Nature of inhomogeneous magnetic state in artificial Fe/Gd ferrimagnetic multilayers


1Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois 60439, USA
2Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA

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The low-field surface nucleation and evolution of the inhomogeneous magnetic state in strongly coupled Fe/Gd ferrimagnetic multilayers is measured via grazing-incidence x-ray magnetic circular dichroism. At \( T \approx 0.7 T_0 \), where \( T_0 \approx 110 \) K is the ferrimagnetic compensation temperature, the inhomogeneous state nucleates at the surface. At nucleation, the surface state extends tens of interatomic distances into the bulk (\( \sim 200 \) Å), a direct consequence of the strong interlayer coupling. At \( T = T_0 \), the inhomogeneous state penetrates throughout the bulk, while homogeneous magnetic states occur far below and above \( T_0 \). Surface termination has a dramatic effect on the nature of the inhomogeneous state.

Understanding the physical interactions that lead to spatially inhomogeneous magnetic ground states remains a challenge for theorists and experimentalists alike. In this respect artificial magnetic nanostructures have proven to be unique model systems, resulting in significant advances in this area. For example, symmetry breaking at interfaces of Fe/Cr artificial antiferromagnets and biquadratic coupling in exchange Fe/SmCo soft/hard ferromagnets induce novel inhomogeneous ground states. \(^4 \) The role of broken symmetry at surfaces and interfaces as a source for chiral effects in magnetic nanostructures has recently been investigated theoretically. \(^3 \)

Fe/Gd artificial ferrimagnets have been used as model systems in several investigations. \(^4,5 \) The markedly different bulk Curie temperatures of Fe (1024 K) and Gd (293 K), together with strong interlayer antiferromagnetic coupling at the Fe/Gd interface, result in a rich phase diagram with magnetic configurations that strongly depend on applied field and temperature. \(^4 \) Although experimental evidence for a magnetic inhomogeneous state has been gathered by several techniques, \(^7 \) a long-standing open question on the mechanism of nucleation of such state has not been resolved experimentally until now. Over a decade ago LePage and Camley \(^6 \) predicted that a phase transition into an inhomogeneous magnetic state would nucleate at the surface of a strongly coupled artificial ferrimagnetic multilayer if the multilayer is terminated by the minority magnetic component. In this “surface-twisted” phase, the magnetization deviates from the applied field direction near the surface while the bulk remains field aligned. This inhomogeneous phase has eluded direct experimental detection due to the difficulty in probing surface and bulk states in the same measurement. The challenge is to observe both the existence of a surface-twisted phase and the absence of a bulk twist. Inhomogeneous magnetic depth profiles in layered nanostructures are commonly probed by scattering techniques (reciprocal space) such as neutron reflectometry and x-ray resonant magnetic scattering. \(^8 \) Although these are powerful probes of magnetism, the measurement of the scattered intensity often results in model-dependent magnetization profiles. In addition, Gd is a strong neutron absorber and, therefore, bulk sensitivity is reduced in neutron measurements.

In this Communication, we report direct, real-space, measurements of the low-field nucleation of an inhomogeneous magnetic state at the surface of a Fe-terminated \([\text{Fe}(35 \text{ Å})/\text{Gd}(50 \text{ Å})]_{15}/\text{Fe}(35 \text{ Å})\) ferrimagnetic multilayer. We exploit the penetration depth tunability of x rays at grazing- and larger-incidence angles to alternately probe the surface and the bulk magnetic states by x-ray magnetic circular dichroism (XMCD). \(^9 \) At \( T = 0.7 T_0 \), where \( T_0 \approx 110 \) K is the ferrimagnetic compensation temperature at which the sublattice magnetizations are equal but opposite, the magnetic twist nucleates at the surface but penetrates \( \approx 200 \) Å (two to three bilayers) into the bulk as a result of strong Fe/Gd interlayer exchange coupling. The reduction in Zeeman energy as \( T \) is approached from below causes the inhomogeneous state to propagate throughout the bulk. Surface nucleation is not observed for a Gd-terminated multilayer showing that surface termination plays a crucial role in defining the magnetic ground state. The results are in excellent agreement with theoretical predictions and help elucidate the driving forces behind inhomogeneous magnetic state nucleation in artificial ferrimagnets.

