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Alternating domains with uniaxial and biaxial magnetic anisotropy in epitaxial Fe films on BaTiO₃

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We report on domain formation and magnetization reversal in epitaxial Fe films on ferroelectric BaTiO₃ substrates with ferroelastic a–c stripe domains. The Fe films exhibit biaxial magnetic anisotropy on top of c domains with out-of-plane polarization, whereas the in-plane lattice elongation of a domains induces uniaxial magnetoelastic anisotropy via inverse magnetostriction. The strong modulation of magnetic anisotropy symmetry results in full imprinting of the a–c domain pattern in the Fe films. Exchange and magnetostatic interactions between neighboring magnetic stripes further influence magnetization reversal and pattern formation within the a and c domains. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4773482]

Ferromagnetic pattern formation via efficient coupling to ferroelectric domain structures has recently been demonstrated.¹–⁶ Direct correlations between ferromagnetic and ferroelectric domains and its persistence during ferroelectric polarization reversal open up promising ways for electric-field control of local magnetic switching¹–⁵ and the motion of magnetic domain walls.⁶ In systems based on interlayer strain transfer, the ferroelastic domain structure of a ferroelectric material induces local magnetoelastic anisotropies in a ferromagnetic film via inverse magnetostriction. Within the ferromagnetic sub-system, the magnetoelastic anisotropy competes with intrinsic magnetic properties including magnetocrystalline anisotropy and exchange and magnetostatic interactions between domains. Consequently, the evolution of the magnetic microstructure in an applied magnetic or electric field depends critically on the two ferroic materials, the ferromagnetic layer thickness, and the ferroelastic domain size.

In a previous study, full pattern transfer from ferroelectric BaTiO₃ substrates with alternating ferroelastic a₁ − a₂ domains to polycrystalline Co₆₀Fe₄₀ thin films was analyzed.¹–⁶ The strain-induced uniaxial magnetoelastic anisotropy axis of this system rotates by 90° at domain boundaries and this fully dominates the local magnetic properties since the magnetocrystalline anisotropy of Co₆₀Fe₄₀ is negligibly small. Here, we report for the first time on full imprinting of ferroelectric BaTiO₃ a–c domain patterns into epitaxial Fe films as schematically illustrated in Fig. 1(a). Magneto-optical Kerr effect (MOKE) microscopy measurements indicate that the magnetic anisotropy is laterally modulated by the alternating in-plane structural symmetry of the BaTiO₃ lattice. Moreover, it is shown that magnetic switching in neighboring a and c domains is strongly coupled via exchange and magnetostatic interactions. The demonstrated ability to initialize a rich variety of micromagnetic configurations in Fe/BaTiO₃ supports the design of electric-field controlled magnetic structures including magnonic crystals and spintronic devices.

In the experiments, 10 nm and 20 nm thick Fe films with a 5 nm Au capping layer were grown onto single-crystal BaTiO₃ substrates using molecular beam epitaxy.⁷,⁸ Film growth at 300°C in an ultrahigh vacuum chamber resulted in epitaxial Fe with an in-plane Fe [110] || BaTiO₃ [100]

Figure 1. (a) Schematic illustration of the domain configuration in the Fe/BaTiO₃ system. The arrows in the BaTiO₃ substrate indicate the direction of ferroelectric polarization, and the double-headed arrows in the Fe film represent the magnetic easy axes. The schematics on the right illustrate the in-plane lattice structure and polarization direction of the BaTiO₃ substrate and the orientation of the Fe film in the a and c domains. The room-temperature lattice parameters of the BaTiO₃ substrate and the in-plane strains of the Fe film are indicated. The strains are calculated relative to the Fe bulk lattice parameter on the basis of full strain transfer between substrate and film. A cross-sectional TEM image and a selected area electron diffraction (SAED) pattern of a 10 nm thick epitaxial Fe film on BaTiO₃ (001) are shown in (b) and (c).

