

Determination of critical thickness of spin reorientation in metastable magnetic ultrathin films

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We investigate the spin reorientation of structurally unstable magnetic ultrathin films (base films) using magnetic capping layers. The capping layers, whose effective anisotropy constant is opposite that of the base films, are grown on the base films with no transformed structure and induce a spin reorientation after exceeding a critical thickness. The critical thickness of the capping layers can be used to deduce the critical thickness as well as the anisotropy constants of the base films. We use Co to cap the well-known Fe/Cu(100) system as an example to demonstrate the proposed approach.

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Magnetic anisotropy plays a crucial role in supporting long-range order in magnetic ultrathin films.^{1,2} Determination of magnetic anisotropy constants requires either sophisticated measurements such as ferromagnetic resonance^{3,4} or Brillouin light scattering,⁵ or hard-axis saturation by a very large external magnetic field.⁶ In systems where a perpendicular magnetization shows up at low thickness, studying the critical thickness of the thickness-driven spin reorientation would also provide reliable information about anisotropy constants.⁷ Unfortunately, the latter approach, although more accessible in laboratories, is not applicable to some of the metastable magnetic ultrathin film systems where the structural transition takes place before the spin reorientation. Even though the structural transition itself could also lead to a “spin reorientation,” it is irrelevant to the conventional spin reorientation and gives no direct information about the magnetic anisotropy constants.

A typical example of such a metastable system is the widely studied Fe/Cu(100) ultrathin film system.⁸⁻¹² When grown on Cu(100), the face-centered cubic (fcc) γ -Fe phase, which exists only above 1100 K in bulk, is stabilized by the Cu substrate. The spin reorientation behavior of the Fe films depends strongly on the growth temperature. For room temperature (RT) grown Fe films, perpendicular magnetization persists up to 10 ML before reorientation to the in-plane direction. It is generally believed that such unusually high critical thickness of the spin reorientation is primarily due to the fact that a structural transformation from face centered tetragonal (fct) to fcc phase occurs at about 4 ML, which results in a spin configuration of the films with ferromagnetic layers on top and nonmagnetic layers underneath in the thickness regime between 4 and 10 ML. The observed spin reorientation is correlated with a fcc \rightarrow bcc structural transformation at 10 ML which is apparently not a conventional thickness driven spin reorientation.

Low temperature [(LT) 130 K or below] grown Fe/Cu(100) films have a considerably lower critical thickness for spin reorientation (i.e., ~ 3.8 ML).¹³ It remains a matter of dispute whether the spin reorientation of the LT Fe films is caused by the fcc to bcc structural transformation¹³ or the

usual balance between the surface and volume anisotropy.⁶ Without doubt, for metastable films like Fe/Cu(100), determination of the “true” critical thickness (i.e., the critical thickness assuming no structural transformation involved) of the spin reorientation would not only provide great insight into the origin of the spin reorientation, but also contribute directly in determining the anisotropy constants of these films.

In this letter, we study the true critical thickness of the spin reorientation for both RT and LT grown Fe/Cu(100) films by capping the perpendicularly magnetized fct Fe/Cu(100) films (<4 ML) with the in-plane magnetized fcc Co(100) films. The competition between the in-plane magnetization of the Co films and the perpendicular magnetization of the Fe films would lead to a spin reorientation from perpendicular to in plane when the Co capping layers exceeds certain critical thickness d_c^{Co} . Such capping layer induced spin reorientation was previously observed in a Co/60 Å Ni/Cu(100) film at Co thickness of about 6 Å.¹⁴ Here, we utilize this phenomena to study the spin reorientation of the metastable base (Fe) films, by determining the dependence of the value of d_c^{Co} on the Fe thickness. The interception with the x axis of the d_c^{Co} vs Fe thickness plot corresponds directly to the true critical thickness of Fe films.

Experiments were performed in an ultrahigh vacuum (UHV) system (base pressure better than 7×10^{-11} Torr) equipped with facilities for low energy electron diffraction (LEED), Auger electron spectroscopy (AES), and magneto-optical Kerr effect (MOKE). The MOKE setup operates in both polar and longitudinal geometries by rotating the magnet around the sample. Prior to Fe deposition, the Cu substrate was cleaned by cycles of Ne ion sputtering and annealing at 900 K until clean AES results and sharp (1×1) LEED patterns were obtained. The Fe (Co) films were evaporated from an Fe (Co) wire heated by e -beam bombardment. The amount of the Fe (Co) deposition was controlled by a flux monitor mounted on the Fe (Co) source, which was calibrated using AES. The accuracy of the AES in thickness determination was significantly improved after being calibrated using scanning tunneling microscopy (STM) in a separate UHV chamber. The Fe films were first deposited at RT and cooled to 110 K for MOKE measurements. The Co films were subsequently deposited on the Fe/Cu(100) film at

