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Domain size and structure in exchange coupled [Co/Pt]/NiO/[Co/Pt] multilayers

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Abstract

We investigate the competing effects of interlayer exchange coupling and magnetostatic coupling in the magnetic heterostructure ([Co/Pt]/NiO/[Co/Pt]) with perpendicular magnetic anisotropy (PMA). This particular heterostructure is unique among coupled materials with PMA in directly exhibiting both ferromagnetic and antiferromagnetic coupling, oscillating between the two as a function of spacer layer thickness. By systematically tuning the coupling interactions via a wedge-shaped NiO spacer layer, we explore the energetics that dictate magnetic domain formation using high resolution magnetic force microscopy coupled with the magneto-optical Kerr effect. This technique probes the microscopic and macroscopic magnetic behavior as a continuous function of thickness and the interlayer exchange coupling, including the regions where interlayer coupling goes through zero. We see significant changes in domain structure based on the sign of coupling, and also show that magnetic domain size is directly related to the magnitude of the interlayer exchange coupling energy, which generally dominates over the magnetostatic interactions. When magnetostatic interactions become comparable to the interlayer exchange coupling, a delicate interplay between the differing energy contributions is apparent and energy scales are extracted. The results are of intense interest to the magnetic recording industry and also illustrate a relatively new avenue of undiscovered physics, primarily dealing with the delicate balance of energies in the formation of magnetic domains for coupled systems with PMA, defining limits on domain size as well as the interplay between roughness, domains and magnetic coupling.
oscillatory coupling that has been observed in FM/AFM/FM heterostructures with PMA arises from exchange coupling at the FM/AFM interfaces, resulting in an out-of-plane canting of Ni spins within the NiO layer that is propagated across the AFM spacer layer via the AFM exchange [2, 3]. In this case, the coupling oscillates with the period of the AFM ordering, transitioning from FM to AFM with each additional monolayer of the AFM thin film.

The question addressed in this paper regards the behavior of coupling as the thickness of NiO is varied between an odd (favors FM ordering) and an even number of layers (favors AFM ordering) [8]. In addition to answering this fundamental question, the results are directly relevant to magnetic recording technology as higher areal densities require the use of spin valves and magnetic junctions with strong PMA. The interplay between coupling, domain size, roughness and the role of the PMA sets limits on the ultimate feature size (i.e. bit element or read head) [9, 10].

Previous magnetization measurements on heterostructures with discrete thicknesses indicate a smooth transition from FM to AFM coupling [2, 8]. Wedge-shaped samples provide a method for exploring the transition regions with magnetic force microscopy (MFM), allowing measurements of domain size and structure with excellent spatial resolution along the wedge. In particular, this paper demonstrates the correlation between magnetic domain size and the strength of the IEC, as well as the coupling between the two [Co/Pt] layers in the transition region from AFM to FM coupling, where the magnitude of the coupling goes through zero.

Two identical samples (A and B) were sputtered simultaneously on a natively oxidized Si substrate from separate Cu, Pt, Co and NiO targets in a similar fashion to [8]. X-ray diffraction shows that the Pt layers are polycrystalline but highly fcc (111) textured, leading to the necessary NiO fcc (111) texturing [8]. Off-axis sputtering produced a NiO layer wedge ranging in thickness from ~6–18 Å (determined below) across a 2” long strip. This shallow wedge angle maintains good (111) texturing of the NiO layer, where higher angle wedges lead to a breakdown of NiO texturing (likely due to strain). The sample schematic for both samples is (figure 1: inset)

\[
\text{Si(111)/Pt(200 Å)/[Pt(6 Å)/Co(4 Å)]_y/NiO(t_{\text{NiO}} Å)/[Co(4 Å)/Pt(6 Å)]_y/Cu(50 Å)}
\]

The shape of the NiO layer was characterized using x-ray reflectivity (XRR) on a thicker wedge and scaled down with time, based on the assumption that both the thickness and the thickness profile scale with time (similar to the wedge study in [8]). The absolute thickness for the center of the NiO layer (along with the other layers in the heterostructure) was checked by placing a recently calibrated in situ crystal thickness monitor at the exact sample position (corresponding to the center of the wedge) and measuring necessary growth times for each layer (including anomalous times such as shutter open and close times). The monitor was then moved and replaced with the sample substrate without breaking the vacuum. This technique has proven quite reliable for us in previous studies [8].

