CA05

Ferroelectricity Induced by Non-collinear Magnetic Order in multiferroics $TbMnO_3$ and $CoCr_2O_4$

Y. Yamasaki[1], T. Arima[2], and Y. Tokura[1,3]

[1]Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan.
[2]Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan.
[3]Multiferroics Project, ERATO, Japan Science and Technology Agency (JST), Tsukuba 305-8562, Japan.

*Corresponding author: yamasaki@cmr.t.u-tokyo.ac.jp, Phone: +81 3 5841 6858, Fax: +81 3 5841 6858

We have investigated the ferroelectricity induced by the non-collinear magnetic ordering, in the spiral and conical magnet, and clarified the correlation between magnetic structure and the lattice modulations including the ferroelectric polarization [1]. The spin helicity, the rotatory direction of spiral spin, was observed by the polarized neutron diffraction experiment on the magnetic ferroelectric perovskite-type $TbMnO_3$. It was revealed that the direction of cooling electric field regulates the direction of ferroelectric polarization and also the spin helicity of spiral magnet. Therefore, the spin helicity can be controlled by the cooling electric field alone (Fig.1) [2]. We have also investigated the conical magnetic spinel-type $CoCr_2O_4$ that is expected to host the spontaneous polarization and magnetization, simultaneously. The ferroelectric polarization observed in the conical magnetic structure can be reversed along with the reversal of magnetization by magnetic field (Fig.2) [3]. We have concluded that this polarization reversal phenomenon is caused by the clamping of magnetic and ferroelectric domain walls.

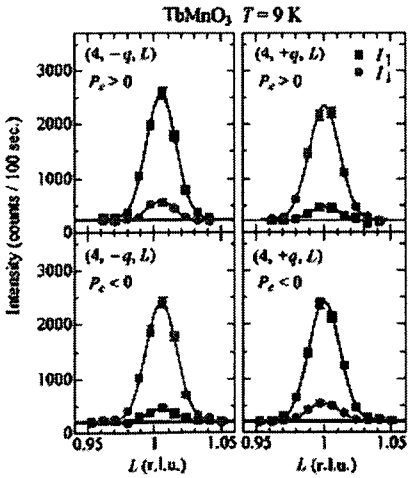

Fig. 1. Electric polarization dependence of intensity of magnetic satellites in $TbMnO_3$.

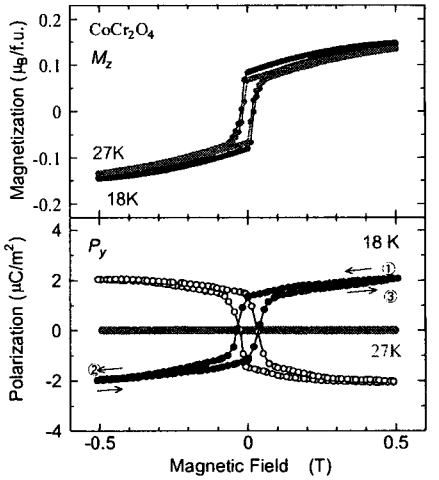

Fig. 2. Magnetic field dependence of magnetization and electric polarization in $CoCr_2O_4$.

CA06

Epitaxially Stabilized New Hexagonal $RMnO_3$ (R = Dy, Tb, and Gd) Thin Films and Their Physical Properties

T. W. Noh*[1], J.-H. Lee[1], D. Lee[1], S. Y. Jang[1], Y. Jo[2], and M.-H. Jung[2]

[1]ReCOE, School of Physics and Astronomy, Seoul National University, Seoul 151-747, Korea
[2]Quantum Material Research Team, Korea Basic Science Institute, Daejeon 305-333, Korea

*Corresponding author: e-mail: twnoh@snu.ac.kr, Phone: +82 2 880 6616, Fax: +82 2 875 1222

The lanthanide based $RMnO_3$ (R = Tb,Dy,..., and Lu) attracted a great deal of attenstion due to their richness in physics caused by the interplay between charge, spin and lattice degrees of freedom. As the Rionic size decreases, the rare earth manganites change their crystal structures from orthorhombic to hexagonal, with a phase boundary between Dy and Ho. The orthorhombic $RMnO_3$ (R = Dy, Tb, and Gd) compounds have quite low ferroelectric and antiferromagentic transition temperatures despite their strong magneto-electric couplings. On the other hand, hexagonal $RMnO_3$ (R = Ho, ..., and Lu) have high ferroelectric transition temperatures (typically, above room temperature)and their remnant polarization values are much larger than those of the orthorhomic phase materials. These facts imply that there is a possibility of controlling the multiferroic properties by modifying the structural phase of the rare-earth manganites.

Here we report the fabrication of the *h*-$RMnO_3$ (R = Dy, Tb, and Gd) thin films using the epitaxial stabilization technique.[1,2] Note that these hexagonal manganites with rather large radius ions do not exist in nature. We could grow such artificial materials using $Pt/Al_2O_3(0001)$ and YSZ(111) substrates, whose surfaces have the hexagonal nets of atomic arrangements. The hexagnoal in-plane symmetry on the substrate surfaces could ensure that the *h*-$RMnO_3$ films grow epitaxially in the hexagnoal phase by maintaing the surface energy.

From the *P-E* hysterisis loop, we measured theelectric properties. These *h*-$RMnO_3$ films showed enhanced ferroelectric properties. For example, *h*-$TbMnO_3$ thin film has a maximum polarization of 1.6 C/cm^2, which is 20 times larger than that of the orthorhombic phase, and its Curie temperature is about 60 K. In addition, we also found an antiferroelectric-like phase above 65 K.

Magnetic property studies showed that they have diverse and very intriguing magnetic phase transitions. All of the *h*-$RMnO_3$ (R = Dy, Tb, and Gd) materials showed an antiferromagnetic Néel temperature around 60 K and a spin reorientation transition around 40 K, just like natural hexagonal manganates. However, they also showed spin-glass-like behavior, which was likely to arise from the geometric frustruation of antiferromagnetically coupled Mn spins with an edge-sharing triangular lattice. In addition, at lower temperature, there exists an ferromagnetic ordering due to the exchange coupling between spins at the rare earth ions.

We also found evidences for the magnetoelectric effects, whose coefficient changes according to the magnetic phase transitions. Especially, at the spin reorientation temperature, the dielectric constant shows an anomaly, whose position varies with the applied magnetic field. All of these intriguing findings suggest the the uniqueness of *h*-$RMnO_3$ compounds in multiferroic materials and their potential applications.

REFERENCES

[1] J.-H. Lee *et al.*, Adv. Mater. **18**, 3125 (2006).
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