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alignment as confirmed by transmission electron microscopy (Fig. 1(b) and (c)). The ferroelastic domain pattern of the BaTiO$_3$ substrates consisted of alternating $a$ and $c$ stripe domains with an average width of 6 $\mu$m and 3 $\mu$m at room temperature. This regular domain structure was formed upon sample cooling through the ferroelectric Curie temperature ($T_C = 120^\circ$C) after Fe film growth. At $T_C$, the structure of BaTiO$_3$ changes from cubic to tetragonal. The ferroelastic $a$ and $c$ domains in tetragonal BaTiO$_3$ impose different local strains on the epitaxial Fe films. For full strain transfer, the in-plane lattice of the Fe films is compressed by $1.6\%$ and $0.6\%$ with respect to that of the bcc Fe bulk structure ($a_{Fe} = 2.870$ Å) as illustrated in Fig. 1(a). On top of the ferroelastic $c$ domains, the Fe lattice remains cubic. However, due to different shear strains in the [110]$_{Fe}$ and [−110]$_{Fe}$ directions, the in-plane structure of the Fe films changes into a diamond shape on top of the BaTiO$_3$ $a$ domains. MOKE microscopy was used to analyze magnetization reversal in the $a$ and $c$ domains as a function of magnetic field. Images were recorded with two orthogonal magneto-optical contrast axes ($\omega$) by rotation of the optical plane of incidence using an adjustable diaphragm. From these two images, the local magnetization direction was extracted. The MOKE microscope setup was also used to measure magnetic hysteresis curves on single $a$ and $c$ stripe domains for different in-plane magnetic field angles ($\theta$).

Figure 2 shows MOKE microscopy images of a 20 nm thick Fe film on top of BaTiO$_3$ for three magnetic field directions. The regular magnetic stripes directly correlate with the ferroelastic $a$–$c$ pattern of the BaTiO$_3$ substrate. The magnetic domain walls are strongly pinned onto the ferroelectric domain boundaries by abrupt changes in local magnetic anisotropy.\(^6\)\(^9\) As a result, the overall stripe pattern does not alter in an applied magnetic field (until the film is saturated) and magnetic switching in the $a$ and $c$ domains differs considerably. For a magnetic field angle of $\theta = 0^\circ$ ($H$ perpendicular to the domains), the magnetization of the $a$ domains rotates coherently as indicated by the white arrows in the images on the top rows of Fig. 2. On the other hand, abrupt switching is observed for $\theta = 90^\circ$ ($H$ parallel to the domains). In the images on the bottom rows of Fig. 2, this switching event can be clearly seen at 2 mT. The difference in magnetization reversal mechanism reflects the uniaxial magnetic symmetry of the $a$ domains, which is also confirmed by the local magnetic hysteresis curves of Fig. 3 and the polar plot of the remnant magnetization in Fig. 4(a). The magnetic easy axis of the $a$ domains in the epitaxial Fe films is oriented parallel to the magnetic domains ([−110]$_{Fe}$ direction) and, thus, perpendicular to the in-plane lattice elongation and polarization direction of the ferroelectric BaTiO$_3$ substrate. This qualitatively

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The same result is obtained for the biaxial compressive strain along the in-plane lattice of BaTiO3 remains cubic when the in-plane Fe lattice exhibits a diamond shape due to a shear strain along the diagonal [110]_Fe direction. Consequently, the uniaxial magnetic easy axis is oriented along the BaTiO3 ferroelastic polarization. The strength of the magnetoelectric anisotropy as determined from the slope and saturation field of hard-axis hysteresis curves is K_{me} = 5 \pm 2 \times 10^6 \text{ J/m}^3. The close agreement between the calculated and experimentally measured uniaxial magnetoelectric anisotropy indicates that the ferroelastic strain of the BaTiO3 substrates is fully transferred to the epitaxial Fe films when the samples are cooled after MBE growth.

Magnetization reversal in the c domains of the Fe films is strikingly different. The polar plot of the remnant magnetization in Fig. 4(a) clearly illustrates that the magnetic anisotropy of the c domains exhibits fourfold symmetry. The easy magnetization axes are aligned along [100]_Fe and [010]_Fe, in agreement with the magnetocrystalline anisotropy of bulk Fe. Moreover, the in-plane lattice of BaTiO3 remains cubic when c domains form during sample cooling through T_C. The lattice mismatch between Fe (a_{Fe} = 2.870 \text{ Å}) and BaTiO3 (a_{BaTiO3} = 3.992 \text{ Å}) at room temperature, therefore, induces a biaxial compressive strain of -1.6\% along the cubic axes. From the first term in Eq. (1) and ε_{cx} = ε_{cy} = -1.6\%, x_z = 1, x_y = 0, and B_1 = -3.43 \times 10^6 \text{ J/m}^3,\text{ }^{11} the magnetoelectric anisotropy along [100]_Fe can be estimated. This gives K_{me} = -5.5 \times 10^4 \text{ J/m}^3.

