Hard x-ray magnetic circular dichroism study of a surface-driven twisted state in Gd/Fe multilayers

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By tuning the incidence angle of hard, circularly polarized x-rays with respect to the sample surface, we are able to independently probe surface and bulk magnetic states in a Fe(35 Å)[Gd(50 Å)/Fe(35 Å)]$_{15}$ ferrimagnetic multilayer by magnetic circular dichroism (MCD). We show that a twisted magnetic state nucleates at the surface of the multilayer at $T \approx 0.7\ T_{\mathrm{comp}}$, where T_{comp} is the ferrimagnetic compensation temperature. At nucleation, the twist penetrates $\approx$170 Å or two bilayers. Despite its theoretical prediction by LePage and Camley more than 15 years ago [J. G. LePage and R. E. Camley, Phys. Rev. Lett. **65**, 1152 (1990); R. E. Camley, Phys. Rev. B **35**, 3608 (1987)], this surface-twisted state has eluded direct experimental detection up to now. Its clear observation here demonstrates the power of grazing incidence, hard x-ray, MCD measurements for studies of magnetic phase transitions in layered structures. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1543875]

I. INTRODUCTION

The ability to separate surface and bulk magnetic states in a measurement is of paramount importance in obtaining a complete understanding of the magnetic properties of the system under study. The spin-flop transition in antiferromagnetically coupled Fe/Cr superlattices, where the spatial homogeneity of the magnetic structure depends on surface termination,[1] is an example of the need for spatially resolved magnetic information in layered nanostructures. Another such example is Fe/Gd ferrimagnetic multilayers, which exhibit antiferromagnetc coupling at the Fe/Gd interfaces and are predicted to exhibit a spatially inhomogeneous magnetic state near the compensation temperature T_{comp}. The prediction by LePage and Camley[2] that a surface-twisted state nucleates in Fe/Gd much before such twist is stabilized in the bulk has eluded experimental detection for 15 years. Unambiguous detection of such a phase requires not only showing the existence of a surface twist, but also showing the absence of such twist in the bulk. Most techniques exclusively probe the surface or the bulk, so a determination of surface and bulk magnetic states cannot be done simultaneously. Here we show that, by tuning the x-rays' incidence angle to near and above the critical angle for total external reflection from the multilayer structure, this separation becomes possible, allowing the direct observation of nucleation of a twisted state at the surface of the multilayer.

II. EXPERIMENTAL PROCEDURE

The multilayers used in this study were sputtered in vacuum onto Si substrates using Nb buffer (100 Å) and cap (30 Å) layers. Superconducting quantum interference device (SQUID) magnetometry shows, and magnetic circular dichroism (MCD) measurements confirm, that the multilayers couple antiferromagnetically at the Gd/Fe interfaces and have coercive fields <50 G at 300 K. X-ray measurements were performed at sector 4 of the Advanced Photon Source at Argonne National Laboratory. Undulator radiation was monochromatized with double Si(111) crystals, and its polarization converted from linear to circular with a diamond (111) quarter-wave plate operated in Bragg transmission geometry.[3] A closed-cycle He refrigerator was used for the temperature-dependent measurements. The sample was placed between the pole pieces of an electromagnet providing a variable field in the range of ± 600 G parallel to the sample's surface. The experimental setup allows for simultaneous measurements of specular reflectivity and MCD in fluorescence geometry. We used the position of the first multilayer Bragg peak to calibrate the grazing incidence angle, θ_i. This is important, since the x-rays' penetration varies rapidly for $\theta_i \gtrsim \theta_c$, where θ_c is the critical angle for total external reflection.

