Curie temperature than LCMO-F layers grown by similar parameters. Magnetic hysteresis curves for LCMO-F/BFO/STO bilayers gave us indication of the existence of exchange bias effect with a Hex of 200 Oe after field cooling at 1 T; we can also conclude that this bilayer configuration offers posibilities to obtain bilayers exhibiting this polarization effect.

1. Introduction

Multiferroics materials are those which exhibit multiple functional properties in which two or all three of the properties ferroelectricity, ferromagnetism and ferroelasticity occur simultaneously in the same phase [1]. $BiFeO_3$ (BFO) is a multiferroic material which structure and magnetic ordering has been study by having two types of long range order: antiferromagnetic ordering below $T_N = 370°C$ and ferroelectric ordering, with a high Curie temperature about $820 - 850°C$. $BiFe0_3$ is known to have a rhombohedrally distorted perovskite structure owing to R_{3C} symmetry [2]. Moreover, mixed manganese perovskite oxides present remarkable properties, like colossal magnetoresistance and non-volatile memory. $La_{2/3}Ca_{1/3}MnO_3$ (LCMO-F) is widely studied due to its magnetic and transport properties; has a ferromagnetic phase and perfect cubic perovskite structure and exhibits paramagnetic-ferromagnetic transition simultaneously with a metal-insulator transition around 270K for thin films systems [3].

Previously, we have grown multiferroic BFO [4] and ferromagnetic LCMO-F thin films with high quality [5], for that reason we can explore, in this work, the behavior of ferromagnetic LCMO and antiferromagnetic BFO bilayers to seek Exchange Bias (EB). This phenomenon manifests itself by a shift in the hysteresis loop of a ferromagnetic in contact with an antiferromagnetic and arises from the exchange coupling at the FM/AF interface [6]. In this report, we shall give an indication of the possible existence of Exchange Bias effect in LCMO-F/BFO/STO bilayers.

2. Experimental Details

We have grown BFO/LCMO-F and LCMO-F/BFO bilayers on (100) $SrTiO_3$ (STO) substrates by sputtering technique at oxygen atmosphere (O_2 99.999%). The LCMO-F layer was deposited using DC sputtering with 830°C susbtrate temperature, oxygen pressure at 3.5 mBar, and a estimated deposition rate of 80 nm/h, whereas BFO layer was grown using RF magnetron sputtering with 600°C as subtrate temperature, oxygen pressure at $4.0\text{x}10^{-1}$ mBar and 72 nm/h deposition rate [4][7]. Samples were structurally characterized by x-ray diffraction (XRD), electrical properties were evaluated by resistivity measurements as function of temperature by DC four points resistance, Van der Pauw technique, in a physical properties measurements sytems (PPMS) quantum design. Magnetic characterizations were carried out by magnetization measurements as function of temperature (10 to 300K) and Field colling (FC) isothermal M(H) loops for temperatures of 10 and 60K using the vibrating sample magnetometer (VSM) probe of the PPMS systemm and SQUID magnetometer (Superconducting Quantum Interference Device) with a magnetic field applied in the plane of the film. We used LCMO-F/BFO/STO and BFO/LCMO-F/STO notation for the two possible configurations of our bilayers.

3. Results

Figure 1, shows the X-ray diffraction pattern for both configurations of our bilayers. LCMO-F/BFO/STO (a) shows characterisitic peaks like [104], [110] of the BFO layer, whereas Bragg peaks from LCMO-F layer are not visible. However, this sample show magnetic properties as you can see later. BFO/LCMO-F/STO bilayer (b), presents a highly oriented growth of the ferromagnetic layer of manganite around (200) reflection of the substrate, respect to BFO layer peak we observed [012] anchored to the [100] peak of the substrate. This sample show us a textured growth regarding to the (100) substrate direction. Bragg peaks noticed with asterisk corresponds to other phases of bismuth oxide present on BFO films.