The induced magnetization rotation in the Fe and BaTiO3 domains instantaneously rotates when the Fe domains switch by 180\˚ between both domains. The second switching in the Fe domains can only be rationalized by considering the magnetic energy of the Fe/BaTiO3 system. If the c domains would switch prior to the a domains, the magnetization angle between both domains would increase from about 45\˚ to 135\˚ (see, circular symbols in Fig. 3). This would enhance both the exchange and magnetostatic energy. Simultaneous switching in both domains is, therefore, energetically more favorable because it limits the maximum magnetization angle between neighboring domains to 45\˚ throughout the reversal process. Since the magnetization of the a domains is parallel to the stripe pattern, their stray field hardly influences the magnetization direction in the c domains. Hence, ferromagnetic coupling between both stripe domains is dominated by short-range exchange interactions.
reversal in the $a$ domains. Both exchange and magnetostatic interactions might contribute to domain coupling in this case.

The polar plot of Fig. 4(b) summarizes the switching fields of the $a$ and $c$ domains as a function of magnetic field angle. Strong coupling between neighboring stripe domains results in identical switching fields for both domains irrespective of field direction. The vertical lines represent magnetic switching events that are triggered by uniaxial $a$ domains. The shape of these curves deviates significantly from the Stoner-Wohlfarth asteroid,\textsuperscript{13} which is obtained when the switching fields of isolated uniaxial domains are simulated (Fig. 4(c)). In the Stoner-Wohlfarth case, a maximum switching field is obtained when the field is applied along the uniaxial magnetic easy axis ($\theta = 90^\circ$). In our Fe/BaTiO$_3$ samples, strong coupling between neighboring stripe domains rotates the magnetization of the $a$ domains away from its magnetic easy axis during magnetization reversal, and this drastically reduces the switching field for $\theta = 90^\circ$. Similarly, the horizontal lines in Fig. 4(b) indicate magnetic switching events that are induced by biaxial $c$ domains. Again, the shape of these experimental curves differs completely from the simulated polar plot of isolated domains with biaxial anisotropy (Fig. 4(c)). The measured polar plot of the switching fields in Fe/BaTiO$_3$ is unique. Both the vertical and horizontal curves have no analogue in systems with uniform magnetic anisotropy. We also note that our microscopic observation of a small intrinsic $c$-domain switching field agrees with previous macroscopic studies demonstrating a reduction of magnetic switching field after the application of an out-of-plane electric field.\textsuperscript{7,14}

Finally, we show that efficient coupling between $a$ and $c$ domains influences the magnetic pattern within $a$ and $c$ stripe domains of the Fe films. If both domains would switch independently, the evolution of the magnetic microstructure of the biaxial $c$ domains would be similar for $\theta = 0^\circ$ and $\theta = 90^\circ$. The MOKE microscopy images of Fig. 2 clearly show that this is not the case. For $\theta = 0^\circ$, the magnetization of the $a$ domains coherently rotates clockwise and, due to ferromagnetic interactions between domains, forces the magnetization of the $c$ domains to reverse in the same direction. In small magnetic field, 90$^\circ$ domains nucleate within the $c$ stripes. These domains are highly mobile and their size increases rapidly with applied field strength. As a result, the magnetic uniformity of the $c$ domains is re-established at a field of about 3 mT. For $\theta = 90^\circ$, the magnetization of the $a$ domains remains fixed along its uniaxial magnetic easy axis. In this case, exchange interactions with neighboring $c$ domains do not favor a particular reversal direction. The $c$ domains, therefore, split up into a regular stripe pattern by alternating clockwise and anti-clockwise magnetization rotation. Due to negligible domain wall motion within the $c$ domains under these conditions, the stripe pattern is robust until it abruptly changes during simultaneous magnetic switching in both domains. In this case, a large magnetic field is required to fully remove the stripe pattern from the $c$ domains.

In summary, the results of this paper clearly demonstrate that magnetic domains can be imprinted into continuous magnetic films via efficient coupling to ferroelastic domains of a ferroelectric material. The magnetization reversal mechanism in such domains depends on the symmetry of the strain-induced magnetoelastic anisotropy and the applied field angle. Moreover, ferromagnetic coupling between neighboring domains strongly influences local magnetic switching events. Pattern formation in epitaxial Fe films on ferroelectric $a$–$c$ domains of BaTiO$_3$ is particularly rich. In this system, magnetic stripe domains with biaxial and uniaxial anisotropy alternate. Moreover, depending on the direction of the applied magnetic field, highly mobile, or robust magnetic patterns form within the $c$ domains, a property that could be exploited in magnetic devices.

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