Temperature dependence of ferromagnet-antiferromagnet spin alignment and coercivity in epitaxial micromagnet bilayers

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Exchange coupling at the interface between ferromagnetic (FM) and antiferromagnetic (AFM) materials has immense technological significance [1]. Implementation of magnetically coupled interfaces into devices typically requires patterning of thin films into micro- or nanoscale features, but it is often seen that the properties of fabricated systems differ from those of thin films. For example, it has long been known that coercivity, the external field required to switch a magnetic bit in the opposite direction, increases with decreasing FM particle dimension, but rapidly decreases once the superparamagnetic regime is reached [2]. The exchange bias field between FM and AFM layers that imposes a unidirectional anisotropy on the FM layer can be used to further modify magnetic switching [1]. This interaction has been studied experimentally in microstructures, and results show that the magnitude of the bias field may be reduced by patterning. However, the origin of the size dependence is less clear [3]. An epitaxial exchange coupled system introduces crystallographic dependence of the coupling behavior that is not present in polycrystalline designs. The interface between AFM LaFeO$_3$ (LFO) and FM La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) is one intriguing example when these materials are grown epitaxially on (001)-oriented SrTiO$_3$. Folven et al. verified this arrangement to be the energetically preferred coupling, yet in patterned bilayer films, a transition exists from spin-flop parallel alignment by varying the temperature between 30 and 300 K. Results show that not only does this spin alignment noticeably influence the bilayer micromagnet coercivity compared to La$_{0.7}$Sr$_{0.3}$MnO$_3$ single-layer micromagnets, but the coercivity within this materials system can be tuned over a wide range by careful balance of material properties.

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I. INTRODUCTION

Exchange coupling at the interface between ferromagnetic (FM) and antiferromagnetic (AFM) materials has immense technological significance [1]. Implementation of magnetically coupled interfaces into devices typically requires patterning of thin films into micro- or nanoscale features, but it is often seen that the properties of fabricated systems differ from those of thin films. For example, it has long been known that coercivity, the external field required to switch a magnetic bit in the opposite direction, increases with decreasing FM particle dimension, but rapidly decreases once the superparamagnetic regime is reached [2]. The exchange bias field between FM and AFM layers that imposes a unidirectional anisotropy on the FM layer can be used to further modify magnetic switching [1]. This interaction has been studied experimentally in microstructures, and results show that the magnitude of the bias field may be reduced by patterning. However, the origin of the size dependence is less clear [3]. An epitaxial exchange coupled system introduces crystallographic dependence of the coupling behavior that is not present in polycrystalline designs. The interface between AFM LaFeO$_3$ (LFO) and FM La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) is one intriguing example when these materials are grown epitaxially on (001)-oriented SrTiO$_3$ (STO) substrates. LFO is a G-type antiferromagnetic material [4] which produces a fully compensated (001) surface of Fe$^{3+}$ moments. Heisenberg modeling predicts such an interface would align the FM and AFM spin axes perpendicular to one another—a spin configuration known as spin-flop coupling [5,6]. Folven et al. verified this arrangement to be the energetically preferred coupling, yet in patterned bilayer films, a transition exists between spin-flop and parallel alignment as a function of both micromagnet aspect ratio and crystallographic orientation of the patterned structures [7]. Understanding the influence of these parameters in micromagnetic structures is critical for device design and suggests that a wide range of magnetic behavior can be tuned within a single material system.

Subsequent work on LFO/LSMO bilayers characterized the switching behavior of 2 $\mu$m x 0.5 $\mu$m patterned micromagnets oriented along the in-plane (100) directions and demonstrated that parallel spin alignment reduces the switching field relative to a patterned LSMO single layer [8]. This decreased coercivity occurred since the AFM and FM moments prefer perpendicular alignment with one another. Thus, when switching the direction of the FM moment by 180°, the midpoint of the rotation of the FM magnetization becomes less unfavorable, thereby reducing the energetic barrier to switching.