Optimal resonant energies for Gd- and Fe-specific hysteresis loops are those that maximize the asymmetry ratio $|(\mu^+ - \mu^-)/(\mu^+ + \mu^-)|$, where $\mu^{+,-}$ are absorption coefficients for incoming left and right circularly polarized x-rays, respectively. These were found by scanning the incident x-ray energy around Gd L_2 ($2p_{1/2}$) and Fe K ($1s$) absorption edges, while switching the helicity of the incident radiation at each energy point. Element-specific hystereses were measured at these optimal energies (7.930 and 7.110 keV for Gd and Fe, respectively) and at selected incident angles by switching the x-rays' helicity at each applied field value. The grazing incidence angle used to enhance surface sensitivity in the measurements was $\theta_i = 0.43°$. As shown in Fig. 1, this measurement provides surface sensitivity at the Gd reso-

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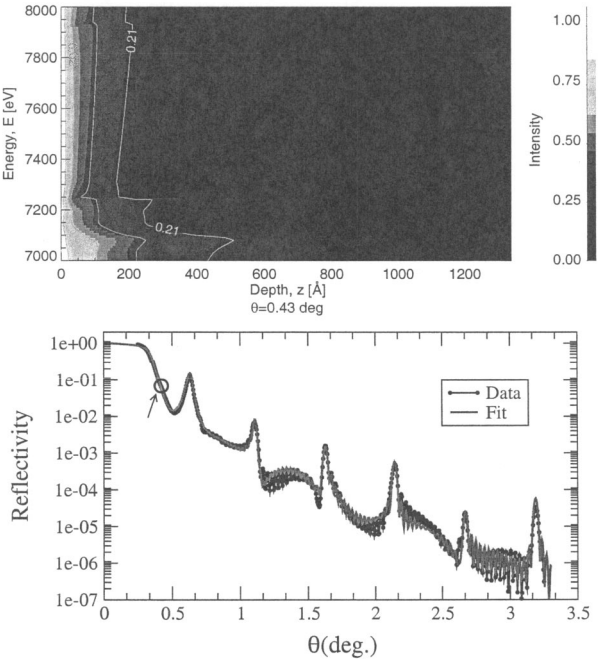

FIG. 1. Top: x-ray intensity depth profile within the multilayer as a function of incident energy calculated at $\theta_i = 0.43°$ grazing incidence angle. Bottom: specular reflectivity data together with corresponding Parratt fit (See Ref. 12). The critical angle for total external reflection is $\theta_c \sim 0.35°$, and the arrow indicates the $\theta_i = 0.43°$ angle used for surface-sensitive measurements.

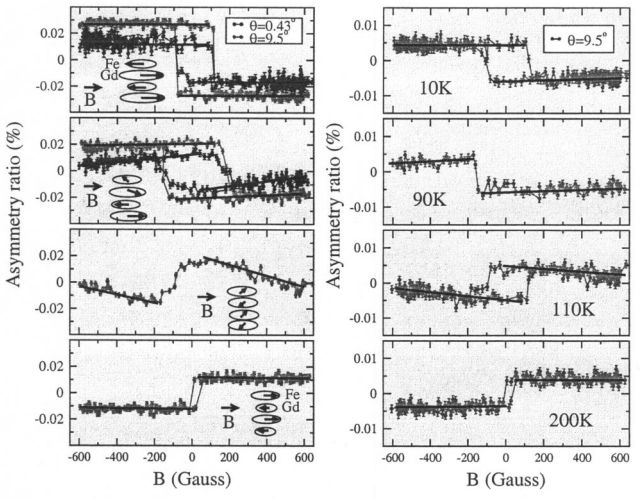

FIG. 2. Element-specific MCD hysteresis loops measured near Gd L_2 ($E = 7.930$ keV, left) and Fe K ($E = 7.110$ keV, right) resonances. The $\theta = 0.43°$ geometry probes the near-surface magnetization state while the whole multilayer is probed at $\theta = 9.5°$. Clear correlated behavior of Gd and Fe layers is seen due to their strong interlayer coupling (see Ref. 10). Surface-sensitive loops are scaled down for clarity. Insets show a schematic representation of the magnetization direction and magnitude in the top and inner parts of the multilayer.

nance energy (probes top 1–2 bilayers), while this sensitivity is greatly diminished at the Fe resonance (probes 4–5 bilayers).[4] The latter is due to relatively low absorption in Gd layers (below Gd $L_{2,3}$ absorption edges) and Fe layers (optimal Fe energy is near the bottom of Fe K absorption edge). Further decreases in incidence angle compromise the data quality as intensity is transferred from absorption to scattering channels degrading the fluorescence signal-to-noise ratio. Bulk sensitive measurements were done at $\theta_i = 9.5°$, where the whole multilayer structure is probed.

