

# Ferromagnetism in $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$ Thin Films and the Correlation Between Ferroelectric and Ferromagnetic Domains

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Multiferroic materials, where ferromagnetic and ferroelectric orders coexist, promise various improvements over singly ordered ferroic materials for the next generation of memory, sensing, and actuation applications.  $\text{BiFeO}_3$  is the most widely studied multiferroic material that has attracted much attention due to its giant electric polarization and room temperature multiferroic properties. It has a cycloidal space-modulated spin structure with a periodicity of 62 nm which generates a parasitic electric polarization superimposed on the G-type antiferromagnetic structure. The presence of cycloidal ordering prohibits the appearance of net ferromagnetic magnetization due to spin canting and a linear magnetoelectric effect. Modifying the spin structure is, therefore, the key for realizing  $\text{BiFeO}_3$ -based ferromagnetic ferroelectrics.

We have recently observed a spin structure transition from low-temperature cycloidal one to high-temperature collinear one at  $\sim 120$  K in rhombohedral  $\text{BiFe}_{0.8}\text{Co}_{0.2}\text{O}_3$  using neutron powder diffraction.[1] Interestingly, magnetization measurements revealed that the collinear phase showed a weakly ferromagnetic behavior with the remanent moments ( $M_r$ ) of  $0.02 \mu_B/\text{f.u.}$  at 300 K, indicating that the spins were canted. In this study, we have fabricated epitaxial  $\text{BiFe}_{1-x}\text{Co}_x\text{O}_3$  (BFCO) thin films on  $\text{SrTiO}_3$  (STO) (111) and  $\text{GdScO}_3$  (GSO) (110) substrates using pulsed laser deposition and their electric/magnetic properties were examined.

The same magnetic transitions were clearly observed in in-plane  $M_r$  versus temperature curves for  $x = 0.10$  and  $0.15$  BFCO thin films on STO substrate at around 220 K and 130 K, respectively, as was observed for bulk samples. These films showed ferromagnetic behavior at 300 K. The value of  $M_r$ ,  $0.038 \mu_B/\text{f.u.}$  is comparable to that reported for bulk BFCO. Temperature-dependent change in spin structure was confirmed by conversion electron Mössbauer spectroscopy measurements performed on a  $x=0.10$  BFCO thin film at various temperatures. From these results, it can be safely concluded that  $x = 0.10$  and  $0.15$  BFCO films are weakly ferromagnetic at room temperature due to spin canting.[2] Ferroelectric and ferromagnetic domain structures of the BFCO films on GSO substrate studied by magnetic force microscopy and piezoresponse force microscopy and the correlation between them will be reported in the presentation.

[1] I. Sosnowska, M. Azuma *et al.*, *Inor. Chem.* **2013**, 52, 13269.

[2] H. Hojo *et al.*, *Adv. Mater.*, *in press*