Here we demonstrate that the FM/AFM spin alignment in (110)-oriented epitaxial micromagnets is highly temperature dependent and that it is possible to stabilize different FM/AFM spin alignments. Relying on element specific domain imaging, we show that parallel, frustrated, and spin-flop states can be reached by varying the temperature between 300 and 30 K within the same micromagnet array. This fluctuation stems from the vastly different ordering temperatures ($T_{N}$ = 670 K [9] for LFO and $T_{C}$ = 270 K [7] for LSMO) of the individual layers. A Stoner-Wohlfarth free energy model developed to quantify the bias field imposed on the LSMO layer by the adjacent LFO layer predicts (relative to an LSMO single layer) an increased switching field in spin-flop systems, reduced in the parallel configuration, and no change in the frustrated case (i.e., one with an equal mix of spin-flop and parallel aligned AFM domains within a single micromagnet). By applying in situ field pulses, we test this model and also show that crystallographic orientation alone has considerable influence on the micromagnet coercivity. In total, this work demonstrates the variety of switching behavior that can be obtained in bit-patterned micromagnets through the combination of exchange-coupled systems with patterning of epitaxial multilayer films.

II. METHODS AND MATERIALS

Two patterned epitaxial films were investigated in this work. The first was a 100 unit cell (u.c.) thick LSMO layer and the...
other was a bilayer consisting of 10 u.c. LFO grown on top of 90 u.c. LSMO. The samples were deposited by pulsed laser deposition on (001)-oriented Nb-doped (0.05 wt. %) STO substrates. The growth and structural characterization were previously described in Ref. [8], and the films showed excellent crystalline and epitaxial quality. Within each film, microstructures are defined by a Cr hard mask using electron-beam lithography. A subsequent flood implantation of Ar⁺ ions modifies the crystalline structure throughout the film thickness in all exposed regions, thus locally eliminating the magnetic order. This technique results in micromagnets embedded within a nonmagnetic matrix. According to Stopping and Range of Ions in Matter simulations, implantation straggle does not appreciably modify the structural or magnetic quality of the microstructure beyond 30 nm of the pattern edge [10]. More details on this patterning process can be found in Refs. [11,12]. The first of two microstructures discussed in this study are zigzag patterns with 2 μm × 0.5 μm segments oriented along alternating in-plane (110) directions, and were measured between 30 and 300 K to evaluate the temperature dependence of the FM/AFM spin alignment. Arrays of one hundred nominally identical 2 μm × 0.5 μm rectangular bits, oriented along the in-plane (110) direction, were also patterned into both films to study the magnetic switching behavior of these micromagnets.

X-ray photoemission electron microscopy (X-PEEM) was used to image the AFM and FM domains as a function of temperature between 30 and 300 K at the PEEM3 endstation on beamline 11.0.1 at the Advanced Light Source [13]. FM domain contrast in X-PEEM emerges from x-ray magnetic circular dichroism (XMCD) at the Mn L₂,₃ absorption edge, where the contrast intensity is proportional to the cosine of the angle between local FM moment orientation and the incident x-ray helicity vector. AFM domain images were obtained by utilizing the x-ray magnetic linear dichroism (XMLD) effect at the Fe L₂,₃ absorption edge which is parametrized by \( I(\theta) = a + b(3\cos^2\theta - 1)(L^2) \), where \( a \) and \( b \) are constants, \( L \) is the AFM moment, and \( \theta \) is the angle between \( L \) and the \( E \) vector of the linearly polarized x rays [9]. X-PEEM images were collected with the x-ray \( E \) vector oriented along the [110] direction at two energies corresponding to the two local maxima of the Fe L₂ absorption edge. It has been shown that these energies possess opposite sign in the XMLD spectra [14]. In order to enhance the AFM domain contrast and to eliminate any topographical contrast, the final AFM domain image is the calculated asymmetry between individual images (i.e., difference normalized by the sum). The sample holder contains an electromagnet that permits application of magnetic fields up to 190 Oe parallel or antiparallel to the in-plane projection of the incident x rays for a duration of 1 s. After each field pulse, a smaller reverse field was applied to remove remanent fields in the magnetic yoke. This compensating field is at most 25% of the initial field pulse, so unwanted switching in the micromagnets does not occur. All images are captured in the remanent, zero field state.

