Spin Engineering in Ultrathin Cu/Co/Cu(110)

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We have studied the effect of depositing Cu overlayers onto CO gas dosed Co/Cu(110) ultrathin films ($6 < d_{\mathrm{Co}} < 30$ monolayers). We find that submonolayer Cu coverages can completely reverse the in-plane 90° easy axis switch caused by the CO adsorption for all Co thicknesses studied and for sufficiently thick Co films ($d_{\mathrm{Co}} > 15$ monolayers). This enables us to "controllably engineer" the direction of the easy axis at a constant Co thickness. [S0031-9007(98)05316-2]

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The magnetic properties of ultrathin films depend intimately on their nanostructure surface morphology and can be drastically altered by the addition of magnetic and nonmagnetic overlayers. For instance, it is now well established that magnetic moments can be enhanced at surfaces in comparison to bulk values and that a further enhancement of the magnetic moment can result for atoms in steps or in generally rough surfaces [1]. Buckley *et al.* [2,3] observed striking, nonmonotonic variations in the coercive field H_c and the ratio of remanent to saturation magnetization S in their M-H loops in the Co/Cu(100) system, upon the deposition of submonolayer quantities of Cu. This behavior was attributed to strong overlayer-induced electronic structure changes as Cu nucleated along surface step edges which were present due to a slight misorientation of the Cu crystal. Weber *et al.* went on to show that if the Co/Cu(100) system contained a deliberately more pronounced miscut then submonolayer coverages of Cu, Fe, Ag, or O could all cause the magnetic easy axis to switch 90°, from a direction parallel to the step edges to one perpendicular to the step edges [4–6]. Recent electronic structure calculations on the same system confirm the importance of the interaction of Cu atoms at Co step sites and show that hybridization effects can result in a noncollinear arrangement of magnetic moments, promoting switching of the easy axis [7].

However, the deliberately stepped Co/Cu(100) system is a highly specific geometry and therefore the question arises as to whether or not other nanostructure surface morphologies can promote switching of the easy axis. To this end we chose to study the Co/Cu(110) system because of its inherent 3D growth mode [8] and low symmetry surface. Furthermore, the unique symmetry of the (110) surface contains three important axes; the ⟨111⟩ axes which are the bulk magnetocrystalline easy axes of fcc Co, as well as the ⟨001⟩ and ⟨110⟩ axes which are the symmetry axes for uniaxial anisotropies. Therefore the (110) orientation allows us to differentiate between the cubic and the uniaxial anisotropy contributions due to different symmetry axes.

Previously we reported a striking time dependent evolution of the magnetic anisotropy of the Co/Cu(110) system [9]. For a given deposition of Co in the thickness range $5 < d_{\mathrm{Co}} < 40$ monolayers (ML), the adsorption of residual CO gas causes the magnetic easy axis to switch 90° from the [001] direction to the $[1\bar{1}0]$ direction over a period of time which is dependent on the thickness of the initial Co film. Dosing experiments reveal that only 0.08 ML of CO is required in order to switch the easy axis of a 15 ML Co film. The effect is chemically specific with oxygen, argon, and hydrogen being unable to switch the easy axis. STM measurements revealed that the 3D growth proceeded via the formation of elongated Co island structures preferentially oriented along the [001] direction. A completely new process for easy axis switching in the Co/Cu(110) system was identified [9]. It is found that a 1 ML Cu overlayer deposited before the CO induced easy axis switch will stop the switch from occurring. This is thought to be the result of the Cu atoms occupying the specific adsorption sites preferred by the CO molecules. The question then arises as to whether or not we can reverse the CO gas induced 90° easy axis switch with Cu overlayers.

In this paper we present the results of depositing submonolayer coverages of Cu onto the Co/Cu(110) system after the easy axis has been switched 90° from the [001] to the $[1\bar{1}0]$ direction by the residual CO in the UHV chamber. We find that Cu coverages can reverse the effect of the CO, thereby switching the easy axis back to the [001] direction. We have identified two distinct modes of switching related to the thickness of the Co film. For thin Co films ($d_{\mathrm{Co}} < 15$ ML), Cu overlayers cause the easy axis to switch back to the [001] direction abruptly. In thicker Co films ($d_{\mathrm{Co}} > 15$ ML) the easy axis is found to shift gradually from the $[1\bar{1}0]$ to the [001] direction, allowing us to controllably engineer the direction of the easy axis due to the presence of a significant cubic anisotropy K_1 in the thicker Co films. A phenomenological model has been developed to explain the two modes of switching. We show that the model is in agreement with *ex situ* Brillouin light-scattering (BLS) measurements by Hillebrands *et al.* [10–12] on the Au/Cu/Co/Cu(110) system which show an unexpected suppression of the magnetocrystalline anisotropy contribution below 50 Å Co, and that the magnetocrystalline contribution K_1

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becomes vanishingly small at Co thicknesses below $\approx$15 ML.

