TSAR

TOPOLOGICAL **S**OLITONS IN **A**NTIFE**R**ROICS

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$BiFeO_3$ thin films were grown by pulsed laser deposition on various substrates using a KrF excimer laser (248 nm) with a fluence of 1 J cm^{-2}. Prior to film growth, the scandate substrates ($DyScO_3$, $TbScO_3$, $GdScO_3$, $SmScO_3$) were *ex-situ* annealed for 3 hours at 1000°C under oxygen flow. The $SrTiO_3$ substrate was chemically etched with a buffered HF solution before following the same annealing procedure. For all the samples, a $SrRuO_3$ bottom electrode (3-5 nm) was first grown at 660°C under 0.2 mbar of oxygen pressure with a laser repetition rate of 5 Hz. The $BiFeO_3$ thin film (30-60 nm) was subsequently grown at the same temperature under 0.36 mbar of oxygen pressure and a repetition rate of 2 Hz. Following the growth of the bilayer, the samples were cooled down to room temperature under an oxygen pressure of 300 mbar. X-ray diffraction shows the high epitaxial quality of the films with Laue fringes attesting for their coherent growth. All films display smooth surfaces with atomic steps, characteristic of a layer-by-layer growth. The (001) $BiFeO_3$ peak evolves from the left to the right of the substrate (001) peak upon increase of the in-plane pseudo cubic lattice parameter of the substrate, as observed in the 2θ-ω scans. Reciprocal space maps indicate that the films are fully strained with only two elastic variants of the $BiFeO_3$ monoclinic phase. Their peak positions enable us to determine a strain value for each film ranging from -1.35% compressive strain to +0.50% tensile strain. The out-of-plane and in-plane variants of polarisation were identified in each sample using piezoresponse force microscopy. For all the samples, the as-grown out-of-plane polarisation is pointing downward, i.e. towards the bottom electrode. The films display similar striped-domain structures with two in-plane ferroelectric variants corresponding to the elastic ones observed in reciprocal space maps.

Combining high-quality strain-engineered epitaxial thin films with highly-sensitive scanning NV magnetometry, the magnetic vs. strain phase diagram has been investigated and demonstrated to be richer than previously depicted [1]. Indeed, using $DyScO_3$(110) or $TbScO_3$(110) substrates, imposing low compressive strain, favors the bulk-like (type I) cycloid with $[\bar{1}10]$ propagation vectors restricted to the film plane of $BiFeO_3$. For large compressive strain (on $SrTiO_3$(001)) or small tensile strain (on $GdScO_3$(110)), the exotic type II cycloid is stabilized with propagation vectors along the $[\bar{2}11]$ and $[1\bar{2}1]$ directions, i.e., as close as possible to the film plane of $BiFeO_3$. Finally, large tensile strain (on $SmScO_3$(110)) seems to destroy the cycloid and favor pseudo-collinear canted antiferromagnetic domains. More recently, highly-ordered $BiFeO_3$ thin films were grown on $SmScO_3$(110) in a new pulsed laser deposition chamber and scanning NV magnetometry revealed a type II cycloid in the as-grown state with a diverging period [2]. Hence, the combination of strain-engineering and scanning probe microscopy allows us to identify critical strains that correspond to magnetic phase boundaries (Figure 1) [2,3].

In addition, resorting to anisotropic in-plane strain in epitaxial (111) thin films of $BiFeO_3$ grown on $DyScO_3$(011) orthorhombic substrates, a single domain ferroelectric domain is stabilized all over the sample with a purely vertical polarization. Interestingly, this in-plane distortion favors a single spin cycloid with a propagation vector contained in the (111) plane of $BiFeO_3$ and parallel to the a-axis of the orthorhombic substrate. The epitaxial thin films of $BiFeO_3$ were grown on $DyScO_3$(011) and $TbScO_3$(011) orthorhombic substrates by pulsed laser deposition using a KrF excimer laser. The scandate substrates were preliminary annealed under constant oxygen flow at 1000°C for 3 hours. Ultrathin bottom electrodes of $SrRuO_3$ were first grown under 0.2 mbar of oxygen at 660°C. The $BiFeO_3$ thin films were grown under 0.36 mbar of oxygen at a temperature of 670°C. The whole heterostructure was elaborated with a laser repetition rate of 5 Hz. X-ray reflectivity experiments were performed to determine the thicknesses of $SrRuO_3$ and $BiFeO_3$.