Experimental results were compared to micromagnetic simulations performed using MuMax³ [15]. Unless otherwise noted, the standard inputs correspond to LSMO parameters at 100 K. These inputs include the saturation magnetization \( M_s \) (400 kA/m), cubic magnetocrystalline anisotropy constant \( K_1 (-2 \text{kJ/m}^3) \), and exchange stiffness \( A_{\text{ex}} (3.8 \text{pJ/m}) \). The simulation cell volume was set to 5 × 5 × 5 nm³ in order to calculate the lowest energy state of a specific magnetic microstructure, in either remanence or applied fields, using the Landau-Lifshitz-Gilbert equation [15]. Materials parameters were obtained from Lee et al. [16].

III. RESULTS

X-PEEM images of the bilayer sample in Fig. 1 present the temperature dependence of FM/AFM spin alignment within zigzag wires with 2 μm × 0.5 μm segments oriented along the in-plane (110) directions. At all temperatures below \( T_C \), the FM domain structure obeys shape anisotropy constraints (i.e., moments aligned along the segment long axis). The (110) directions also coincide with the FM easy axis in continuous LSMO films grown on (001)-oriented STO substrates [17], further stabilizing the domain configuration seen in Fig. 1. At elevated temperatures near and above \( T_C \) of the LSMO sublayer, the AFM domains also follow a shape-induced anisotropy, leading to parallel alignment of the FM and AFM spin axes. This observation is consistent with prior work on similarly fabricated LFO microstructures that showed shape-induced AFM domain formation resulting from

FIG. 1. (Left column) XMLD images of the LFO layer in the zigzag wires at temperatures between 30 K, where the AFM spin axis is perpendicular to the wire edge (spin-flop), through frustrated alignments at 100 and 200 K, to 300 K, where the AFM spin axis becomes parallel to the wire edge. (Right column) Crystallographic legend, XMCD image of the LSMO zigzag wire at 100 K with FM domains oriented parallel to edges of the wire, and a schematic ofnanowire dimensions. The incident x rays are along the [110] direction for all X-PEEM images.
structural constraints present in a patterned, epitaxial film in a surrounding nonmagnetic matrix [18]. This parallel orientation of spin axes breaks down below 200 K as the frustrated state emerges, and the LFO layer breaks down into many smaller domains approximately a few hundred nanometers in diameter aligned along [110] and [110] directions. These respective directions correspond to the dark and light regions observed in the XMLD images of the zigzag nanowire acquired at 100 and 200 K in Fig. 1. Reducing the temperature further pushes the system fully into the spin-flop configuration, as the AFM spin axis is now always perpendicular to the FM moments and the long axis of the patterned segments. Within this experimental range, the magnetization in the LSMO layer changes considerably, from zero at 300 K to 400 kA/m at 30 K. Our results suggest that for low LSMO magnetization, the shape-induced effects dominate the AFM domain structure, while at high LSMO magnetization, the exchange coupling between the FM and AFM spins prevails. These two regimes are separated by a frustrated region where a mixture of parallel and perpendicular spin alignment is found.

