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Anomalous magnetic anisotropy in ultrathin transition metals

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We have used *in situ* polar Kerr effect measurements to probe directly the magnetic anisotropy of ultrathin MBE-grown Co films with and without overlayers of various nonmagnetic metals (X=Ag, Cu, Pd). To investigate the Co/X interface, we measured perpendicular hysteresis curves as a function of overlayer coverage. We find the magnitude of the perpendicular anisotropy is strongly peaked near ~1 atomic layer (1.5–2 Å) coverage for Ag and Cu overlayers. For Cu, the overlayer with the strongest effect, the total anisotropy energy rapidly decreases by a factor of 3 from its peak value after a total coverage of only ~2 atomic layers (4 Å) of Cu.

In spite of considerable progress in modern magnetism, the origin of the perpendicular anisotropy observed in certain layered magnetic thin films[1] remains an important unresolved problem.[2] Possible explanations advanced for this strong anisotropy include the reduced coordination symmetry,[3] altered electronic structure,[4] and possible localized expitaxial strain at the interface between two different materials.[5] However, to date information has been lacking on the roles of material species, crystal structure, orientation, lattice strain, and electronic band-structure interactions.[4] Basic to developing a theoretical understanding of the origin of interface-induced magnetic anisotropy is determining the relative importance of the structural and electronic contributions.

To date, work in the area of interface anisotropy has concentrated on the study of two-component multilayers[1,6–11] and magnetic/nonmagnetic overlayer-substrate systems.[11–14] Calculations of the spin anisotropy have been performed for the limiting case of unsupported ferromagnetic monolayers,[15] certain overlayer-substrate combinations,[16] and particular metallic superlattices.[17,18] These calculations rely greatly on the details of the electronic band structure of the individual materials and have had limited success due to the difficulty of solving this strongly interacting problem.

In this paper we present results from trilayer experiments designed to investigate the evolution of perpendicular magnetic anisotropy during the controlled formation of a magnetic/nonmagnetic metal interface. Previous evidence of changes in magnetic behavior in the low-coverage regime include a reduction in Curie temperature of ultrathin Co(001) films when covered by Cu (Ref. 19) and an epitaxial structure-induced change in the in-plane easy-axis of Fe(110) films when covered by Au.[14]

In this work, we have used *in situ* Kerr effect measurements in our molecular-beam epitaxy (MBE) system to study the effect of different nonmagnetic metal interfaces (X=Ag, Cu, and Pd) on the magnetic properties of ultrathin single-crystal Co films. We have performed a systematic study of the structural and magnetic behavior as a function of Co film thickness, overlayer coverage, and material species.

The Co films in this study were epitaxially grown at room temperature on thick, 300–500 Å, Pd(111) buffer layers deposited onto Co-seeded GaAs(110) substrates.[20] The background pressure during deposition was $\leq 5\times10^{-10}$ Torr and was composed predominantly of H_2. We used effusion cells for Ag and Pd deposition at 0.1 Å/s and 0.15 Å/s, respectively, and optical-feedback-controlled *e*-beam evaporators to deposit the Co (0.25 Å/s) and Cu (0.1 Å/s). All deposition rates were determined from Rutherford backscattering spectrometry analysis of calibration films and were reproducible to within ±10%. Film quality and crystal structure were monitored during growth with reflection high energy electron diffraction (RHEED). The RHEED pattern images were captured and digitized with a computer-based video system capable of resolving changes in surface lattice spacings on the order of 1%.

A sample can be transferred between the deposition chamber of our MBE system and another connected ultrahigh vacuum chamber ($P<2\times10^{-10}$ Torr) where it is aligned between the poles of an external electromagnet. The perpendicular field at the sample can be swept to ±2.2 kOe, with optical access provided by a hole along the axis of one pole. The sample can be repeatedly moved from the measurement chamber to the deposition chamber and back without need for optical realignment. We use a 50 kHz photoelastic modulator and lock-in amplifier based detection scheme to measure the polar (perpendicular) Kerr ellipticity of the sample, thereby eliminating the background Faraday rotation from the quartz vacuum window. Stated in terms of bulk Co properties, sensitivity of this system is sufficient to detect the equivalent of 0.5 monolayers of bulk Co.

We grew our samples on Pd(111) buffer layers so that the initial Pd/Co interface would provide a perpendicular anisotropy in competition with the in-plane shape contribution of the Co film. We could therefore adjust the initial total anisotropy of individual samples to be either perpendicular or in plane by selecting the proper Co film thickness. This allowed us to work within the 2.2 kOe limit of our perpendicular magnetic field. Using this idea of designing samples which balance the relatively strong shape and interface anisotropies against each other allows us to very easily observe small changes in the total anisot-

ropy energy due to overlayer deposition.

