



“Understanding Spin Textures in (110)-oriented Epitaxial BiFeO₃”

Dr. Daniel Sando

School of Materials Science and Engineering,
UNSW

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Abstract: The room-temperature multiferroic perovskite oxide BiFeO₃ (BFO) has a complex magnetic structure [1]. In the bulk, superimposed on G-type antiferromagnetic (AFM) order is a long-period ($\lambda \approx 64$ nm) spin cycloid, whose propagation vector is typically confined to high-symmetry $\langle -110 \rangle$ -type directions. In (001)-oriented epitaxial BFO films [2] strain plays a decisive role, with high compressive and tensile strains destabilizing the cycloid in favor of pseudo-collinear AFM order [3]. On the other hand, the magnetic structure of (110)-oriented BFO films has been comparatively less studied and is thus not well understood. In such orientation, the cycloid possesses a unique [11-2] propagation vector [4], different from the directions observed in bulk. Here, we combine several experimental techniques – neutron diffraction (ND), Mössbauer spectroscopy (MS), and low-energy Raman spectroscopy (RS) – with first-principles-based calculations to map out the cycloid stability in (110)-oriented BFO films under a wide range of growth, thickness (25-200 nm), strain gradient, and temperature conditions. We reveal using ND that upon reducing the thickness (and increasing temperature toward the AFM transition), the cycloid period increases, possibly related to spin pinning effects at the interface and surface. We show using MS that not only does the average strain level define the existence (or not) of the cycloid, but the lattice distortion of the base plane of the unit cell (i.e. a/b) also plays a critical role. We also show using RS and MS that films subject to large strain gradients (which result in high flexoelectric fields) do not sustain the cycloid. Finally, first-principles-based calculations, taking into account nearest and next-nearest neighbors [5], reproduce the experimentally-observed [11-2] unique propagation direction, and explain the dependence of the cycloid stability on unit cell distortion. These results provide strict guidelines for the design of future magnonic and multiferroic devices that rely on the existence of the spin cycloid in BiFeO₃ and demonstrate that effects such as flexoelectricity can be harnessed to engineer desired spin textures in multiferroics.

[1] J.-G. Park, M. D. Le, J. Jeong, and S. Lee, *J. Phys. Condens. Matter* 26, 433202 (2014). [2] D. Sando, A. Barthélémy, and M. Bibes, *J. Phys. Condens. Matter* 26, 473201 (2014). [3] D. Sando, et al., *Nat. Mater.* 12, 641 (2013). [4] W. Ratcliff, et al., *Adv. Funct. Mater.* 21, 1567 (2011). [5] B. Xu, B. Dupé, C. Xu, H. Xiang, and L. Bellaiche, *Phys. Rev. B* 98, 184420 (2018).