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Authors
Lee, Michael S
Wynn, Thomas A
Folven, Erik
et al.

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Tailoring Spin Textures in Complex Oxide Micromagnets

Michael S. Lee,†,⊥ Thomas A. Wynn,†,⊥ Erik Folven,‡ Rajesh V. Chopdekar,† Andreas Scholl,§ Anthony T. Young,§ Scott T. Retterer,‖ Jostein K. Grepstad,‡ and Yayoi Takamura*†

†Department of Materials Science and Engineering, University of California—Davis, Davis, California 95616, United States
‡Department of Electronics and Telecommunications, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway
§Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94703, United States
‖Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States

Supporting Information

ABSTRACT: Engineered topological spin textures with submicron dimensions in magnetic materials have emerged in recent years as the building blocks for various spin-based memory devices. Examples of these magnetic configurations include magnetic skyrmions, vortices, and domain walls. Here, we show the ability to control and characterize the evolution of spin textures in complex oxide micromagnets as a function of temperature through the delicate balance of fundamental materials parameters, micromagnet geometries, and epitaxial strain. These results demonstrate that in order to fully describe the observed spin textures, it is necessary to account for the spatial variation of the magnetic parameters within the micromagnet. This study provides the framework to accurately characterize such structures, leading to efficient design of spin-based memory devices based on complex oxide thin films.

KEYWORDS: complex oxides, micromagnetics, magnetic anisotropy, X-ray photoemission electron microscopy

Spin textures consist of spatially varying spin arrangements that form within patterned magnetic materials with nano-/microscale dimensions and are considered to have substantial promise as the basis for spin-based memory devices.1−7 In particular, magnetic skyrmions have received significant interest for future spintronic devices as they exhibit phenomena including topological electric and spin Hall effects8−10 and microwave magnetoelastic effects11 and they require much lower critical current densities for propagation by spin-transfer torque than other forms of spin textures such as domain walls.6,12,13 In an effort to find materials and feature geometries more amenable to the incorporation of magnetic skyrmions into functional devices, recent work has focused on specially designed metal heterostructures with spatially divergent spin−orbit torque14 and embedding a magnetic vortex in a perpendicular spin environment.15 However, a thorough understanding of the spatial variation of fundamental magnetic parameters in nano/microstructures remains largely unexplored despite the fact that in many classes of materials, but especially complex oxides, functional properties are sensitively correlated to lattice strains.16 This issue will only grow in significance as spin-based devices with increasingly complex spin textures become more commonplace, such that the integration of the magnetic layers with various device components including the substrate, contacts, and isolations regions must be considered. For example, synchrotron X-ray microdiffraction experiments have shown lattice nonuniformities within the strained silicon gate of a metal-oxide semiconductor field-effect-transistor due to the different types of interfaces and surfaces, which ultimately modify the local electron mobility.17 Similar work utilizing coherent X-ray diffraction on individual LiNiO3−δ cathode nanoparticles demonstrated that inhomogeneous strain influences electrochemical performance and is dependent on environment, size, and geometry.18

In order to effectively probe the modification in magnetic behavior when a continuous epitaxial thin film is patterned into micromagnets of varying size and shape, a material with observable properties sensitive to slight perturbations caused by...
the fabrication process must be studied. Complex oxides such as La_{0.7}Sr_{0.3}MnO_3 (LSMO) satisfy this requirement as they offer a wide range of highly tunable functional properties and display strong correlations between the spin, charge, orbital, and lattice degrees of freedom. These factors may also provide a versatile route to control spin textures and magnetic behavior in nano/micro-patterned structures. This is extremely relevant when designing vortex core dynamics for application in future data storage technologies, as the stability and control of such systems are heavily influenced by the magnetic energy landscape.\(^\text{19}\) LSMO is a soft ferromagnetic metal, which displays coincident ferromagnetic/paramagnetic and metal/insulator transitions, a large peak in magnetoresistance (i.e., colossal magnetoresistance, CMR) at its Curie temperature \((T_C)\), and a high-degree of spin-polarization, which is essential for efficient spin-transfer torque applications.\(^\text{20,21}\) In epitaxial thin films, magnetocrystalline anisotropy directly impacts the formation and configuration of spin textures and can be the dominant driving force in stabilizing the magnetic state of spin-based memory devices.\(^\text{2}\) Enabled by a unique patterning technique designed for complex oxides\(^\text{22,23}\) and high-resolution imaging of spin configurations using X-ray photoemission electron microscopy (X-PEEM), we show the evolution of spin textures in complex oxide micromagnets and devise a framework in which the lateral variation of fundamental materials’ parameters, including saturation magnetization and magnetocrystalline anisotropy constants, can be determined as a function of temperature with approximately 0.1 \(\mu\)m spatial resolution.

