Investigation of Dzyaloshinskii-Moriya Interaction in Rhombohedral and Tetragonal BiFeO₃/CoFe Bilayers

Koki Mukaiyama, Hiroshi Naganuma, Mikihiko Oogane and Yasuo Ando

Department of Applied Physics, Graduate School of Engineering, Tohoku University, Aoba-yama 6-6-05, Sendai 980-8579, Japan Phone: +81-022-795-7949 E-mail: kokim@mlab.apph.tohoku.ac.jp

1. Introduction

Multiferroic BiFeO₃ (BFO) is attractive material because ferroelectricity and antiferromagnetism coexist above room temperature (RT). In the BFO/ferromagnets bilayers, electric polarization switching can change the magnetization direction in ferromagnetic layer through exchange bias (EB) effect at the interface between BFO/ferromagnet bilayers. This mechanism is useful for voltage operated magnetic random access memory (V-MRMA) for non-volatile memory with low power consumption. Some studies have already demonstrated the electrical manipulation of the magnetization direction in ferromagnetic layer for BFO/ferromagnets bilayers [1-3]. In this mechanism, EB between BFO and ferromagnetic layer is very important. However, there are a few reports on EB for BFO/ferromagnets bilayers [4-6] and the understanding of EB is not enough. For earlier realization of V-MRAM, investigation of EB for more understanding is necessary. Then, we focused on that the crystal symmetry of BFO epitaxial films changed among rhombohedral(R), tetragonal(T) and monoclinic(M) structure owing to sputtering gas, power, and substrate temperature on the SrTiO₃(STO) substrates [7, 8]. EB observed ever was mostly using *R*-BFO/ferromagnets bilayers because EB operation is based on Dzyaloshinskii-Moriya (DM) interaction in rhombohedral crystal symmetry [9]. Therefore, there are no reports on EB for T- and M-BFO/forromagnets bilavers, and it has never been clarified that the relationships between the crystal symmetry of BFO and EB for BFO/ferromagnets bilayers, and the role of DM interaction in exchange bias system is not clear.

In this study, we prepared *R*- and *T*-BFO/CoFe bilayers and investigated the EB properties.

2. Experiments

The BFO films (80 nm) were deposited on STO (100) substrates with substrate temperature (T_s) 600°C by r.f. magnetron sputtering. To modify the crystal symmetry of BFO, partial oxygen gas pressure (P_{O2}) during the sputtering was changed from 0 to 36 mPa. Then, CoFe was deposited 4.0 nm on BFO with applying 150 Oe external magnetic field ($H_{dep.}$) with $T_s = RT$. Film structure was evaluated by θ -2 θ , ω , φ scan and x-ray reciprocal space mapping (XRSM). The EB properties of BFO/CoFe bilayers were evaluated by VSM and SQUID.

3. Results and Discussions

Figure 1 shows θ -2 θ profiles of BFO films with various

 P_{O2} (mPa). For the BFO films with $P_{O2} = 0$ mPa, secondary phase such as Bi₂O₃ was appeared. When P_{O2} was 18 and 36 mPa, single phase of BFO was formed. In addition, the crystallinity was found to become lower with increasing P_{O2} by ω scan, and epitaxial growth for each P_{O2} was confirmed by φ -scan measurement (not shown).

Figure 2 shows XRSM images for the $P_{O2} = 0$, 18, and 36 mPa. For $P_{O2} = 0$ mPa, the (202) diffraction was confirmed as single spot. On the other hand, two split peaks of BFO (202) were observed for $P_{O2} = 18$ and 36 mPa. According to *K. Saito et al* report [10], these (202) peak splits indicate that the BFO film with $P_{O2} = 0$ mPa has T (c/a = 1.05), and films with $P_{O2} = 18$ and 36 mPa have *R* symmetry.



Fig. 1 θ -2 θ profiles of BFO single layers.



Fig. 2 XRSM images of BFO single layers.

