Proceedings of the European Conference "Physics of Magnetism" (PM'08), Poznań 2008

# Structure and Magnetic Characterization of BiFeO<sub>3</sub>/YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> Bilayers

# K. WERNER-MALENTO<sup>a</sup>, A. TSAROU<sup>a</sup>, P. DLUZEWSKI<sup>a</sup>, W. PASZKOWICZ<sup>a</sup>, R. MINIKAYEV<sup>a</sup>,

M. Sawicki<sup>a</sup>, Kees van der Beek<sup>b</sup>, M. Konczykowski<sup>b</sup> and P. Przyslupski<sup>a</sup>

<sup>a</sup>Institute of Physics, Polish Academy of Sciences

al. Lotników 32/46, 02-668 Warsaw, Poland

<sup>b</sup>Laboratoire des Solides Irradies, Ecole Polytechnique, Palaiesau, France

Bilayered epitaxial BiFeO<sub>3</sub>/YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films were fabricated on (100) [(LaAlO<sub>3</sub>)<sub>0.3</sub>(Sr<sub>2</sub>TaAlO<sub>6</sub>)<sub>0.7</sub>] substrates by sputtering method. For structural comparison the bilayered BiFeO<sub>3</sub>/La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> films were also deposited on (100) SrTiO<sub>3</sub> substrates. A weak ferromagnetic moment is observed in BiFeO<sub>3</sub>/YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films. The mechanism responsible for weak ferromagnetic moment arises presumably from the epitaxy strain induced canted antiferromagnetic structure.

PACS numbers: 74.25.Ha, 74.78.Fk, 75.70.Cn, 77.84.Bw

#### 1. Introduction

Multiferroic (MF) materials demonstrate both ferroelectric and ferro or antiferromagnetic order. The coupling between the order parameters is interesting from the point of view of fundamental physics as well as can lead to applications in spintronics [1, 2]. In such compounds due to occurrence of ferroelectric and magnetic states the change of magnetization can be induced by electrical field and vice versa which is known as magnetoelectric effect (ME). The integration of dissimilar materials such as superconductor and MF can create a new type composite which could be interesting in development of the new type functionality of such heterostructures. Bulk  $BiFeO_3$  (BFO) exhibits a rhombohedrally distorted perovskite structure with space group R3c and aniferromagnetic ordering of  $Fe^{3+}$  ions [3]. In the case of (001) oriented thin films deposited on substrates with cubic symmetry the epitaxial strain will induce lattice distortion and also can change the crystallographic symmetry. In epitaxial films due to relaxation of strain by formation of structural defects with increasing thickness the structure of the films changes as a function of their thickness [4, 5]. In this paper we report our preliminary studies on the preparation and characterization of BFO films deposited on the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO) films. Our motivation to prepare high quality BFO/YBCO bilayers is a search for a possibility of ME effect which could occur in BFO/YBCO heterostructure induced by the electromagnetic interaction of the screening currents when cooling the BFO/YBCO bilayer below its superconducting transition temperature with magnetic moment of the BFO layer.

# 2. Experimental

BFO films were deposited by rf sputtering with rf power of 120 W from polycrystalline BFO target. The BFO target with nominal composition plus excess of 10% of Bi was prepared by sintering constituent oxide mixture  $Bi_2O_3$  and  $Fe_2O_3$  at 830°C. The deposition was carried out on (100)  $[(LaAlO_3)_{0.3}(Sr_2TaAlO_6)_{0.7}]$  (LSAT, a = 0.3868 nm) substrates in oxygen pressure of 1.5 mbar at 720°C. Prior to deposition of BFO film the YBCO buffer layer of 40 nm in thickness was first deposited at substrate at 760°C by high pressure dc sputtering [6] in 3 mbar of oxygen pressure. The epitaxial growth of BFO/YBCO or BFO/La<sub>0.67</sub>Sr<sub>0.33</sub>MnO<sub>3</sub> (LSMO) bilayers was characterized by X-ray  $\theta - 2\theta$  diffractometer using Cu  $K_{\alpha}$  radiation. Sharp interfaces and uniformly continuous layers in bilayer structures was confirmed by transmission electron microscopy (TEM). The magnetization of bilayers was measured by SQUID magnetometer.

### 3. Results and discussion

In order to confirm that the growth of the BFO film is epitaxial first we have prepared the BFO(40 nm)/LSMO(20 nm) bilayer deposited on (100)  $SrTiO_3$  substrate. Figure 1a shows a typical X-ray diffraction (XRD) linear scan of the BFO/LSMO bilayer, where only (001) reflections from BFO and LSMO were observed indicating the epitaxial *c*-axis growth of the BFO and LSMO films (along the film normal direction). The epitaxial growth of BFO film on LSMO films was observed using pulsed laser deposition [7, 8]. Figure 1b shows X-ray diffraction scan of

BFO(55 nm)/YBCO(40 nm) bilayer. The X-ray diffraction scan indicates that film is free from impurity phases and highly oriented along c-axis with only (001) peaks. It is found that epitaxial growth of the BFO film on YBCO film and the diffraction patterns originating from BFO overlap with those from YBCO. The epitaxial growth is also confirmed by the cross-sectional TEM studies as shown in Fig. 2. TEM image shows that the interface between BFO film and YBCO film is atomically sharp.