The multilayers were sputtered onto Si substrates using Nb buffer (100 Å) and cap (30 Å) layers. Superconducting quantum interference device (SQUID) magnetometry shows, and XMCD measurements confirm, that the multilayers couple antiferromagnetically at the Fe/Gd interfaces and have coercive fields \( < 50 \text{ Oe} \) 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. \(^10 \) A closed-cycle He refrigerator, mounted on the \( \phi \) circle of a diffractometer, was used for the temperature-dependent measurements. The sample was placed between the pole pieces of an electromagnet delivering magnetic fields in the range of \( \pm 600 \text{ Oe} \) in a direction parallel to the sample’s surface and nearly coincident with the x-ray wave vector.

The experimental setup permits simultaneous measurements of specular reflectivity and XMCD in fluorescence geometry. Figure 1 shows the sum \( \mu_{\phi} + \mu_{\phi} \) and asymmetry
ratio \( \frac{\mu^+ - \mu^-}{\mu^+ + \mu^-} \) of fluorescence signals, measured near the Gd \( L_2 \) and Fe \( K \) absorption edges (+ and − denote opposite helicites of circularly polarized x rays). While the sum is proportional to the sample’s absorption coefficient, the asymmetry ratio is proportional to the element-specific magnetization. Element-specific hysteresis loops were then measured at the resonant energies for which the magnetic contrast is the largest (7.93 and 7.11 keV for Gd and Fe, respectively) by reversing the x-ray helicity at each applied field value and measuring the asymmetry ratio.

The specularly reflected signal was used to accurately determine the x-ray incidence angle. This is important since the penetration depth varies rapidly for \( \theta_i = 0.43^\circ \), is slightly above the critical angle \( \theta_c \approx 0.35^\circ \) (see Ref. 11). Further decreasing the incidence angle towards \( \theta_c \) was not practical, since the Nb cap and Fe layers (above Gd) and Nb cap layer (above Fe) transfer intensity from absorption to scattering channels degrading the fluorescence signal-to-noise ratio. Despite these structural constraints, which are typical in magnetic heterostructures, we were able to retrieve surface-enhanced magnetic information from Gd layers. At the Fe resonance, surface sensitivity was reduced due to smaller absorption in Gd layers (below Gd \( L_{2,3} \) edges) and Fe layers (resonance occurs near the bottom of the Fe absorption edge) (Fig. 1). A Parratt fit to the specular reflectivity data (modified to include roughness) confirmed the nominal structural parameters of the multilayer.

Gd and Fe hysteresis loops for selected temperatures below, near, and above \( T_0 \) in a Fe-terminated multilayer are shown in Fig. 2 (left). For the Gd loops, at each temperature, two sets of data are shown corresponding to surface-enhanced loops at \( \theta_i = 0.43^\circ \) (probes approximately two bilayers) and bulk-sensitive loops at \( \theta_i = 9.5^\circ \) (probes the whole multilayer). We did not detect significant differences between Fe loops at these angles due to the diminished surface sensitivity at the Fe resonance and the larger error bars associated with the smaller dichroic signal, so we only show bulk-sensitive Fe loops. Since XMCD is proportional to the projection of the magnetization along the x-ray wave vector, a “flat” hysteresis loop in Fig. 2 indicates that this projection remains unchanged as a function of \( H \). This is the situation at 10 K, where Gd dominates the Zeeman energy and aligns with \( H \), while Fe is constrained antiparallel by interlayer exchange. At 70 and 90 K, increasingly “tilted” Gd loops are measured at the top part of the multilayer, while bulk-sensitive Gd loops show less tilting. Tilting indicates a de-
crease in the projection of the moment along $H$; i.e., a
gradual twist of the probed magnetization away from
the field direction. At 110 K, the tilting or twist already propa-
gates throughout the multilayer, as evidenced from the now
significantly tilted bulk 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 con-
tribution with its net component, averaged over depth, hav-
ing a positive projection along the field. At 200 K, the loops
are again flat, with the Fe aligning along the field and Gd anti-
parallel to it.