List of fully-characterized samples available to the consortium:

REAL1519B BiFeO3 (62 nm) / SrRuO3 (5 nm) // DyScO3 (011)

REAL1519C BiFeO3 (47 nm) / SrRuO3 (5 nm) // TbScO3 (011)

TURF1091 BiFeO3 (36 nm) / SrRuO3 (5 nm) // TbScO3 (011)

REAL326 BiFeO3 (78 nm) / SrRuO3 (5 nm) // DyScO3 (110)

REAL1035 BiFeO3 (37 nm) / SrRuO3 (5 nm) // DyScO3 (110)

REAL1240 BiFeO3 (60 nm) // DyScO3 (110)

REAL 1352 BiFeO3 (60 nm) // GdScO3 (110)

REAL1369 BiFeO3 (30 nm) / SrRuO3 (5 nm) // DyScO3 (110)

REAL1506B BiFeO3 (33 nm) / SrRuO3 (5 nm) // DyScO3 (110)

TURF716 BiFeO3 (56 nm) / SrRuO3 (5 nm) // GdScO3 (110)

TURF772 BiFeO3 (25 nm) / SrRuO3 (5 nm) // SmScO3(110)

TURF776B BiFeO3 (54 nm) / SrRuO3 (5 nm) // SmScO3 (110)

TURF1070 BiFeO3:Mn (30 nm) / La0.7Sr0.3MnO3 (4 nm) // DyScO3 (110)

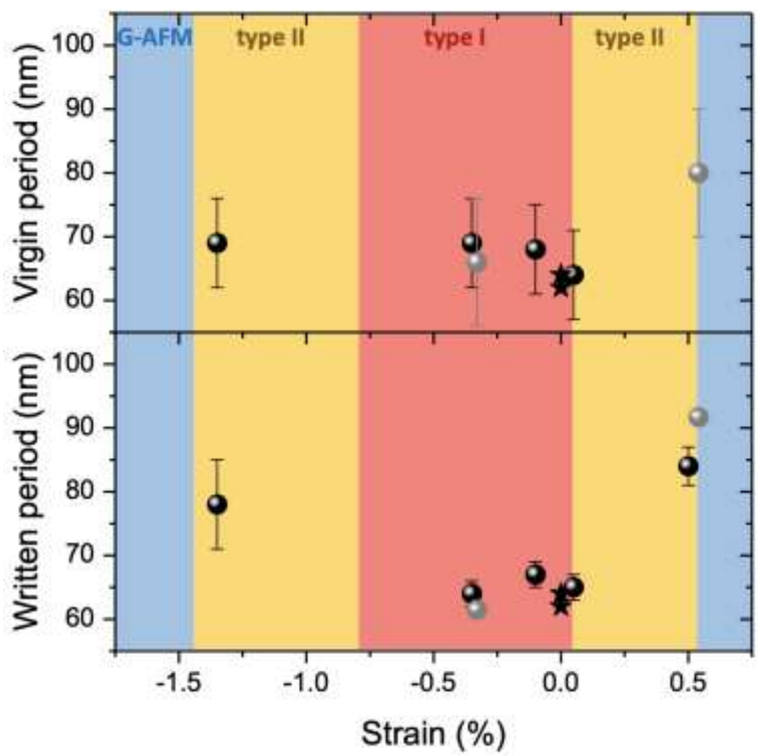

Figure 1. Magnetic state vs strain phase diagram of (001)-oriented $BiFeO_3$ thin films

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[1] D. Sando et al., *Crafting the Magnonic and Spintronic Response of BiFeO3 Films by Epitaxial Strain*, Nature Mater **12**, 641 (2013).
[2] H. Zhong et al., *Quantitative Imaging of Exotic Antiferromagnetic Spin Cycloids in Bi Fe O 3 Thin Films*, Phys. Rev. Applied **17**, 044051 (2022).
[3] A. Haykal et al., *Antiferromagnetic Textures in BiFeO3 Controlled by Strain and Electric Field*, Nature Communications **11**, 1704 (2020).

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