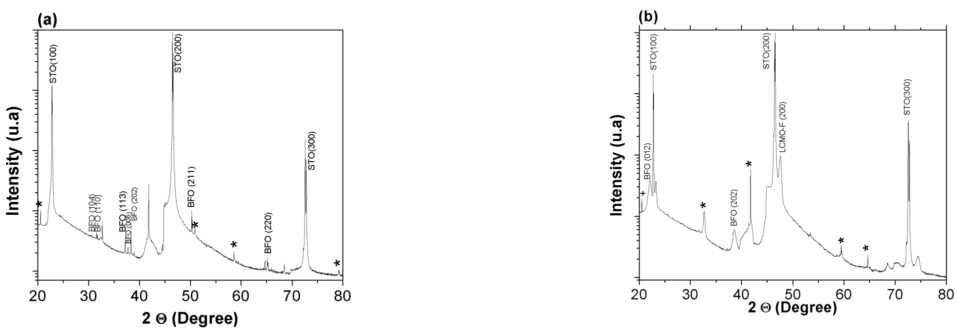

Figure 1. XRD patterns for LCMO-F/BFO/STO (a) and BFO/LCMO-F/STO (b) configurations.

Figure 2, displays the temperature dependence of the resistance curves, in the range of 10 - 300 K for 100 nm LCMO-F film (a), LCMO-F/BFO/STO (b) and BFO/LCMO-F/STO (c). LCMO-F/BFO/STO (b) shows typical ferromagnetic metal isolator transition characteristic of the LCMO system, with a transition temperature, T_{IM} of 225±25K; this transition temperature is lower than that for a 100 nm LCMO-F film (a) grown under identical parameters. Moreover, BFO/LCMO-F/STO sample (c) contact electrodes are on the BFO insulator layer and the resistance curve exhibits predominant behavior of insulating phase at low temperatures, even then it is possible to observe a decrease of the resistivity for temperatures below 150 K arising from the metal behavior of the LCMO-F layer. Our resistance measurements are carried out in the plane of the layers. Thus, we have contribution to the resistance from the two layers and also from the interface, but we cannot discriminate each.

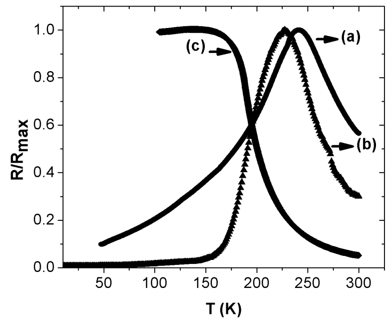

Figure 2. Resistance (normalized) as function of temperature forLCMO-F (a), LCMO-F/BFO/STO (b) and BFO/LCMO-F/STO (c).

Figure 3, shows the zero filed cooling (ZFC) thermal demagnetization behavior for LCMO-F/BFO/STO sample (b), and BFO/LCMO-F/STO sample (c). The behavior are dramatically different. BFO/LCMO-F/STO bilayer, where the LCMO-F layer grew directly on substrate, shows the typical demagnetization behavior of the LCMO-F system (a), whereas the LCMO-F/BFO/STO bilayer an atypical ferromagnetic response, with a maximun in M a very low temperature (50 K) and for temperatures below the magnetization drecreases dramatically; these results indicate that the magnetic property of the LCMO-F film is strongly dependent of its crystalline structure and morphology. LCMO-F grew very well on STO substrates; but after growing the BFO layer on top of LCMO-F, its magnetic properties decrease. We could determine the Curie temperature of 247.1 ± 0.1 K for the BFO/LCMO-F/STO bilayer using a least squares fit with a gaussian distribution of ferromagnetic regions [7].

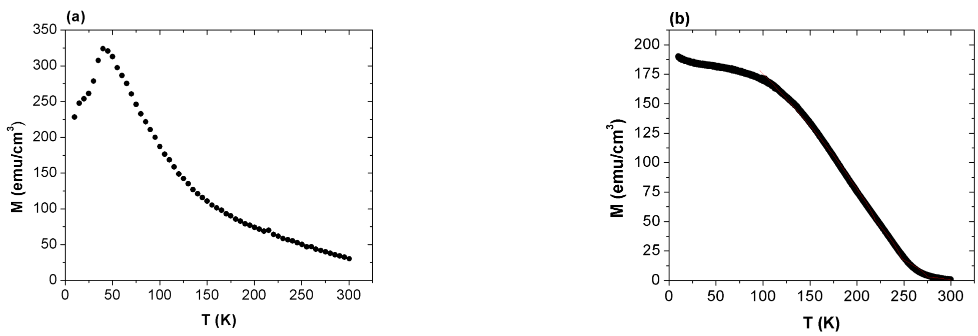

Figure 3. Magnetization as function of temperature, ZFC, for LCMO-F/BFO/STO sample (a), and BFO/LCMO-F/STO bilayer (b).