All experiments were carried out under UHV conditions with a base pressure of 1.0×10^{-10} mbar. The single crystal Cu(110) substrate was prepared via cycles of 1 kV Ar^+ sputtering and annealing to 700 K, until Auger electron spectroscopy and low energy electron diffraction measurements indicated a clean, well-ordered surface. Co was evaporated from an Omicron commercial electron beam evaporator with an integral flux monitor, at a constant rate of 0.4 ML min^{-1}. Cu was evaporated from a home made e-beam evaporator at a rate of 0.1 ML min^{-1}. The pressure remained below 5.0×10^{-10} mbar during deposition and the thickness of the Co and Cu films was evaluated from the relative heights of the Co and Cu Auger peaks recorded at the end of the experiment. This procedure gives an error in the absolute thickness of $\approx$25%. However, by keeping the evaporation rate constant, the relative thicknesses are known much more accurately.

To study the influence of Cu overlayers as a function of Co thickness we grew 6 and 30 ML Co films. In both films, the magnetic easy axis was found to switch from the [001] to the [1−10] direction over a period of about 1 and 2 hours, respectively, after the end of the Co deposition as minute quantities of residual chamber CO was adsorbed. Figure 1 (top and middle panels) shows the evolution of the magneto-optical Kerr effect (MOKE) loop squareness S for 6 and 30 ML Co films, respectively, as a function of Cu overlayer thickness. The loop squareness has been defined as the ratio of the remanent to saturation magnetization $S = M_{\text{rem}}/M_{\text{max}}$, where M_{max} is the loop amplitude measured from the maximum Kerr intensity. For the 6 ML Co film (Fig. 1, top panel), we see that, for fields applied parallel to [1−10], S remains at unity until about 0.9 ML of Cu has been deposited, at which time it drops sharply to $S = 0$ over the next tenth of a monolayer of Cu. We observe a mirror image of this behavior when the field is applied parallel to the [001] direction, with $S = 0$ until about 0.9 ML Cu then rising sharply to $S = 1$ over the next tenth of a monolayer of Cu as the easy axis switches to the [001] direction. This abrupt switching behavior contrasts with that observed for a 30 ML Co film; the behavior of which is shown in Fig. 1 (middle panel). Initially S is just below unity with the field applied parallel to [1−10], falling smoothly to $S = 0$ over a Cu deposition of 1 ML. When the field is applied parallel to [001], we see a smooth rise in S from $S = 0$ at 0 ML Cu to $S = 1$ at 1 ML Cu as the easy axis switches to the [001] direction.

Noting that the shape anisotropy keeps the magnetization in the plane of the film [10], we can model the magnetic anisotropy of the system phenomenologically as follows:

$$E_{\text{tot}} = \frac{K_1}{4}(\sin^4\varphi + \sin^2 2\varphi) + K_U^{\text{eff}} \cos^2\varphi\,, \quad (1)$$