In above experiments, it was found that BFO epitaxial films with *T*-symmetry were successfully obtained at $P_{O2} = 0$ mPa and *R*-symmetry at $P_{O2} = 18$, 36mPa respectively. So, in the following, we fabricated *T*- and *R*-BFO/CoFe bilayers and investigated EB properties.

Figure 3 shows the magnetization curves for the BFO/CoFe bilayers measured by VSM at 300 K. Magnetic field (*H*) was applied to the same direction as H_{dep} . For the *R*-BFO/CoFe bilayers, the magnetization curves shifted by 50 Oe (b: $P_{O2} = 18$ mPa) and 20 Oe (c: $P_{O2} = 36$ mPa) to negative field. On the contrary, for the *T*-BFO/CoFe bilayer showed increase of coercive field; however, the shift of the magnetization curve was not observed (c: $P_{O2} = 0$

mPa).

Figure 4 shows the applied magnetic field angle ($\theta_{\rm H}$) dependence of magnitude of magnetization curve's shift from zero field ($H_{\rm ex}$) and coercive field ($H_{\rm c}$). We defined the applied field direction when CoFe layers were depositted as $\theta_{\rm H} = 0^{\circ}$. For *R*-BFO ($P_{\rm O2} = 18, 36$ mPa)/CoFe bilayers, unidirectional anisotropy was observed, which clearly indicates that the EB resulted from DM interaction occurs at the interface between *R*-BFO/CoFe bilayers. On the contrary, for *T*-BFO ($P_{\rm O2} = 0$ mPa)/CoFe bilayer, $\theta_{\rm H}$ dependence of $H_{\rm ex}$ is quite different from those of *R*-BFO/CoFe bilayers and it seems that unidirectional anisotropy due to EB was not observed, which was suggested that DM interaction disappeared at the interface between *T*-BFO/CoFe bilayers.

For more investigation for *T*-BFO/CoFe bilayer, the temperature dependence of H_{ex} and H_c for *T*-BFO/CoFe bilayer was measured by SQUID. In addition, CoFe directly deposited on STO substrate was also measured for comparison. In this measurement, magnetic field was applied to the angle at $\theta_{\rm H} = 0^{\circ}$. As shown in figure 5, H_c increased with decreasing temperature for *T*-BFO/CoFe bilayer in comparison with STO/CoFe, which indicates the increase of H_c is contribution from *T*-BFO. In addition, H_{ex} was observed and increased monotonically bellow 200 K.







Fig. 4 Applied magnetic field angle dependence of H_{ex} and H_c for BFO/CoFe bilayers.



Fig. 5 Temperature dependence of H_{ex} and H_c for *T*-BFO/CoFe bilayer

4. Summary

R- and *T*-BFO/CoFe bilayers were prepared, and the EB effect of them was investigated. The EB induced by DM interaction was clearly observed for the *R*-BFO/CoFe bilayers at 300K. For *T*-BFO/CoFe bilayer, on the other hand, EB was observed at only low temperature bellow 200 K and it was revealed that the DM interaction may disappear at 300K.

Acknowledgements

This work was supported by Grant-in-Aid for Exploratory Research (No. 23656025), the FIRST program, and research promotion by Tanaka foundation.

References

- [1] Y. H. Chu et al., Nature mater. 7, 478 (2008)
- [2] J. T. Heron et al., Phy. Rev. Lett. 107, 217202 (2011)
- [3] J. Allibe et al., Nano Lett. **12**, 1141 (2012)
- [4] J. Dho et al. Adv. Mater., 18. 1445 (2006)
- [5] X. QI et al. Philos. Mag. Lett., 87. 175 (2007)
- [6] H. Naganuma et al. J. Appl. Phys., 109, 07D736 (2011)
- [7] H. Liu et al., Appl. Phys. Lett. 98, 102902 (2011)
- [8] P. Ren et al., AIP Adv. 3, 012110 (2013)
- [9] S. Dong et al., Phy. Rev. B. 103, 127201 (2009)
- [10] K. Saito et al. Jpn. J. Appl. Phys. 45, 7311 (2006).