### RESULTS AND DISCUSSION

Arrays of micromagnets were defined in a 100 unit cell thick epitaxial film of LSMO using a patterning technique based on the local modification of the structural and magnetic order by Ar\(^+\) ion implantation.\(^\text{22,23}\) Three different geometries were patterned: squares with 2 \(\mu\)m long edges oriented along the in-plane \((110)\) substrate directions, squares of the same dimensions with edges along the in-plane \((100)\) substrate directions (referred to as diamonds), and circles with a diameter of 2 \(\mu\)m. Each shape provides a unique perspective on the balance of relevant magnetic energies. For LSMO films grown under tensile strain on \((001)\)-oriented SrTiO\(_3\) (STO) substrates, the magnetic easy axes have been shown to lie along the in-plane \((110)\) directions.\(^\text{24}\) Thus, in the square micromagnets, both shape and magnetocrystalline anisotropy promote the same Landau flux closure structure with moments aligned parallel to the edges, whereas those energy terms are at odds with one another in the diamond micromagnets. Similarly, circular micromagnets typically display a vortex structure characterized by a continuous rotation of moments tangential to the edge at the expense of magnetocrystalline anisotropy energy. The 2 \(\mu\)m wide micromagnets studied here lie in the size regime where new domain walls will not be injected to form more complicated multidomain states but they are still large enough to observe subtle details in domain structure due to magnetocrystalline anisotropy.\(^\text{25,26}\) For micromagnets with smaller dimensions (e.g., 500 nm wide) patterned in LSMO as well as metallic nano- and micromagnets, domain patterns dictated by shape anisotropy were observed irrespective of the shape (i.e., square, diamond, or circle).\(^\text{19,27}\) Theoretically, many types of ferromagnetic materials with a similar balance of saturation magnetization, \(M_S\), and magnetocrystalline anisotropy, \(K\), \((K/M_S \sim −0.01\ T\) for LSMO) should produce comparable magnetic domain structures including metals, other oxides, and Heusler alloys (e.g., Fe, Fe\(_2\)O\(_4\) and Co\(_2\)FeAl); however, it is necessary that they are epitaxial or single crystalline and are characterized by a technique sensitive to the in-plane magnetization (e.g., X-PEEM or spin-polarized low energy electron microscopy).

The magnetic domain patterns (Figure 1) were imaged with high spatial resolution (\(\sim 40\) nm) and as a function of temperature from \(T_C\) to 32 K using X-PEEM at the PEEM endstation on beamline 11.0.1 of the Advanced Light Source. The magnetic contrast in X-PEEM arises due to X-ray magnetic circular dichroism (XMCD) at the Mn \(L_{3,2}\) absorption edges of LSMO, and it scales with the dot product between the local magnetization and the helicity of the X-rays beam (horizontal in Figure 1, oriented along the [110] easy axis).\(^\text{28}\) Thus, domains with the brightest/darkest/intermediate contrast correspond to regions with the magnetic moments aligned antiparallel/parallel/perpendicular to the helicity of the X-rays, respectively. The schematics in Figure 1 highlight the magnetic domain orientation based on the XMCD contrast.