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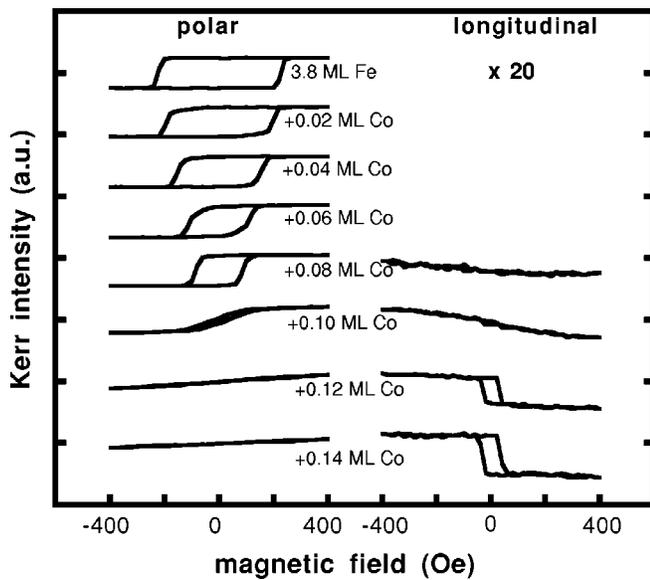


FIG. 1. Kerr hysteresis loops of a room temperature grown 3.8 ML Fe/Cu(100) film with Co capped layers of various thickness. When capped by 0.1 ML of Co, the Fe film starts to show canted axis as evidenced by the appearance of canting curves in both polar and longitudinal geometry. Well-defined in plane rectangular loops were measured with additional Co capping.

110 K using 0.02 ML steps. Polar and longitudinal MOKE data were recorded after each deposition step.

Figure 1 demonstrates the typical spin reorientation induced by Co capping of a 3.8 ML Fe/Cu(100) film grown at RT. Polar and longitudinal MOKE hysteresis curves are shown for various Co thicknesses. With increasing Co thickness, the coercivity H_c of the films decreases quickly although the easy magnetization axis remains perpendicular to the surface until 0.08 ML is deposited, below which no longitudinal magnetic signal can be detected. After an additional 0.02 ML of Co deposition, the film starts to show canted hysteresis loops in both polar and longitudinal geometries (i.e., a spin reorientation occurs after a total amount of 0.1 ML of Co deposition on top of the 3.8 ML Fe film). Further increasing the Co thickness immediately results in well-defined in-plane rectangular loops indicating the completion of the spin reorientation. We thus determine the value of d_c^{Co} for the 3.8 ML Fe film to be about 0.1 ML, with an error bar of 0.02 ML. It is important to point out that the error bar is determined by the incremental step of the Co deposition (0.02 ML), which can be improved by choosing a smaller value.

Similar Co capping experiments have been performed on Fe films grown at LT. Figure 2 shows a series of polar (left panel) and longitudinal (right panel) hysteresis loops of a 2.4 ML LT-grown Fe film as a function of the Co capping thickness. The value of d_c^{Co} is again determined to be 0.1 ML.

Following the described procedure, we are able to obtain d_c^{Co} values for both RT- and LT-grown Fe films of various thickness within the untransformed fct thickness regime (<4 ML). Figure 3 shows the values of d_c^{Co} as a function of thickness for Fe films grown at RT (solid circles) and LT (open circles). In both cases, the data points follow clearly a linear fit as indicated by the dashed lines. The x -axis intercepts, 4.8 and 3.5 ML, are the values of the critical thick-