The hysteresis loops of Fig. 2 can be classified, following
Camley, as (a) Gd-aligned, (b) surface-twisted, (c) bulk-
twisted, and (d) Fe-aligned phases. These phases are pre-
dicted to emerge as a result of a delicate balance between
intralayer and interlayer exchange and Zeeman energies. A
deviation from a collinear, antiparallel arrangement of the Gd
and Fe magnetizations along $H$ can only be driven by a re-
duction in Zeeman energy, since the exchange energy is al-
ready minimized in the collinear arrangement. In order to be
energetically favorable, a twist configuration has to satisfy
the Zeeman energy “boundary” $M_{\text{Fe}} \cos \alpha_{\text{Gd}} - M_{\text{Fe}} \cos \alpha_{\text{Gd}} > |M_{\text{Gd}}(T) - M_{\text{Fe}}|$, where $\alpha_{\text{Gd}}, \alpha_{\text{Fe}}$ are depth-
averaged twist angles. A gain in the difference between the
projected magnetizations along $H$ has to take place for a twist to occur. Here, the $T$ dependence in the magnitude of the Gd magnetization is explicitly noted, while that of Fe is neglected. For $M_{\text{Gd}} > M_{\text{Fe}}$, this can only be satisfied if $\alpha_{\text{Fe}} > \alpha_{\text{Gd}}$; i.e., the “minority” sublattice has to twist more than the “majority” sublattice in order to compensate for the in-
creased Zeeman energy of the latter. These different twist
angles, however, result in an increased exchange energy, ei-
ther due to deviations from antiferromagnetic alignment at
the Fe/Gd interfaces or deviations from ferromagnetic align-
ment within the Fe and Gd layers themselves. It is the com-
petition between this increased exchange energy and the re-
duction in Zeeman energy that determines the magnetic
ground state.

At 10 K, $M_{\text{Gd}} \approx 1.55M_{\text{Fe}}$, and a twist would be too
 costly in exchange, favoring a Gd-aligned phase. At 70 and
90 K, the spontaneous Gd magnetization is lowered by
$\sim 20\text{--}25\%$, as seen from the reduced Gd hysteresis jump,
resulting in $M_{\text{Gd}} \approx 1.20(4)M_{\text{Fe}}$. Under these conditions, the magnetization twists away from $H$ only at the top (and bot-
tom) of the multilayer, while the interior remains field
aligned. For example, at 90 K the Gd surface-sensitive
XMCD is reduced by $\sim 65\%$ at $H=600$ Oe, while the bulk
XMCD decreases only by $\sim 20\%$. Considering the probing
depth of approximately two bilayers at $\theta_i=0.43\degree$, and
given that top and bottom parts of the multilayer are equiv-
lent, the average reduced magnetization $m$ in the inner $\sim 11$
bilayers can be obtained from $m = 0.96$; i.e., the interior of the multilayer re-
mains mostly field aligned. The reduction in the bulk XMCD
at 70 and 90 K is mainly due to the surface contribution.

It is instructive to compare the experimental results with
theoretical calculations of the static magnetization profile.
The Landau-Lifshitz equation of motion with energy dissipation
is an efficient and robust energy minimization scheme
that is very often used in micromagnetic calculations.\textsuperscript{15} We
implemented this approach to obtain static solutions of the
one-dimensional spin chain, as described in Ref. 16. The
chain consisted of 262 sites as the Fe and Gd layers were
artificially divided into seven and ten sublayers, respectively.
Ferromagnetic exchange coupling in Fe and Gd layers were
scaled to their bulk $T_C$, $J_{\text{Gd}}=0.28J_{\text{Fe}}$, while interlayer an-
tiferromagnetic exchange was set to $-\langle J_{\text{Fe}}+J_{\text{Gd}}\rangle/2 = -0.64J_{\text{Fe}}$ (alloy mean-field calculations\textsuperscript{3} yield
$-0.706J_{\text{Fe}}$). The saturation magnetization $M_s(T)$ in Gd lay-
ers was independently obtained from integrated areas of
angle-dependent XMCD measurements and $T$-dependent
hysteresis jumps. Results from these calculations are shown
in the right panel of Fig. 2. The surface nucleation of the
inhomogeneous state is clearly observed. To compare with the
experimental data, the calculated magnetization depth
profiles were weighted, each element separately, to account
for the depth selectivity of our XMCD measurements at
the different incidence angles, $\langle M \rangle = \sum_{n} M_{n}(H) W_{n}/\sum_{n} W_{n}$,
where $W_{n} = e^{-\mu_{z_{n}}/\sin \theta}$ and $\mu_{z_{n}} = \mu_{z_{n}}$, i.e., the com-
pound absorption coefficient at depth $z_{n}$ (for surface-
sensitive averaging we used the refracted angle $\theta_i=0.25\degree$,
see Ref. 12). The results of this averaging are shown by solid
lines in the left panel of Fig. 2, where the agreement with ex-
periment supports the conclusion of the extent of the pen-
etration depth at nucleation. This is an important result, as
previous attempts to determine the penetration depth placed
it at several thousand angstroms.\textsuperscript{17}