As shown in Figure 1b (left column), at 230 K, the micromagnets display domain patterns dominated by shape anisotropy (i.e., Landau or vortex domain states where the magnetization follows the edges of the structures). For the diamond micromagnets, the two top (bottom) domains are
indistinguishable from one another since they have the same component of magnetization in the direction of the helicity of the X-rays. A vertical stripe of higher/lower intensity through the center of the diamond arises due to the domain wall where the moments are aligned antiparallel/parallel to the X-ray beam, whereas the moments in the two adjacent domains form a 45° angle. This domain structure agrees with previous results from 500 nm wide structures in LSMO and metal alloys with a low magnetocrystalline anisotropy. As the temperature is reduced to 32 K, no change is observed in the domain patterns for the square micromagnets.

For both the diamond and circular micromagnets, the domain patterns change substantially: a clear Landau pattern in the central of the structure can be seen with the magnet domain patterns change substantially: a clear Landau pattern in the perimeter region (see Supporting Information Figure S1). This analysis predicts the existence of these unique spin textures in the form of double closure domains in the diamond and circular micromagnets, whereas the moments in the two adjacent domains form a 45° relative to one another. Similarly for the circular micromagnets, the domain pattern consists of a (110)-oriented Landau pattern surrounded by a thin region with a vortex structure. The derivatives of the domain images (differential images in Figure 1b) highlight the locations of the domain walls associated with the Landau patterns. For these micromagnets, the central Landau pattern gives rise to domain walls forming a central “X”. For the diamond micromagnets, two horizontal spurs emanate from the right/left corners resulting from the outer Landau pattern. A gradual development of the inner Landau pattern can be seen at intermediate temperatures. The existence of these unique spin textures in the form of double concentric flux closure patterns results from the delicate, spatially dependent balance between shape and magnetocrystalline anisotropy energies, and it demonstrates our control of spin textures in complex oxides structures.

To understand the formation of the double concentric flux closure domains in the diamond and circular micromagnets, micromagnetic simulations were performed using MuMax, which takes the sample geometry, as well as values for $M_s$, $K_1$, and exchange stiffness ($A_{ex}$) as inputs to calculate the lowest energy state of a magnetic feature using the Landau–Lifshitz–Gilbert equation. The temperature dependence of $M_s$ was determined for an unpatterned LSMO film grown with the same deposition conditions using a superconducting quantum interference device magnetometer. The intensity difference between the bright and dark domains of the X-PEEM images for the square micromagnets was used to determine the local saturation magnetization of the patterned structures. The experimental domain contrast was found to be homogeneous throughout each domain at each measurement temperature (see Supporting Information Figure S1). This analysis predicts a $T_C \sim 270$ K, comparable to the as-deposited film, confirming that bulk magnetic behavior is retained within the patterned structures. The relationship between $M_s$ and $A_{ex}$ can be described as

$$A_{ex} = \frac{D_s q M_s}{2 g \mu_B}$$

(1)

where $g$ is the Landé factor, $\mu_B$ is the Bohr magneton, and $D_s$ is the spin wave stiffness constant. Temperature-dependent values of $D_s$ for LSMO films on STO substrates, as determined by Golosovsky et al. using magnetic-field-dependent microwave absorption, were used to calculate $A_{ex}$. The remaining unknown is the magnetocrystalline anisotropy constant, $K_1$. Previously, Steenbeck et al. examined the temperature dependence of $K_1$ in 26 nm thick LSMO films grown on STO substrates by torque magnetometry and found a monotonic increase with decreasing temperature (Figure 3). The simulation cell volume of $5 \times 5 \times 40$ nm$^3$ assumes magnetic parameters are constant throughout the depth of the film.

The X-PEEM images for the circular micromagnets were quantitatively compared with micromagnetic simulations as a function of temperature using a metric termed the vortex...
fraction \( f_v \). Assuming a pure vortex structure, the contrast around a circular profile (such as along the blue circle in the inset to Figure 2a) should be sinusoidal as a function of angle. For a pure Landau structure (as in the square micromagnets), the contrast should remain constant within a single domain, and the circular intensity profile will result in a step function with sharp steps every 90° (see Supporting Information Figure S2). The experimental profile obtained from a circle with radius \( r = 0.3 \, \mu m \) (blue circle in the inset to Figure 2a) has abrupt changes in contrast at the domain walls but also displays an intensity that is not constant within each of the four domains (blue curve in Figure 2a). The deviation from the step function of the Landau structure is particularly apparent within the gray domains (highlighted regions in Figure 2a). This behavior indicates the local moments are not perfectly parallel within each magnetic domain but rather are characterized by some degree of rotation akin to a vortex structure. A superposition of a step function (Landau structure) and a sine function (vortex structure) more accurately describes the spin texture of the micromagnets. By subtracting a sine function from the experimental data, the difference (green curve in Figure 2a) has the expected step profile of a Landau pattern. The vortex fraction, \( f_v \), quantifies the relative proportions of Landau and vortex structures as