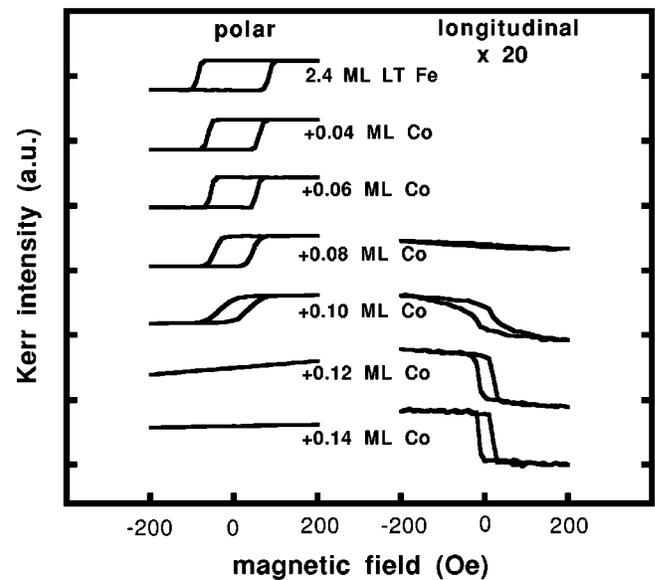


FIG. 2. Kerr hysteresis loops of a low temperature grown 2.4 ML Fe/Cu(100) film with Co capped layers of various thicknesses. When capped by 0.1 ML of Co, the Fe film starts to show canted axis as evidenced by the appearance of canting curves in both polar and longitudinal geometry. Well-defined in plane rectangular loops were measured with additional Co capping.

ness for spin reorientation of the Fe/Cu(100) films without Co capping for RT and LT films, respectively. In other words, the easy magnetization axis of the Fe/Cu(100) films would reorient from perpendicular to in plane at 4.8 ML for a RT film and 3.5 ML for a LT film if there had been no structural transformation.

Comparing the extrapolated true critical thickness to the measured critical thickness of the spin reorientation is crucial for understanding the origin of the spin reorientation. For the films grown at RT, all literature data including our own show that the measured critical thickness is about 10 ML, which is twice as high as the extrapolated value from Fig. 3. The significant delay of the spin reorientation, as mentioned, is a direct result of the fct \rightarrow fcc structural transformation which occurs between 4 and 5 ML. For the LT-grown Fe films, we notice that the extrapolated true critical thickness is practically the same as the reported critical thickness,¹³ which implies that the origin of the spin reorientation is the usual balance between the surface and shape anisotropy, rather than the formation of bcc structure.

The magnetic anisotropy constants of Fe/Cu(100) films can be estimated from Fig. 3. We use a phenomenological ansatz to express the second-order anisotropy constant K_1

$$K_1 = K_1^V + \frac{K_1^S}{d}, \quad (1)$$

where K_1^V is the volume anisotropy, K_1^S is the sum of surface and interface anisotropies, and d is the thickness of the film. At the critical thickness of spin reorientation, the second-order magnetic anisotropy constant K_1 and the shape anisotropy roughly cancels out, i.e., $K_1 - 2\pi M^2 = 0$. For the Co capped Fe/Cu(100) films, K_1^S has to include the contribution from both the Fe and Co films weighted by their exposed surface area, i.e.,

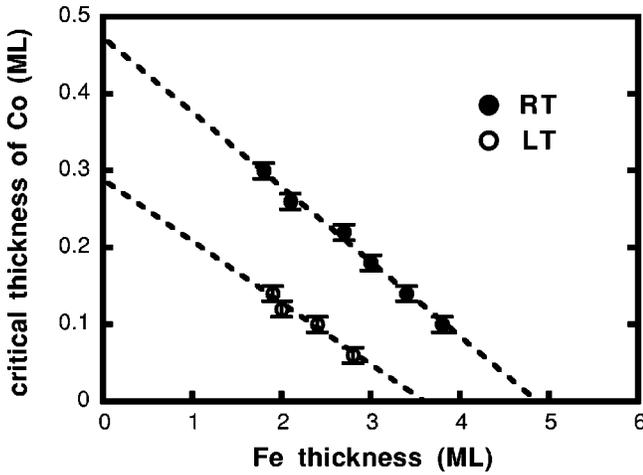


FIG. 3. Critical thickness of Co capping layers (upper panel) as a function of the thickness of the underneath Fe layers. The dashed lines serve as a guide for the eye.

$$K_1^S = \varphi * (K_{1,\text{Co-Vac}}^S + K_{1,\text{Co-Fe}}^S) + K_{1,\text{Fe-Cu}}^S + (1 - \varphi) * K_{1,\text{Fe-Vac}}^S, \quad (2)$$

where $K_{1,\text{Fe-Vac}}^S$, $K_{1,\text{Fe-Cu}}^S$, $K_{1,\text{Co-Vac}}^S$, and $K_{1,\text{Co-Fe}}^S$ are the interface anisotropy constants of Fe–vacuum, Fe–Cu, Co–vacuum, and Fe–Co, respectively. Here φ is the Co coverage of the surface area and equals the nominal thickness of Co assuming the Co islands have monatomic step height. This assumption should be roughly correct since the Co thickness is very low (<0.3 ML). Obviously, a more accurate value for φ requires a STM study. When the Co induced spin reorientation occurs, we have

$$(K_{1,\text{Fe}}^V - 2\pi M_{\text{Fe}}^2) * d_{\text{Fe}} - 2\pi M_{\text{Co}}^2 * d_{\text{Co}} + K_{1,\text{Fe-Cu}}^S + (1 - \varphi) * K_{1,\text{Fe-Vac}}^S + (K_{1,\text{Co-Vac}}^S + K_{1,\text{Co-Fe}}^S) \varphi = 0. \quad (3)$$