Figure 1. Room temperature \(J_{\text{IEC}}\) values (obtained from minor loop shifts) as a function of NiO thickness along the wedge (as indicated in the illustration). Above 8 Å NiO, the coupling smoothly oscillates with NiO thickness from AFM to FM coupling. Below 8 Å the existence of pinholes leads to dominant FM coupling. The coupling strength follows a simple cosine function with exponential damping, indicated by the solid red line fit. In the lower inset, the major and minor hysteresis loops are shown for \(t_{\text{NiO}} = 10.3\) Å. The minor loop of the top [Co/Pt] layer (red) is shifted toward positive field indicating AFM coupling.

Magnetization measurements were performed at room temperature on sample A using the perpendicular magneto-optical Kerr effect (PMOKE) while scanning along the length of the NiO wedge (figure 1). The difference in the saturation magnetization for the two [Co/Pt] layers (figure 1, inset) can be attributed to a change in the microstructure of the upper multilayer due to the intervening NiO spacer layer, which also leads to the difference in coercive values [8]. PMOKE measurements of the minor loop of the upper [Co/Pt] layer (figure 1, inset), indicate a positive (negative) shift in the minor loop. This shift determines the magnitude of the AFM (FM) coupling [2, 8], given by \(J_{\text{IEC}} = M_s H_{\text{IEC}}\), where \(M_s\) is the saturation magnetization, \(H_{\text{IEC}}\) is the minor loop shift and \(t\) is the Co thickness. A cross calibration against the XRR data correlates the coupling strength, \(J_{\text{IEC}}\), with NiO thickness (figure 1). The coupling oscillates smoothly, transitioning from AFM to FM and back to AFM with increasing NiO thickness, with a period of 5.1 Å. This smooth transition must arise from incomplete monolayers of NiO, with the net coupling (whether FM or AFM) arising from a complex interplay between the various energies involved.

Sample B was maintained in a virgin state. MFM images of this sample, taken along the length of the wedge, are shown in figure 2. These images, corresponding to varying coupling strengths, were taken in tapping/lift mode at a lift height of 5 nm under ambient conditions. The magnetization of the MFM tip is perpendicular to the sample surface, pointing downward [11]. Using a mechanical translation stage, the NiO thickness (from XRR) and the corresponding \(J_{\text{IEC}}\) (from PMOKE) were calibrated and are both indicated on each panel of figure 2. Note that positive (negative) \(J_{\text{IEC}}\) values correspond to AFM (FM) coupling.
MFM measures the net magnetization through the depth of the sample, including the top and bottom [Co/Pt] layers. For these coupled samples, three levels of contrast are observed. FM coupled samples, in which the domains are aligned parallel in both layers, display both up-up (light colored) and down-down (darkest colored) domains, with a sharp contrast between the two, typified in the samples with $t_{\text{NiO}} = 8.5$ and 13.2 Å. In the AFM coupled samples, anti-parallel alignment of top and bottom domains results in zero net magnetization and an intermediate level of contrast. In addition, at the domain walls a slight shift of the two domains (upper and lower) leads to the observation of FM stripes. This phenomena leads to three separate levels of contrast, as in the sample with $t_{\text{NiO}} = 10.3$ Å. These FM stripes have been well characterized and are a result of competition between the AFM coupling and the magnetostatic interlayer interaction (favoring FM alignment) between the two [Co/Pt] layers [8, 12–15].

Using ImageJ, a public domain Java image processing program [16], the average domain size was determined for each MFM image. ImageJ was designed with an open architecture that provides extensibility via Java plug-ins. One particular plug-in allows the user to define a boundary (domain edge), determined by a controlled threshold, and then digitizes the image into a bilevel representation (i.e. domain A cf domain B). For the FM coupled regions, this boundary was defined by the sharp contrast across a domain wall (transition from up to down domains), where the mask edge was defined as

![Figure 2](image_url)

**Figure 2.** MFM images of coupled [Co/Pt] multilayers with different thicknesses of the NiO spacer layer. The strength of the IEC listed (in units of merg cm$^{-2}$) is based on PMOKE data taken at each thickness. The images display three levels of contrast, as discussed in the text—light (yellow) areas indicate a magnetization pointing up (out of the page), dark (red) areas correspond to magnetization pointing down and the intermediate color (orange) indicates AFM coupled domains with zero net magnetization. The domain size and structure vary non-monotonically with NiO thickness. Each image is $5 \times 5$ µm$^2$ in size.
The center of this sharp contrast. Thus, we only measured the size of up–up domains. For the AFM coupled regions, the FM stripe in the vicinity of the domain wall defined the boundary between domains, where the center of this FM stripe defined the mask edge. For the AFM coupled case, the up/down and down/up domains are indistinguishable, so we decided to choose the smaller of the two regions, consistent with the FM coupled case (this has previously been confirmed with x-ray magnetic circular dichroism photoemission electron microscopy, XMCD-PEEM [8]). The areas of these masked domains are binned and plotted as a histogram, giving both the average size and standard deviation. A direct, monotonic correlation between the domain size and the magnitude of the coupling strength (not NiO thickness) is established (figure 3).