After deposition of the Pd buffer layer and Co film, *in situ* polar Kerr hysteresis curves were measured from the initial uncovered sample. Kerr measurements were then repeated after each of the many overlayer depositions spanning the coverage range $0 \leq t_X \leq 100$ Å in small 0.5–10 Å steps. The entire deposition/measurement cycle for each coverage step required ≈ 10 min. in a background pressure of $\approx 1\text{–}5 \times 10^{-10}$ Torr.

For 2 Å $< t_{Co} \leq 6$ Å the perpendicular hysteresis curves of the initial uncovered Co films are nearly square, with coercive fields of $H_c \leq 200$ Oe. Above this Co thickness, the shape anisotropy dominates that provided by the Pd/Co interface and the easy-axis becomes in plane. This results in a linear hysteresis curve when measured in the perpendicular direction. However, we find for 6 Å $\leq t_{Co} \leq 10$ Å subsequent deposition of approximately one atomic monolayer [ML ≈ 2 Å for the (111) direction of these fcc metals] of any of the nonmagnetic metals Ag, Cu, or Pd results in strongly perpendicular square hysteresis loops.[21] This surprising phenomenon indicates that a large change in the perpendicular anisotropy results from a very small amount of overlayer material. To understand the origin, additional amounts of nonmagnetic metal were deposited. As shown in Fig. 1, further coverage causes a marked change in the perpendicular coercivity suggesting concurrent nonmonotonic changes in the total anisotropy with coverage. The most striking feature of Fig. 1 is the pronounced peak at 1.5±0.2 Å coverage for both Co thicknesses which, to within our resolution and control, is located at or just below 1 ML. Increasing the coverage beyond ~2 ML (4 Å), H_c begins to increase again, reaching a value near that of the peak coercivity by ~100 Å.

To determine if changes in the saturation magnetization as a function of coverage might be responsible for the behavior shown in Fig. 1, we monitored the saturation ellipticity of these square hysteresis curves. In contrast to the coercivity shown in Fig. 1, the ellipticity displays only a slow monotonic decrease as the Kerr signal from the magnetic Co is progressively reduced as it is buried by the nonmagnetic Cu.[21] Hence, the saturation magnetization of these films is not changing significantly with overlayer coverage. Neither is there any evidence from our simultaneously measured RHEED data of any large structural changes. The RHEED streak spacings change continuously from those of the Co surface to those of bulk Cu (~2% difference) by 6 Å Cu coverage. We cannot, however, rule out any subtle structural changes below our ~1% resolution. Because RHEED measures only the in-plane lattice spacing it is not sensitive to any changes in the perpendicular atomic spacings. Also, because we cannot say at present whether there is complete monolayer coverage of Cu at 2 Å, it is difficult to determine the presence of interfacial strain from RHEED alone. However, the behavior shown in Fig. 1 appears to be quite general, in that we observe similar behavior for Ag, Cu, and Pd overlayers each of which has a different lattice mismatch with Co (14%, 2%, and 9%, respectively). Figure 2 is a plot of coercivity versus overlayer thickness of 8 Å Co covered by Ag, Cu, and Pd. For the Pd overlayers there is a large increase in coercivity, although the peak in H_c is broad and much less pronounced than for the other two metals and is shifted to ~3 Å coverage.

The pronounced peak in coercivity we observe at ~1 ML is highly suggestive of an abrupt change in anisotropy. However, coercivity is the result of the interaction of several parameters rather than being a fundamental magnetic quantity. Consequently, we also have directly measured the uniaxial anisotropy of Co films *in situ* as a function of nonmagnetic overlayer coverage. To accomplish this, we measured the slope of the hard-axis magnetization curves and deduced the anisotropy field. Because our MBE machine is equipped with only a 2.2 kOe perpendicular magnet, we are limited to making these mea-