Note that in Eq. (3) we assume the volume anisotropy of Fe films $K_{1,\text{Fe}}^V$ were not affected by the Co capping layers, which are justified by both the linear dependence of $d_{\text{Co}}^{\text{Co}}$ vs Fe thickness (see Fig. 3), and the fact that the magnetization of Fe has hardly been changed after Co capping (see Figs. 1 and 2). The number of unknowns in Eq. (3) can be reduced by roughly assuming that the magnetic film–vacuum interface anisotropy and the magnetic film–nonmagnetic substrate interface anisotropy have the same value, i.e., $K_{1,\text{Fe-Cu}}^S = K_{1,\text{Fe-Vac}}^S = K_{1,\text{Fe}}^S$. Since the slope and the interception in Fig. 3 would only give two independent equations with three unknowns ($K_{1,\text{Fe}}^S$, $K_{1,\text{Co-Fe}}^S$, and $K_{1,\text{Co-Vac}}^S$) to be solved, one needs to know the value of $K_{1,\text{Co-Fe}}^S$ by other means. We created a RT-grown bilayer system of 1 ML Co/1 ML Fe/Cu(100) which has an in plane easy magnetization axis. Capping this bilayer film with Fe leads to an inverse spin reorientation from in plane to perpendicular with about 0.35 ML of Fe capping.¹⁵ The third equation can thus be obtained as follows:

$$(K_{1,\text{Fe}}^V - 2\pi M_{\text{Fe}}^2) * d_{\text{Fe}} - 2\pi M_{\text{Co}}^2 * d_{\text{Co}} + 0.65 * K_{1,\text{Co-Vac}}^S + 1.35 * K_{1,\text{Co-Fe}}^S + 1.35 * K_{1,\text{Fe}}^S = 0. \quad (4)$$

We take the literature value¹⁶ obtained from Co/Cu(100) at 77 K (-0.508 ergs/cm²) for $K_{1,\text{Co-Vac}}^S$, and bulk values of

bcc Fe (1714 G) and hcp Co (1422 G) at RT¹⁷ for M_{Fe} and M_{Co} , respectively. After some calculation, Eqs. (3) and (4) yield $K_{1,\text{Fe}}^S = 0.16$ ergs/cm², $K_{1,\text{Fe}}^V = 1.49 \times 10^7$ ergs/cm³, and $K_{1,\text{Co-Fe}}^S = 0.35$ ergs/cm² for the RT Fe film. Putting the $K_{1,\text{Co-Fe}}^S$ value into Eq. (4) would then yield $K_{1,\text{Fe}}^S = 0.08$ ergs/cm², $K_{1,\text{Fe}}^V = 1.59 \times 10^7$ ergs/cm³ for the LT grown Fe film. It is interesting to note that the Fe–Co interfacial anisotropy, $K_{1,\text{Co-Fe}}^S$, is a positive value favoring perpendicular magnetization.

The smaller surface anisotropy of the LT Fe film is consistent with the fact that the LT film has a significantly rougher surface (compared to the RT film)¹⁸ which is known to reduce the effective surface anisotropy.¹⁹ From our data, it is clear that the surface anisotropy constants for both the RT and the LT Fe/Cu(100) films, though being positive, is too small to overcome the shape anisotropy at any given thickness above 1 ML. It is the large positive volume anisotropy, which most likely originates from the tetragonal expansion of the film, that supports the perpendicular magnetization in this system. This also implies that a cubic-like Fe/Cu(100) would have an in plane easy magnetization axis, which has been recently demonstrated experimentally with Fe/Cu(100) films grown by pulsed laser deposition.²⁰

In summary, we have used magnetic capping layers to study the spin reorientation and the anisotropy constants of metastable magnetic ultrathin films. The anisotropy constants from the tested system, Fe/Cu(100), have been calculated from the experimental data. We believe that this method is applicable to study the spin reorientation of other metastable magnetic ultrathin film systems.

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