Equilibrium domain sizes in coupled FM layers with PMA are determined by various competing terms in the free energy [14, 17]. These include the magnetostatic intralayer ($E_{M}^{(0)}$) and interlayer ($E_{M}^{(1)}$) energy, the IEC ($J_{IEC}$) between the two [Co/Pt] layers and the domain wall energy ($E_{dw}$). An estimate of the energy contributions using a stripe domain model [14] is shown in table 1. The magnetostatic energies favor smaller domains, competing with $E_{dw}$, which favors the formation of larger domains. $E_{M}^{(0)}$ (several orders of magnitude larger than any contributing energy) is constant over the entire range of NiO thickness and $E_{M}^{(1)}$ varies very little compared to $J_{IEC}$.

The additional interfacial energy contribution from $J_{IEC}$ ranges from $-7.46-8.17 \times 10^{-3} \text{ erg cm}^{-2}$ (figure 1). Scaling by the Co thickness, $t$, gives an effective field term, $J_{IEC}/t = M_{s}H_{IEC}/t = M_{s}J_{IEC}$. The energy density values for this Zeeman-like term range from $-0.062-0.068 \times 10^{6} \text{ erg cm}^{-3}$. At the IEC maxima the energy values are roughly four times larger than $E_{M}^{(1)}$, and are comparable with $E_{dw}$, which is why the FM stripe is observed in the AFM coupled samples [10].

The effect of purely magnetostatic coupling will be to increase the domain size across the wedge, as $E_{M}^{(1)}$ decreases due to increased NiO spacing [14, 17]. In contrast, the domain size, governed by the local minimum in energy of the ‘as-deposited state’, correlates directly with the strength of the IEC, since this effective field field term is linear with $J_{IEC}$. However, unlike externally applied fields [18], the favorable, or lower energy, domain configuration is determined by the sign of the coupling, where AFM coupling favors a growth in anti-parallel aligned domains and FM coupling favors parallel ones. Thus, a decrease in the IEC lowers the energy cost for domain formation and $E_{M}^{(1)}$ will dominate, leading to the formation of smaller domains for the weakly coupled samples to minimize the magnetostatic energy.

In the transition regions, where $J_{IEC}$ is comparable to or smaller than $E_{M}^{(1)}$, $E_{M}^{(0)}$ will result in a characteristic domain size for each [Co/Pt] layer. However, the domains in the upper and lower [Co/Pt] layers may not be in registry due to the weak interlayer interactions. MFM images of weakly coupled samples in figures 2 and 4 display all three levels of contrast (for example, see figure 2 for $t_{\text{NiO}} = 11.6 \text{ Å}$) corresponding to roughly equivalent areas for both FM and AFM coupled domains. To further investigate the transition region, MFM images were measured at 100 µm steps through the region at $t_{\text{NiO}} = 11.6 \text{ Å}$. A visual comparison of figures 4(a) and (i), over which the nominal NiO thickness changes by only ~0.2 Å, suggests two differences: (i) a decrease in the area of the intermediate level of MFM contrast, which corresponds to AFM coupled domains, and (ii) a slight increase in the FM coupled domain size.* As was done for figure 2, using ImageJ, the up–up (FM coupled) domains were masked off and the average sizes of these domains were measured. A slight monotonic increase in domain size (ranging from 0.16 ± 0.05 to 0.27 ± 0.04 µm²), corresponding to a small increase in FM coupling, is observed (figure 5). Note that the domain sizes in this transition region are consistent with the minimum domain size obtained from the inset of figure 3. In this transition