In order to study the magnetic switching behavior, 2.μm × 0.5 μm rectangular micromagnets were characterized from a series of X-PEEM images captured under remanent conditions between in situ magnetic field pulses of approximately 1 s in duration at temperatures ranging from 30 to 200 K. The applied fields induce no observable change in XMLD images of the LFO sublayer, but 180° switching is readily seen in XMCD images of the LSMO sublayer, where the contrast switches completely from dark to light. This contrast change indicates a complete reversal of magnetization between the two stable monodomain states of the FM layer within individual micromagnets. Tallying the switching events after each field pulse allows for the construction of the magnetization switching curves shown in Fig. 2(c), which are fit to a normal cumulative distribution function. No unidirectional exchange bias is observed as the switching curves are experimentally identical upon application of field pulses in the opposite direction. Coercivity of both single-layer and bilayer samples increases as the temperature decreases, but the bilayer micromagnets consistently require a greater applied field to reverse magnetization. The spread in switching fields for an individual micromagnet consistently requires a greater applied field to reverse magnetization between 0° and 180°. This parallel orientation is the free energy minimum defined by the derivative of $F$ with respect to $\varphi_{FM}$. This point becomes unstable as the second derivative also approaches zero under the application of an external field, leading to the magnetic reversal of the FM layer (e.g., $\varphi_{FM}$ rotates from 0° to 180°). Since $\varphi_{FM}$ is constant at a specific temperature while rotating the FM moment via an applied field, the $F_{AFM}$ term is not present in the derivatives and therefore has no influence on the equilibrium and reversal conditions. Thus, the relevant free energy landscape is determined by the characteristics of the FM layer, its response to applied fields, and the angle between the FM and the AFM spin axes used to calculate $F_{coupling}$. It can now be seen how $F_{coupling}$ modifies the energy barrier between $\varphi_{FM} = 0°$ and $\varphi_{FM} = 180°$ that must
ends pinned to atomic scale defects 
propagation of flux-closure domain walls nucleated at wire 
bistable magnetic microwires that are known to switch via 
switching field distribution width increases as temperature 
both 
be overcome by the external field for both parallel alignment, 
\[ F_{\text{parallel}}^{\text{coupling}} = \frac{1}{2} M_0 V_0 H_{\text{coupling}} \cos^2(\phi_{\text{FM}} - 0^\circ) \]
and spin-flop alignment, 
\[ F_{\text{spin-flop}}^{\text{coupling}} = \frac{1}{2} M_0 V_0 H_{\text{coupling}} \cos^2(\phi_{\text{FM}} - 90^\circ) \]

Relative to the single layer LSMO, involving only \( F_{\text{FM}} \),
the parallel configuration reduces the barrier height, whereas 
spin-flop does the opposite. In the frustrated case, where 
there is an equal mixture of parallel and spin-flop domains in 
The AFM layer, then \( F_{\text{coupling}} = F_{\text{parallel}}^{\text{coupling}} + F_{\text{spin-flop}}^{\text{coupling}} \), which 
can be reduced to a constant and is no longer present in 
the derivatives that determine the equilibrium and reversal 
conditions.

As outlined above, aspects of Stoner-Wohlfarth theory 
apply to this system to guide our understanding, but 
rigorous, analytical calculations cannot be accurately applied. 
The primary complication stems from the Stoner-Wohlfarth 
assumption that switching occurs through coherent rotation 
within a fully saturated magnet. This notion is valid for 
structures with dimensions on the order of the material’s 
exchange length [19] (approximately 6 nm for LSMO [20]), 
but the micromagnets of this study are much larger. Incoherent 
switching and flux-closure domains (shown in Fig. 3) at 
the short ends of the rectangular micromagnets prevent accurate 
determination of \( H_{\text{shape}} \). It can be seen in Fig. 2 that the 
switching field distribution width increases as temperature 
decreases, which is consistent with experiments on amorphous bistable magnetic microwires that are known to switch via 
propagation of flux-closure domain walls nucleated at wire 
ends pinned to atomic scale defects [21]. MuMax\(^3\) simulations of the 
LSMO micromagnets also indicate that domain wall motion 
plays a dominant role in their switching mode [15]. 
These reversal processes dominate at the macroscopic level to 
determine the coercivity of these micromagnets, but aspects of 
Stoner-Wohlfarth theory appear to be applicable at the mesoscopic level. Namely, \( F_{\text{coupling}} \) can show how structures with equivalent switching mechanisms, but different coupling 
interactions, should behave relative to one another. Individual FM 
moments are still sensitive to adjacent AFM domains during 
rotation and switching, even if the interplay between FM and 
AFM moments is not coherent throughout the micromagnet.