III. RESULTS AND DISCUSSION

Figure 2 shows Gd and Fe hysteresis loops for selected temperatures below, near and above T_{comp}. Both surface sensitive ($\theta_i = 0.43°$) and bulk-sensitive ($\theta_i = 9.5°$) loops are shown for Gd, while only bulk-sensitive loops are shown for Fe [we found no significant differences between surface and bulk loops at the Fe resonance, as expected from the much larger probing depth at this energy (see Fig. 1)]. Since MCD is proportional to the projection of the magnetization along the photon wave vector (the latter nearly coincides with the applied field direction), a ''flat'' hysteresis loop in Fig. 2 indicates this projection remains unchanged as a function of applied field. This is the situation at T=10K, where the Gd magnetization dominates the Zeeman energy and aligns with the applied field, while Fe is constrained antiparallel by interlayer exchange. At $T = 90$ K, increasingly ''tilted'' Gd loops are measured in the top part of the multilayer ($\theta = 0.43°$), while bulk-sensitive Gd and Fe loops show related but largely diminished tilting. Tilting indicates a decrease in the projection of the moment along the field direction with increased applied field, that is, a gradual twist of the probed magnetization away from the applied field direction. Self-consistent analysis of surface- and bulk-sensitive loops shows that, at this temperature, a twist occurs in the top (and bottom, by symmetry) two bilayers approximately, while the interior of the multilayer remains in a nearly collinear state. At $T = 110$ K, the tilting or twist already propagates throughout the multilayer, as evidenced in the now significantly tilted, bulk-sensitive Gd and Fe loops. A correlated reversal in the sign of Gd and Fe loops at this temperature indicates that the Fe magnetization now dominates the Zeeman energy contribution with its net component, averaged over depth, having a positive projection along the field, while that of Gd has a negative projection. At $T = 200$ K the loops are again flat, with the Fe magnetization aligning along the field and Gd constrained antiparallel.

We note that Gd L_3 and Fe K MCD signals have opposite signs for antiparallel alignment of Gd and Fe magnetizations, while same sign MCD signals are measured here at Gd L_2 and Fe K resonances. This is due to opposite signs of spin-orbit coupling at the $2p_{1/2}$ and $2p_{3/2}$ core levels involved in Gd L_2 and L_3 resonances, respectively.

The different regimes observed in the hysteresis loops of Fig. 2 can be classified, following Camley and Tilley,[5] as Gd-aligned, surface-twisted, bulk-twisted, and Fe-aligned phases. These phases are predicted to emerge as a result of a delicate balance between exchange and Zeeman energies. Experimental evidence supporting the existence of Gd(Fe)-aligned and bulk twisted phases has been reported using a variety of techniques, including magnetization measurements,[6] polarized neutron reflectometry,[7] Mössbauer spectroscopy,[8] and MCD measurements in transmission geometry.[9] The surface-twisted phase, however, has eluded

direct experimental detection since its prediction by LePage and Camley,[2] due mostly to the difficulty in probing surface and bulk states simultaneously. Proving the existence of a surface-twist phase requires proving the absence of such a twist in the bulk. The loops in Fig. 2 clearly show that grazing and larger incidence angle MCD measurements in fluorescence geometry can unambiguously detect the presence of all postulated phases, including the surface-twisted phase.