The energy barrier for a twist of the minority sublattice
(Fe) towards the applied field direction $H$ is decreased at the
surface due to the absence of Fe/Gd interlayer exchange cou-
pling at the end sides of terminal Fe layers. This results in
surface nucleation of the inhomogeneous state, while the in-
creased exchange energy cost in the bulk does not allow the
 twist to penetrate past the first few Fe/Gd bilayers. We had
mentioned that for $M_{\text{Gd}} > M_{\text{Fe}}$, the minority sublattice has to
twist more toward $H$ than the majority sublattice twists away
from it. This is seen in the data by the larger XMCD reduc-
tion at nucleation. This is an important result, as the ex-
pected suppression of the penetration depth placed it at several
thousand angstroms.

At 110 K, $M_{\text{Gd}} \approx 0.98M_{\text{Fe}}$ and the twist penetrates
throughout the multilayer. Here, the nearly identical sublat-
tice magnetizations allow for the Zeeman energy boundary
inequality to be satisfied at similar (and opposite) twists of
the two sublattice magnetizations. This is seen by the similar
XMCD reduction, with $H$, in Fe and Gd bulk loops at 110 K.
The low exchange-energy cost of this locally more collinear
structure stabilizes the twisted state throughout the entire
multilayer. Note that both data and simulations show that, at
this $T$, the interior of the multilayer twists more than the
surface. This is because the Fe layers already dominate the
magnetization and favor an aligned structure; the twist is

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|}
\hline
$H$ (Oe) & $M_{\text{Gd}}$ & $M_{\text{Fe}}$ \\
\hline
0 & 1.55 & 1.20 \\
600 & 1.35 & 1.18 \\
\hline
\end{tabular}
\caption{Comparison of calculated and experimental magnetization.
}\end{table}
now driven by the Gd minority layers. At 200 K, $M_{Fe} \approx 1.6 M_{Gd}$, and the roles of Gd and Fe are reversed compared to those at 10 K. The dominant Fe magnetization aligns with $H$, Gd being constrained antiparallel by the interlayer exchange.

The effect of surface termination upon nucleation of the inhomogeneous state was further studied on a Gd-terminated multilayer with identical composition, i.e., Gd(50 Å)/Fe(35 Å)Gd(50 Å)$_{15}$. Here, we find no evidence for nucleation of a surface-twisted phase below $T_0$. Although increasing the magnetization component of the minority sublattice (Fe) along $H$ remains the driving force for nucleation of a twist, the energy advantage encountered at the surfaces of the Fe-terminated multilayer is no longer present. Thus the phase transition into a twisted state is a bulk phenomenon which, at low fields, is confined to the vicinity of $T_0$.

In summary, we directly probed, in real space, the nucleation and evolution of an inhomogeneous magnetic state in a model artificial ferrimagnetic system with strong interlayer coupling. Nucleation occurs at the surface only when the multilayer is terminated by the minority (Fe) component. The surface state penetrates $\approx 200$ Å into the bulk due to strong interlayer coupling at Fe/Gd interfaces. Our results are the first direct confirmation of the long-ago predicted inhomogeneous magnetic phase in the strongly coupled model system. Furthermore, our method opens a way towards distinguishing surface from bulk states in inhomogeneous magnetic systems.

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