Pattern Transfer and Lateral Switching Modulation in Multiferroic Heterostructures

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The ability to tailor magnetic properties via coupling to ferroelectric domains is of great interest for the design of electric-field tunable magnetic devices. One-to-one imprinting of ferroelectric domains into continuous magnetic films would enable local control over magnetization dynamics but requires strong interfacial coupling to overcome exchange and magnetostatic interactions within the ferromagnet. In this paper, full pattern transfer in multiferroic heterostructures consisting of a ferroelectric substrate and a thin magnetic film with in-plane magnetization is demonstrated. Simultaneous imaging of ferroelectric and ferromagnetic domains and local magnetization reversal analysis using polarization microscopy reveals strong lateral modulations of magnetic hysteresis due to strain coupling to the underlying ferroelectric lattice. The presented experiments open up new avenues for local actuation of magnetic functionalities by external magnetic- and electric fields. Beyond this, the work provides a framework for exploring ferroelectric, ferroelastic, and ferromagnetic domain interactions in multiferroic hetero-structures.

Domain formation and hysteresis in magnetic thin films depend on internal material parameters (exchange stiffness, magnetization), film thickness and shape (magnetostatics), and various other sources of magnetic anisotropy including crystal symmetry, lattice deformations (strain), and interfaces. The interplay between these film properties and an external magnetic field usually results in a laterally uniform energy landscape with random dispersions due to defects and interface roughness. The magnetic hysteresis therefore hardly depends on sample position. Area specific magnetic responses in continuous films require a local modification of the magnetic properties. This can, for example, be accomplished by the growth of ferromagnetic films onto pre-patterned substrates or local ion irradiation. In these cases, the magnetic anisotropy remains fixed after sample preparation. Interface strain coupling between ferroelectric crystals with alternating ferroelastic domains and ferromagnetic films provides an alternative route towards lateral anisotropy control in continuous magnetic media. Moreover, such multiferroic heterostructures hold the potential of electrically tunable magnetism, a phenomenon that has attracted major scientific interest in recent years.

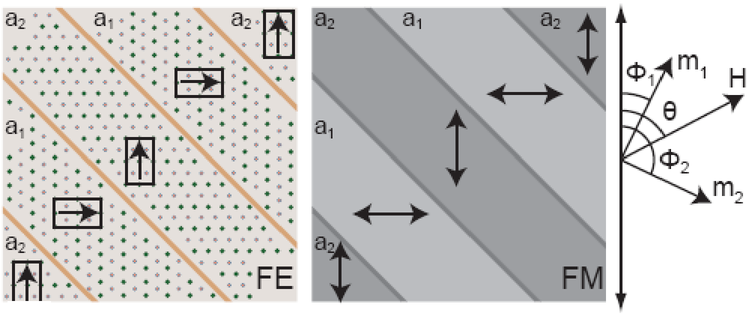

Figure 1 – Schematic illustration of the ferroelastic $BaTiO_3$ domain structure (FE) and the induced magnetic stripe pattern in the Fe film (FM). The arrows in FE indicate the orientation of the ferroelectric polarization and elongated *c*-axis in the *a*1 and *a*2 domains of the $BaTiO_3$ substrate. The uniaxial magneto-elastic easy axes in the Fe film are collinear with the ferroelectric polarization as illustrated by the arrows in FM.

In our approach to manipulate local magnetic properties we used tetragonal $BaTiO_3$ substrates with in-plane polarization. Stress relieve in these substrates results in a regular ferroelastic stripe pattern whereby the elongated *c*-axis (c = 4.036 Å, a = b = 3.992 Å) rotates by 90° in the substrate plane as illustrated in Fig. 1. Thus, the alternating *a*1 and *a*2 domains provide a maximum lateral strain modulation of about 1.1%. Efficient coupling of a magnetic thin film to this ferroelestic template induces uniaxial magnetoelastic anisotropy with orthogonal magnetic easy axes in neighboring domains. A demonstration of one-to-one imprinting of $BaTiO_3$ domains into a continuous 10 nm thick Fe film is shown in Fig. 2. Here, strong interfacial strain coupling induces different magnetic responses in the Fe film on top of the *a*1 and *a*2 domains leading to an exact copy of the ferroelastic stripe pattern. Polarization analysis of the birefringent ferroelectric domains and the magnetization reversal process indicate that the ferroelectric polarization of the $BaTiO_3$ substrate and the strain-induced uniaxial magnetic easy axes of the Fe film are collinear. This multiferroic configuration results in coherent magnetization rotation towards the magnetic easy axes of the *a*1 and *a*2 domains when the applied magnetic field is reduced from saturation to zero. Thus, in the remnant state the film magnetization is laterally modulated by near 90° rotations at domain boundaries. Reversal of the applied magnetic field induces inverse domain nucleation in *a*2 domains and more gradual magnetization reversal in *a*1 domains. The imprinted stripe pattern in the Fe film remains visible until the magnetization is fully saturated.

The dependence of multiferroic pattern transfer on the direction and strength of the applied magnetic field, exchange and stray-field coupling between magnetic stripe domains, and other anisotropy contributions will be discussed in detail.

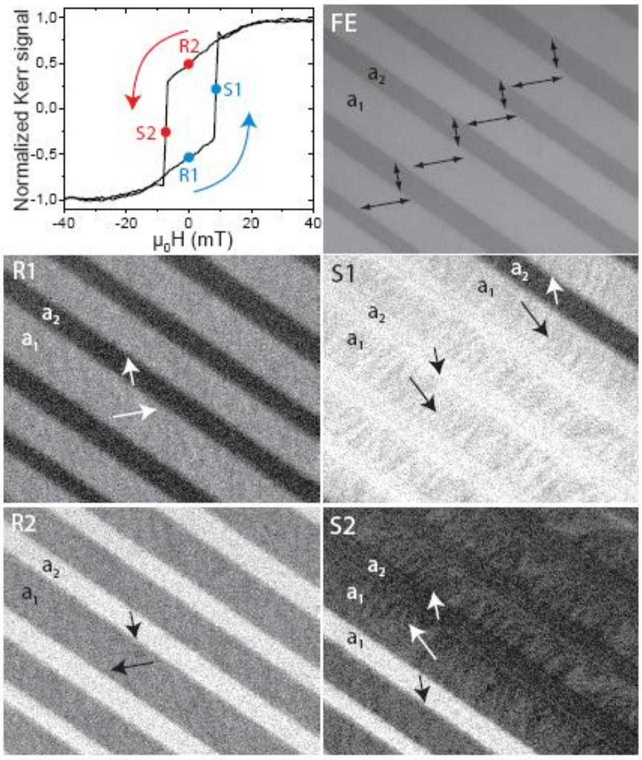

Figure 2 – Magnetic hysteresis curve and polarization microscopy images of the ferroelectric domain structure and magnetic stripe pattern during several stages of the magnetization reversal process. The figure depicts experimental data for a magnetic field angle of 10°. The hysteresis curve is a global measurement and thus an average magnetic response from several *a*1 and *a*2 domains. The arrows in the images indicate the orientation of ferroelectric polarization (FE) and film magnetization in the remnant state (R1 and R2) and during abrupt magnetic switching (S1 and S2).