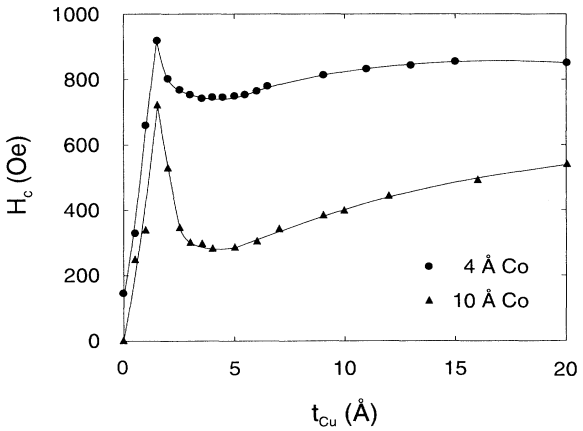

FIG. 1. Perpendicular coercivity vs Cu coverage for two samples with Co thicknesses of 4 Å and 10 Å. As discussed in the text, the pronounced peak at 1.5±0.2 Å Cu coverage results from an increase in perpendicular anisotropy at ~1 ML overlayer coverage. Uncertainties in H_c are the size of the data points. Estimated relative and absolute thickness uncertainties are ±5% and ±10%, respectively.

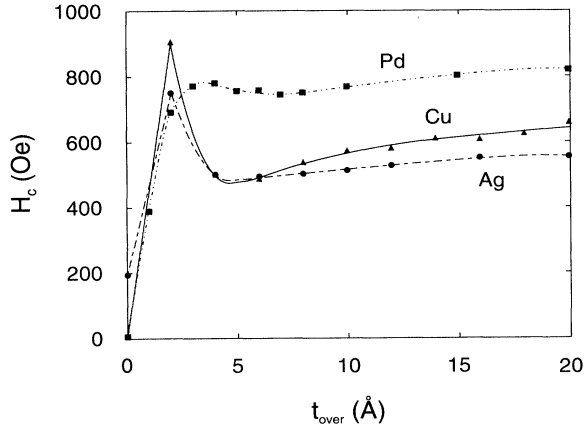

FIG. 2. Perpendicular coercivity vs overlayer coverage for three samples with 8 Å Co and Pd, Cu, and Ag overlayers. The behavior is qualitatively similar, however, the Pd overlayers display a broad, less pronounced peak in H_c that is shifted to ~3 Å coverage. Uncertainties in H_c are the size of the data points. Estimated relative and absolute thickness uncertainties are ±5% and ±10%, respectively.

surements on moderately anisotropic films with an in-plane easy axis. We can create films of this type by carefully selecting the proper Co film thickness that we grow on Pd(111) before overlayer coverage. Figure 3 is a series of hard-axis (perpendicular) hysteresis loops of a 12 Å Co film on Pd(111) covered by ultrathin overlayers of Cu. The top curve is that of the uncovered film; the linear behavior indicates it has an in-plane easy axis. However, upon coverage by 1 Å of Cu the slope increases. This indicates the Co/Cu interface has contributed a perpendicular component to the anisotropy, although, the overall anisotropy still remains in plane. Continuing the coverage to 2 Å Cu causes a further increase in the perpendicular component of the anisotropy, allowing the magnetization to be saturated by our 2.2 kOe field. It is clear from the figure that further Cu coverage above 2 Å then decreases the perpendicular anisotropy contribution. This behavior is the same as that exhibited by the coercivity discussed above.

Since we can saturate the moments in this sample, we are able to deduce the anisotropy field from extrapolation of the hard-axis curve to saturation and hence directly calculate the total anisotropy energy. Figure 4 is a plot of the anisotropy constant K_1 vs Cu coverage for Cu thicknesses to 50 Å. We determined the values from the anisotropy field H_k using the relation for a uniaxial crystal, $K_1 = H_k M_s/2$, where $M_s = 1422$ emu/cm^3 is the bulk saturation magnetization of Co. Here we have adopted the convention used by many multilayer researchers, where a positive K_1 indicates perpendicular anisotropy. After concluding our complete set of *in situ* measurements we removed the samples and verified their bulk magnetization behavior with a vibrating sample magnetometer. The overall negative anisotropy values of Fig. 4 indicate an in-plane easy axis was maintained throughout this coverage series.

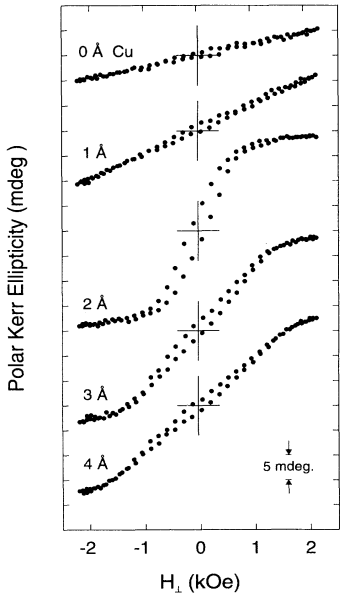

FIG. 3. Hard-axis (perpendicular) hysteresis curves from a 12 Å Co film as it is progressively covered with Cu. The perpendicular anisotropy contribution from the Co/Cu interface is seen to be a maximum at 2 Å Cu coverage.