Figure 4, shows isothermal magnetic loops at 10 K (blue) and 60 K (red) for LCMO-F/BFO/STO sample (a) and BFO/LCMO-F/STO bilayers (b). Both samples are ferromagnetic at the measured temperatures. However, BFO/LCMO-F/STO bilayer is magnetically very poor,

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and we can not measured a shift in the hysteresis loop after field cooling, as we can see in the inset. On the other hand, LCMO-F/BFO/STO bilayer show clearly ferromagnetic behavior exhibiting a weak exchange bias effect, see inset. Coercive field, width of the hysteresis curve, exchange bias magnitud increase with the decreasing of the temperature as expected for the LCMO system.

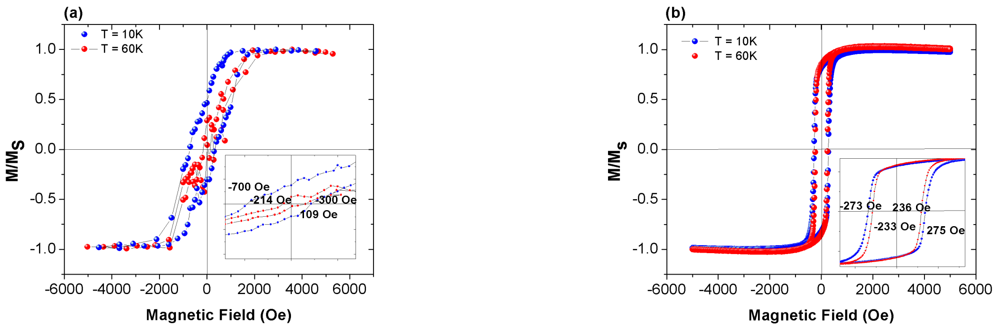

Figure 4. Magnetic hysteresis loops at 10 and 60K for LCMO-F/BFO/STO (a) and BFO/LCMO-F/STO (b).

The magnitude of the shift, denoted as exchange field (H_{EX}) was measured over hysteresis isotherms according to the average between the two coercive fields and we found the most significant result for the LCMO-F/BFO/STO bilayer at 10K obtaining Hex = 200 Oe.

4. Conclusions

We have suscefully grown BFO/LCMO-F and LCMO-F/BFO bilayers on (100) $SrTiO_3$ substrates, where LCMO-F/BFO/STO system are exhibiting exchange bias effect with a Hex of 200 Oe after field cooling at 1 T. We can also conclude that from magnetic properties point of view, this bilayer configuration offers posibilities to obtain bilayers exhibiting this polarization effect.

Acknowledgments

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References

[1] H Schmid 1994 *Ferroelectrics.* **162** 317
[2] I Sosnowskat, T Peterlin-Neumaier and E Steichele 1982 *J. Phys. C: Solid State Phys.* **15** 4835-4846
[3] P Vivas, E Baca, G Campillo, B Maiorov, N Haberkorn and J Guimpel 2002 *Revista Colombiana de Física.* **34** 625-629
[4] J Zapata, J M Caicedo, W Lopera, M E Gomez, F Cuellar and P Prieto 2008 *Colombian Journal of Physics.* **40** 77-80
[5] M E Gómez, G Campillo, J G Ramírez, P Prieto, A Hoffmann, J Guimpel, N Haberkorn, A Condó and F Lovey 2006 *IEEE Transactions on Magnetics.* **42** 10
[6] J Nogués and I K Schuller 1999 *J. Magn. Magn. Mater.* **192** 203-232
[7] G Campillo, A Berger, J Osorio, J E Pearson, S D Bader, E Baca and P Prieto 2001 *Journal of Magnetism and Magnetic Materials.* **237** 6168