Deviation from a collinear, antiparallel arrangement of Gd and Fe magnetizations along the applied field direction can only be driven by a Zeeman energy gain, as exchange energy is already minimized in the collinear arrangement. A net gain in the *difference* between the Gd and Fe projected magnetizations along the field direction has to take place for a twist to occur. Clearly, for $M_{\mathrm{Gd}} > M_{\mathrm{Fe}}$, this requirement can only be satisfied if $\theta_{\mathrm{Fe}} > \theta_{\mathrm{Gd}}$, that is, the ''minority'' sublattice has to twist more than the ''majority'' sublattice in order to compensate for the Zeeman energy loss in the latter. The different twist angles, however, result in increased exchange energy, either due to deviations from antiferromagnetic alignment at the Gd/Fe interfaces[10] or deviations from ferromagnetic alignment within the Fe and Gd layers themselves.[5] It is the competition between this increased exchange energy and the Zeeman energy gain that determines the magnetic ground state.

At $T=10$ K, $M_{\mathrm{Gd}} \approx 1.5 M_{\mathrm{Fe}}$[11] and a twist would be too costly in exchange, favoring a Gd-aligned phase. At $T=90$ K, the spontaneous Gd magnetization has decreased by $\approx 25\%$ (as seen from the decreased Gd-hysteresis jump), and now $M_{\mathrm{Gd}} \approx 1.18 M_{\mathrm{Fe}}$. The data indicate that under this condition, the magnetization twists away from the field direction only in the top (and, by symmetry, bottom) parts of the multilayer, while in the interior it remains nearly aligned with the field. The stabilization of a surface twist at a lower temperature (i.e., larger $|M_{\mathrm{Gd}}|$) than its bulk counterpart shows that the transition from aligned to twisted states nucleates at the surface in an Fe-terminated multilayer. The energy barrier for a twist is decreased at the surface due to the absence of Gd/Fe interlayer exchange on end sides of top (bottom) Fe layers allowing for a larger average twist angle in these layers with a resultant lower exchange price than in their bulk counterparts. At $T=110$ K, $M_{\mathrm{Gd}} \approx 0.99 M_{\mathrm{Fe}}$, and the twist penetrates throughout the multilayer. In this case, the nearly identical sublattice magnetizations allow for a net Zeeman energy gain at almost equal (and opposite) twists of the two sublattice magnetizations. The low exchange energy cost of the resulting nearly collinear magnetic structure stabilizes the twisted state throughout the entire multilayer. At $T=200$ K, $M_{\mathrm{Fe}} \approx 1.6 M_{\mathrm{Gd}}$, and the roles of Gd and Fe are nearly reversed compared to those at $T=10$ K. The dominant Fe magnetization stabilizes an aligned phase with the Fe magnetization along the applied field and Gd constrained antiparallel.

IV. SUMMARY AND CONCLUSIONS

In summary, we provide clear experimental evidence showing the surface nucleation of a twisted magnetization state in a ferrimagnetic multilayer. Simultaneous measurements of surface- and bulk-sensitive element-specific hysteresis loops show that surface nucleation occurs at $T \approx 0.7 T_{\mathrm{comp}}$, and that the twist propagates throughout the bulk at $T \sim T_{\mathrm{comp}}$. At nucleation, the surface twist penetrates approximately two bilayers or ≈ 170 Å, for $B < \sim 600$ G. The results demonstrate the ability of grazing incidence MCD in fluorescence geometry to study complex layered magnetic systems near instabilities in their phase diagrams.

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[4] The angle between the refracted beam and the sample's surface is given by $\alpha_r = \sqrt{\alpha_i^2 - \theta_c^2}$, which for $\alpha_i = 0.43°$ and $\theta_c = 0.35°$ is $\alpha_r = 0.25°$. The sample's absorption lengths at 7930 and 7110 eV are 3 μm and 8 μm, respectively (using experimentally determined optical constants for Gd and Fe). This gives penetration depths of 131 Å (1.5 bilayers) and 350 Å (4.1 bilayers) at Gd L_2 and Fe K resonances, respectively. Accurate calculations of depth-resolved electric field intensity profiles using Parratt's recursive formalism and experimental Gd and Fe optical constants yield nearly identical depths for a $1/e$ decrease in incident intensity.

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