The data to test this hypothesis are presented in Fig. 2(c), 
which captures the distribution of switching events as a 
function of applied field for the bilayer as the spin alignment 
transitions from parallel at 300 K towards spin-flop at 30 K 
alongside a similar set of curves for the FM single layer, for 
reference. To more intuitively compare the switching behavior 
over this temperature range, Fig. 4 plots the switching field 
declared as the value where 50% of the micromagnets have 
switched, as determined by the respective fit curves in Fig. 2(c). 
At all temperatures, the bilayer requires a higher applied field 
to switch. Additionally, the (100) data from [8] has been 
included for a comprehensive depiction of the system.

Above 30 K, the single layer and bilayer show the same 
gradual decrease in switching field with temperature, but with 
an offset of 10%–15%. In this regime, frustrated spin alignment 
dominates and equivalent coercivities would be expected for 
the two structures. The slight offset is a result of the difference 
in FM thickness between the single layer (100 u.c. LSMO) 
and bilayer (90 u.c. LSMO). MuMax\(^3\) simulations show that a 
90 u.c. FM micromagnet at these dimensions will switch at a 
field \( \sim 10\% \) greater than the 100 u.c. layer [15], which 
suggests that the experimental switching behavior between 
60 and 200 K is consistent with a thickness-dependent effect.

The switching fields near 30 K show a clear divergence 
from the trend present between 60 and 200 K. The coercivity 
of the bilayer jumps to a value larger than can be accounted
by the difference in LSMO layer thickness, and the X-PEEM images (e.g., Fig. 1) verify the spin-flop alignment emerging at 30 K that should lead to an increased coercivity. This outcome is again in agreement with the influence of the interfacial coupling component of the free energy model. Folven et al. found that parallel coupling in (100)-oriented micromagnets at 110 K reduced the AFM/FM bilayer switching field by 30% relative to the FM single-layer micromagnets [8]. The spin-flop alignment that emerges at 30 K in the (110)-oriented micromagnets shown here increases the switching field by approximately 25% (after correcting for the difference in LSMO thickness).

The large difference in switching field between the (100)- and (110)-oriented micromagnets at 110 K emerges because of the remanent FM domain structure in each type of micromagnet as described by both X-PEEM data and simulated domain pattern images in Fig. 3. As stated earlier, switching in this system occurs via a domain wall nucleation and propagation mechanism. Rectangular micromagnets oriented along (110) directions are likely to form vortex flux-closure domains at the ends, which greatly reduces the switching field as domain walls are already nucleated within the structure. This FM domain structure persists at 30 K in the bilayer sample despite strong spin-flop coupling to the LFO layer. Micromagnets oriented along (100) directions do not form this structure, as doing so introduces too large of a magnetocrystalline anisotropy energy cost, and domain walls must be nucleated by the applied field. It is expected that minor modifications to the pattern geometry should provide even further control over remanent magnetization states and thus coercivity.

V. CONCLUSIONS

In summary, we demonstrate that considerable variation in coercivity (30 to 190 Oe) takes place when the spin alignment at the FM/AFM interface in an epitaxial micromagnet is altered. Depending on the temperature and crystalline orientation of such micromagnets, parallel, frustrated, and spin-flop alignment can be stabilized. Using a combination of micromagnetic simulations and free energy modeling, the switching behavior of this complex system can be accurately characterized. While maintaining many aspects of the micromagnet constant such as crystallinity, chemistry, or fabrication conditions, our findings emphasize the sensitivity of spin alignment on material properties of the individual layers and the resulting effect on functional properties.

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