\[
f_v = \frac{A_{\text{sine}}}{A_{\text{sine}} + A_{\text{step}}}
\]

where \( A_{\text{sine}} \) is the amplitude of the subtracted sine function (vortex component) and \( A_{\text{step}} \) is the amplitude of the remaining step function (Landau component). Figure S4 illustrates fit quality for the raw data in Figure 2a. In addition to the sine and step functions, linear regions are spliced into the fit accounting for rapid changes in contrast due to domain walls but do not influence the amplitude used to calculate \( f_v \). The vortex fraction was extracted as a function of both the radius within the circular micromagnet (Figure 2b) and as a function of temperature. Shape anisotropy dominates at the perimeter of the circular micromagnet, so that \( f_v \) approaches unity at larger radii. The vortex fraction decreases as the radius decreases as the influence of magnetocrystalline anisotropy overcomes that of shape anisotropy and the Landau structure emerges. The magnitude of this decrease is enhanced at lower temperatures. Finally, near the center of the micromagnet, \( f_v \) increases again due to the formation of the vortex core with an out-of-plane component, convoluted with the limitations in the spatial resolution of the PEEM3 microscope.

Micromagnetic simulations also yield the superposition of the Landau and vortex components in the spin texture of the micromagnets, thus allowing \( f_v \) to be used to accurately select the simulation parameters that best correspond with X-PEEM images at a given temperature. A series of simulations with a range of \( K_1 \) values and with varying \( K_1 \) profiles was carried out for each pair of \( M_S \) and \( A_{K_S} \) values as a function of radius for the temperature range from 32–230 K. The simple assumption of a uniform \( K_1 \) profile throughout the micromagnet (green line in Figure 2b) only matches the observed \( f_v \) values in the central region (0.2 \( \mu m < r < 0.4 \, \mu m \)). At larger radius, the simulations underestimate \( f_v \), suggesting that \( K_1 \) decreases toward the perimeter of the micromagnet (see Supporting Information Figure S3). Simulations with a gradually decreasing \( K_1 \) profile also underestimate \( f_v \) at large radius. Of all tested decay models, the full \( f_v \) radial dependence (blue line in Figure 2b) was best represented using a sigmoid function with an abrupt change in \( K_1 \) at a radius = 0.6 \( \mu m \). The X-PEEM and MuMax3 images do not show a corresponding abrupt change in the domain patterns at this radius due to the fact that the exchange energy works to smooth out any sharp changes in local magnetization. These result highlight the fact that spatial variation in all fundamental magnetic parameters must be accounted for to accurately model the magnetic domains in patterned media, as \( K_1 \) values at the inner region of the micromagnet directly affect the domain patterns of the outer region, and vice versa. The different radial dependencies of \( M_S \) and \( K_1 \) are surprising, however, it has been reported that substrate-induced strain modifies the \( T_C \) and low temperature \( K_1 \) values of an epitaxial LSMO film. In addition, Thiele et al. studied the relationship between strain-induced changes in \( T_C \) and magnetization and discovered that though the \( T_C \) may change with strain, changes in saturation field and \( M_S \) at low temperatures are negligible. Therefore, it is possible that at low temperatures, \( K_1 \) is reduced by strain from the surrounding matrix, whereas \( M_S \) remains unchanged.