Fig. 1. (a) XRD patterns of the BFO/LSMO bilayer deposited at (100) SrTiO<sub>3</sub> substrate, and (b) BFO/YBCO bilayer deposited on LSAT substrate.



Fig. 2. TEM cross-section of BFO(55 nm)/ YBCO(40 nm) bilayer deposited at (100) LSAT substrate.

The out-of-plane magnetic moment M(T) measurements of BFO(55 nm)/YBCO(40 nm) bilayer indicate that the onset of diamagnetic transition  $T_{\rm co}$  begins at 62 K as presented in Fig. 3. Relatively high superconducting transition temperature indicates that the BFO/YBCO bilayer are fully oxygenated and the concentration of the oxygen vacancies in both systems is low.



Fig. 3. Zero field cooling (ZFC), magnetic moment vs. temperature of the BFO/YBCO bilayer.

Figure 4 shows the magnetization hysteresis loops, M(H), measured for the BFO(55 nm)/YBCO(40 nm) bilayer recorded above and below superconducting transition both in parallel and perpendicular magnetic field to sample surface. The data indicate superconducting response at low temperatures. Above the superconducting transition a weak ferromagnetic response is seen. The estimated magnetic moment of 12 emu/cc was observed. The mechanism underlying the weak ferromagnetic behavior of BFO films is complicated. Theoretical model [9] indicates that oxygen vacancies associated with  $Fe^{2+}$ ions could enhance the effective magnetization. Also experimental observation [10] support this supposition. Experimental observation [11] indicate also an important role of strain imposed on BFO films. An enhancement of magnetic moment was observed for BFO thin films with thickness lower than 80 nm. The BFO/YBCO bilayer shows (Fig. 4b,d) M-H curves characteristic of antiferromagnetic order accompanied by weak ferromagnetism with well-defined saturation, remanent magnetization and coercive field.



Fig. 4. Magnetization hysteresis loops of the BFO(55 nm)/YBCO(40 nm) bilayer recorded in magnetic field perpendicular (a,b) and parallel (c,d) to the sample.

In summary, the bilayered epitaxial BFO/YBCO films were successfully prepared on (100) LSAT substrates by sputtering method. These films demonstrate a superconducting state and weakened ferromagnetism, which is attributed to strain induced antiferromagnetic structure.

### Acknowledgments

This work was supported by the Ministry of Science and Higher Education under research project for the years 2006–2009 (MNiSW-1 P03B 122 30, N N202 0708 33).

# References

- W. Erenstein, N. Mathur, J.F. Scott, *Nature Mater.* 442, 759 (2006).
- [2] R. Ramesh, N.A. Spaldin, Nature 6, 21 (2006).
- [3] D. Lebeugle, D. Colson, A. Forget, M. Viret, Appl. Phys. Lett. 91, 022907 (2007).
- [4] J. Wang, J.B. Neaton, H. Zheng, V. Ngarajan, S.B. Ogle, B. Liu, D. Veihland, V. Vaityhyanathan, D.G. Schlom, U.V. Waghmare, N.A. Spaldin, K.M. Rabe, M. Wuttig, R. Ramesh, *Science* 299, 1719 (2003).

- [5] Y.H. Chu, T. Zhao, M.P. Cruz, Q. Zhan, P.L. Yang, L.M. Martin, M. Huijben, C.H. Yang, F. Zavaliche, H. Zheng, R. Ramesh, *Appl. Phys. Lett.* **90**, 252906 (2006).
- [6] P. Przysłupski, I. Komissarov, W. Paszkowicz, P. Dluzewski, R. Minikayev, M. Sawicki, *Phys. Rev.* B 69, 134428 (2004).
- [7] H. Bea, M. Bibes, M. Sirena, G. Harranz, K. Bouzehouane, E. Jacquet, S. Fusil, P. Paruch, M. Dawber, J.-P. Contour, A. Barthelemy, *Appl. Phys. Lett.* 88, 062502 (2006).
- [8] Guo-Zhen Liu, Can Wang, Chun-Chang Wang, Jie Qiu, Meng He, Jie Xing, Kui-Juan Jin, Hui-Bin Lu, Guo-Zhen Yang, Appl. Phys. Lett. 92, 122903 (2008).
- C. Ederer, N.A. Spaldin, *Phys. Rev. B* **71**, 224105 (2008).
- [10] M.C. Li, J.L. MacManus-Driscoll, Appl. Phys. Lett. 87, 252510 (2005).
- [11] D.S. Rana, K. Takahashi, K.R. Marvani, I. Kawayama, H. Murakami, M. Tonouchi, T. Yanagida, H. Tanaka, T. Kwawai, *Phys. Rev. B* **75**, 060405 (2007).