<table>
<thead>
<tr>
<th>$E_{M}^{(0)}$ (erg cm$^{-3}$)</th>
<th>$E_{M}^{(1)}$ (erg cm$^{-3}$)</th>
<th>$E_{dw}$ (erg cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$3.60 \times 10^{6}$</td>
<td>$(0.014-0.016) \times 10^{6}$</td>
<td>$0.57 \times 10^{6}$</td>
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Table 1. Comparison of the calculated energies using the stripe domain model for [Co/Pt] layers separated by a spacer layer (as described in the text [14]) with the following parameters: Co thickness $t = 1.2 \times 10^{-7} \text{ cm}$ (from XRR), separation between the two [Co/Pt] layers $d = 1.1 \times 10^{-7} \text{ cm}$ (from XRR), saturation magnetization $M_{s} = 760 \text{ emu cm}^{-3}$ (from superconducting quantum interference device measurements), average domain stripe width $L = 1.0 \times 10^{-4} \text{ cm}$ (for a micron domain size neither $E_{M}^{(0)}$ nor $E_{M}^{(1)}$ depend strongly on the value of $L$).
region of small $J_{IEC}$, the dominant interlayer coupling is magnetostatic in origin, favoring FM coupled domains. Traversing the wedge from (a) to (i) in figure 4, over a nominal thickness change of NiO equivalent to less than one-tenth of a monolayer, the coupling goes from very weakly AFM (as evidenced by the presence of some intermediate contrast) to very weakly FM. The magnetostatic coupling biases the MFM images in the direction of parallel coupled domains and is expected to be nearly constant over this region; hence, the exact position at which $J_{IEC} = 0$ is hard to determine. One approach we propose for assisting in this determination would be to isolate the domain structure to a single [Co/Pt] layer by first growing the lower layer, saturating it into a single domain state, and then growing the NiO interlayer and upper [Co/Pt] layer. This method would result in data similar to those from XMCD-PEEM images of these types of structures, where only the topmost layer was able to be imaged [8].

However, from the present data, it is clear that the transition from AFM to FM coupling occurs over ~0.2 Å, comparable to the transition region determined by scanning electron microscopy with polarization analysis (SEMPA) experiments on Fe/Cr/Fe structures with in-plane anisotropy [19]. The roughness of the sputtered NiO wedge is larger than the evaporated Cr wedge in the SEMPA experiments, where atomic force microscopy measurements on sputtered NiO films grown under similar conditions indicate an RMS roughness of ~4 Å, with lateral grain sizes of <0.1 µm [20].

The transition from AFM to FM coupling in figure 2 occurs as the NiO film transitions between odd ($n$) and even ($n+1$) numbers of monolayers. On the sub-micron length scale of MFM measurements, domain-by-domain coupling is observed in the transition region, with areas of both FM and AFM coupling. The areas of NiO thickness variation are significantly smaller in lateral size [21] than a magnetic domain. Thus, within a single magnetic domain, the dominant coupling mechanism will be determined by the preponderant layer thickness (slightly biased toward FM coupling, as mentioned above, due to the weak but persistent $E_M^{(1)}$). Although it is not possible to image these regions of differing NiO thickness, a consequence of this effect is apparent in the domain wall region of AFM coupled samples, where a FM stripe is formed, governed by a $1/J_{IEC}$ width dependence [8, 15]. As the AFM IEC decreases, the width of this FM stripe increases as $E_M^{(1)}$ is nearly constant and favors FM alignment [15]. As the IEC decreases below ~25% of its maximum, $E_M^{(1)}$ dominates (table 1) and the FM stripes now evolve.

Figure 4. MFM images of weakly coupled [Co/Pt] multilayers, with slightly different thicknesses of the NiO interlayer corresponding to the position along the wedge. Each image corresponds to a 100 µm step along the wedge in the transition region from AFM to FM coupling ($t_{NiO} = 11.6$ Å). The total change in NiO thickness for the entire series is ~0.2 Å (i.e. ranges from (a) 11.5 Å to (i) 11.7 Å). Each image is 5 × 5 µm² in size.
into FM domains, governed by $E^{(0)}_m$, $E^{(1)}_m$ and $E_{av}$. Note that unlike other experiments with coupling across AFM spacer layers with an in-plane FM, for example Co/NiO/NiFe [21], averaging within a domain will not lead to non-collinear coupling. This is due to the strong PMA. On the macroscopic length scale probed by PMOKE, the net coupling is determined by whichever NiO thickness dominates the area, with a magnitude weighted by the presence of both $(n)$ and $(n + 1)$ monolayers.

In conclusion, an investigation of the correlation between magnetic domain formation and IEC on the only system with PMA that directly displays an oscillatory IEC (traversing from FM to AFM, as indicated by the minor loop shift) indicates that there exists a direct, monotonic relationship between domain size and the magnitude of the IEC. The IEC serves as an effective field that favors larger domains based on the sign of the coupling. In the transition region between FM and AFM coupling, the IEC is small and the magnetostatic energy dominates, leading to smaller domains that are preferentially FM coupled. The transition region is extremely narrow, and the scale for this transition region arises from a combination of atomic scale roughness in the NiO layer as well as fundamental limits on magnetic domain sizes in PMA materials.

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References