The temperature dependence of \( K_1 \) was determined assuming both a uniform and abruptly varying \( K_1 \) profiles as shown in Figure 3. For the uniform \( K_1 \) profile, the \( K_1 \) values were determined as the average value in the central region (0.2 \( \mu m < r < 0.4 \, \mu m \)), while for the abruptly varying \( K_1 \) profile, the maximum value of the sigmoidal function was used. Both calculated values display a similar trend with temperature as those presented by Steenbeck et al.3 Because the LSMO thin films studied by Steenbeck et al. had higher \( M_S \) and \( T_C \) values, the data is plotted on normalized axes in the inset to Figure 3, which confirms that all three curves possess analogous temperature dependence. In order to illustrate that the temperature-dependent \( K_1 \) values extracted from the X-PEEM images are the essential parameters for the determination of the experimentally observed spin textures, micromagnetic simulations of the square, diamond, and circular micromagnets are...
shown in Figure 4 for the three temperatures from Figure 1. Excellent agreement exists between the two sets of images.

![Figure 4. Simulated magnetic domain images. Simulated magnetic domain images at the same temperatures from Figure 1 and associated differential images (T = 32 K) using the calculated values of $K_1$.](image)

METHODS/EXPERIMENTAL

A 100 unit-cell-thick LSMO thin film was grown epitaxially by pulsed laser deposition on a (001)-oriented Nb-doped (0.05 wt % Nb) STO substrate. During growth, the substrate temperature was held at 680 °C, and the oxygen pressure was 2.0 × 10⁻¹ mbar. A KrF excimer laser was operated at fluence of 2.0 J/cm² and 1 Hz repetition rate. The growth was monitored with in situ reflection high-energy electron diffraction, and unit-cell intensity oscillations of the specular reflection were observed throughout the growth. Structural analysis was performed with X-ray diffraction and X-ray reflectivity using a four-circle Bruker D8 Discover to verify film crystallinity, epitaxial nature, and thickness. Microstructures (~100 of each geometry) were defined using a patterning technique based on local modification of the structural and magnetic order by Ar⁺ ion implantation through a Cr hard mask. Scanning electron microscopy was performed at room temperature to verify the shape and size of the nanostructures. X-PEEM images were acquired using right and left circularly polarized (RCP and LCP, respectively) X-rays at a photon energy corresponding to the maximum XMCD signal at the Mn $L_2$, edge of the film. The domain images were obtained using an asymmetry equation, $I = (I_{RCP} - I_{LCP})/(I_{RCP} + I_{LCP})$, which removes contributions from topography and work function differences from the individual images. The X-ray beam was incident along the [110] direction at 30° from the sample surface, and the PEEM3 chamber was shielded from external magnetic fields so that all images correspond to the demagnetized state. K$_f$ error bars are derived from variations in the experimental $f_z$ value obtained from four individual micromagnets taken with high resolution images. X-PEEM image contrast is only sensitive to the in-plane magnetization component parallel to the incident beam, but MuMax³ simulations output a complete vector field. The images generated by MuMax³ shown and studied here are obtained by using only the $x$ component of the vector field, which is analogous to the X-PEEM measurements. A full listing of simulation parameters can be found in Table S1 of the Supporting Information.

ASSOCIATED CONTENT

## Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsnano.6b03770. Supporting Information describing radial dependence of magnetization within an individual micromagnet (Figure S1), a full spectrum of radial profiles (Figure S2), further detail of magnetocrystalline anisotropy constants (Figure...
S3), an example fit of circular profile data (Figure S4), and a complete list of the simulation parameters used (Table S1) are presented. (PDF)

AUTHOR INFORMATION
Corresponding Author
*E-mail: ytkamura@ucdavis.edu. Phone: 530-754-7124. Fax: 530-752-1031. Address: Department of Materials Science and Engineering University of California—Davis, One Shields Avenue, Davis, CA 95616, United States.

Author Contributions
1M.S.L. and T.A.W. are co-first authors. Y.T., E.F., and J.K.G. designed the experiment. S.T.R. performed the electron-beam lithography. M.S.L, T.A.W, R.V.C., A.S., and A.T.Y. carried out the X-PEEM measurements. M.S.L. and T.A.W. analyzed the data. M.S.L, T.A.W., and Y.T. cowrote the manuscript.

Notes
The authors declare no competing financial interest.

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