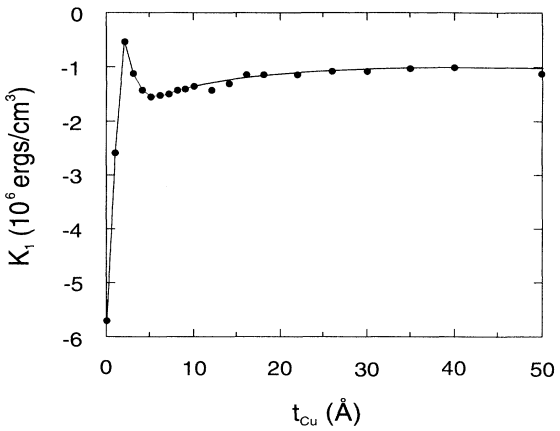

FIG. 4. The total uniaxial anisotropy energy of a 12 Å Co film vs Cu overlayer coverage. Values of $K_1 < 0$ indicate an in-plane easy axis. K_1 becomes increasingly in plane above 2 Å coverage changing by nearly a factor of 3 between $t_{Cu} = 2$ Å and 4 Å. Estimated relative and absolute thickness uncertainties are $\pm 5\%$ and $\pm 10\%$, respectively.

The total anisotropy energy shown in Fig. 4 displays the same peaked behavior with Cu coverage as we observed in the coercivity of the perpendicular films discussed earlier. It can be seen that the initial uncovered Co film displays a strong in-plane anisotropy (negative K_1). As the Cu coverage is increased from 0 to 2 Å, the magnitude of this anisotropy energy rapidly approaches zero, indicating the presence of an increasing perpendicular contribution. This large increase in perpendicular anisotropy after Cu coverage is surprising in light of the very weak interface anisotropy displayed in Co/Cu multilayers.[1] One possibility is the existence of a large, in-plane Co/vacuum interface anisotropy that is being replaced by a perpendicular contribution from the forming Co/Cu interface. Such an in-plane vacuum interface anisotropy has recently been observed in fcc Co(100) films.[22] Work is in progress to determine directly this vacuum interface contribution in our (111)-oriented films.

Above 2 Å Cu coverage, the total anisotropy becomes increasingly negative indicating a reduction of the perpendicular contribution. One possible explanation of this phenomenon is the existence of a changing interfacial strain in the Co film due to the increasing overlayer thickness. As discussed earlier, we find no evidence from our RHEED data of any abrupt in-plane structural change greater than our resolution of $\sim 1\%$. However, changes in the out-of-plane atomic spacing, undetectable with RHEED, could influence the magnetic interface anisotropy. Detailed *in situ* structural characterization using low energy electron diffraction (LEED) is in progress to clarify these points.

Another possibility is that the magnetic interface anisotropy is very sensitive to details of the electronic band structure.[4] Recent photoemission measurements of Cu monolayers deposited on ferromagnetic transition-metals

have found electronic states that deviate significantly from bulk behavior.[23,24] If the hybridization of electronic states at the Co/X interface plays an important role in magnetic anisotropy, then variations of the overlayer band structure could cause significant alterations of the total anisotropy. It is interesting to note that the very recent photoemission measurements of 1 ML of Cu deposited on Co(0001) found the Cu peak at significantly lower binding energy than that of bulk (a shift of approximately 0.25 eV).[23] With further Cu coverage the bulk peak rapidly emerged, so that by 2 ML only the bulk Cu peak remained. This coverage dependence of the Cu electronic band structure is very similar to that which we observe for the magnetic anisotropy of our Co/Cu structures. This suggests a possible relation between the two phenomena, although more work is needed before this can be determined.

In conclusion, we have used *in situ* polar Kerr effect measurements to study the perpendicular magnetic behavior of MBE-grown Pd/Co/X sandwich structures where X=Ag, Cu, and Pd. For all three nonmagnetic overlayer materials, we find a rapid increase in the perpendicular anisotropy after only 2 Å ($\sim$1 ML) overlayer coverage. We find this perpendicular anisotropy is anomalously peaked at a coverage of $\sim$1 ML and significantly decreases with further overlayer deposition. The results reported here will provide a rigorous test for theoretical explanations of the magnetic anisotropy at surfaces and interfaces in layered metallic systems.

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