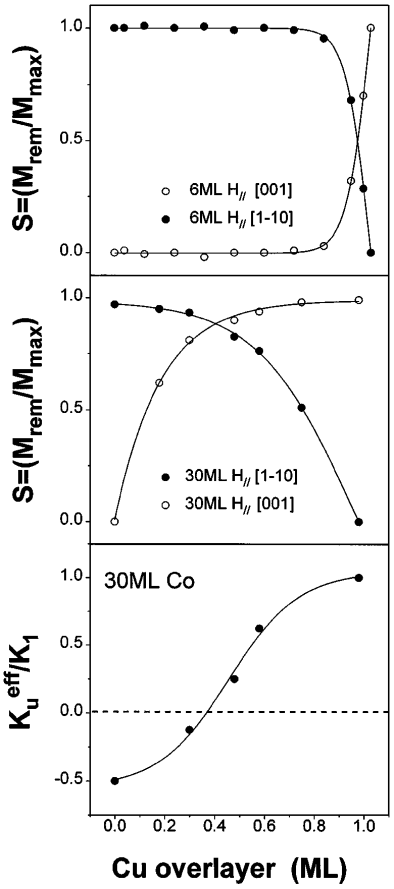

FIG. 1. (Top panel) the evolution of the loop squareness S as a function of Cu coverage on a CO dosed 6 ML Co/Cu(110) film with the field applied parallel to the [1−10] and [001] directions. The curves show that there is an abrupt switch of the easy axis after 0.9 ML of Cu is deposited. (Middle panel) the evolution of the loop squareness S as a function of Cu coverage on a CO dosed 30 ML Co/Cu(110) film. The smoothly rising and falling S curves can be interpreted as a gradual rotation of the easy axis from [1−10] to [001] due to the competing cubic and uniaxial anisotropies at this Co thickness. (Bottom panel) the calculated ratio of K_U^{eff}/K_1 for different orientations of the magnetic easy axis as a function of Cu overlayer thickness.

where K_1 is the cubic anisotropy constant and the angle φ is measured from the [001] axis. The second term is the effective in-plane uniaxial anisotropy term which is known to change sign from negative to positive as CO is adsorbed onto the Co. When K_U^{eff} is negative it favors the $\langle 001\rangle$ axes as easy and when it is positive it favors the $\langle 1-10\rangle$ axes. K_1 is known to be negative [10] and so the cubic term favors the $\langle 111\rangle$ axes. In our model we assume that the Cu overlayer is able to completely reverse the effect of the adsorbed CO gas and thereby change the sign of K_U^{eff} again and hence return the easy axis back to the [001] direction. It is possible that the Cu displaces the CO gas from the Co surface. If this is so, the CO will quickly be desorbed since the desorption temperature for CO on Cu is well

below room temperature [13]. The observation that the same amount of Cu (approximately 1 ML) is needed to completely switch the easy axis of both the 6 and 30 ML Co films supports this view. Our anisotropy model can be applied to the data in Fig. 1 (top panel) as follows. As Cu is deposited onto the surface of the Co-CO interface, K_U^{eff} tends to zero before changing sign. Therefore for a certain coverage of Cu we can reduce K_U^{eff} to zero leaving only the cubic anisotropy term in Eq. (1); the easy axis would shift to the $\langle 111 \rangle$ axes when $K_U^{\text{eff}} = 0$. However, for a gradual shift of the easy axis to the $\langle 111 \rangle$ axes, S would not remain at unity as is observed. The fact that S remains at unity at all Cu coverages, except briefly at the sharp crossover region, implies that the cubic anisotropy K_1 is negligible for a Co thickness of 6 ML. Therefore our results are in agreement with the *ex situ* BLS measurements of Hillebrands *et al.* which show that the cubic anisotropy component of the total anisotropy is vanishingly small for Co thicknesses below 15 ML [10] in the Au/Cu/Co/Cu(110) system. When $K_U^{\text{eff}} = 0$ the sample becomes magnetically isotropic with no preferred easy axis and further Cu deposition will establish a small negative K_U^{eff}, and thereby immediately switch the easy axis to the [001] direction. Figure 2 shows the result of an experiment that tries to reproduce this magnetically isotropic state in a 6.5 ML Co film. After a Cu deposition of 0.96 ML, angular dependent studies show that the surface is almost perfectly isotropic with no preferred easy axis direction, and therefore we believe that 0.96 ML of Cu has reduced K_U^{eff} to zero.

Since K_1 becomes comparable to K_U^{eff} for Co thicknesses greater than 15 ML [10], the smoothly varying S curves in the 30 ML sample (Fig. 1, middle panel) can be interpreted as due to a gradual rotation of the easy axis in-plane due to the changing competition between K_U^{eff} and K_1 as Cu is deposited. Figure 3 shows the direct observation of this competition for a 30 ML Co film. The figure clearly shows the shift of the easy axis direction (indicated by the square loops with thicker lines) from $[1-10]$ to 60° to 45° to 30° to [001] as a function of Cu thickness. For each Cu thickness a full angle dependent MOKE study was performed to study the changing anisotropy. The middle panel of Fig. 3 shows that a deposition of 0.48 ML of Cu has created a fourfold surface anisotropy with 45° being the easy axis and $[1-10]$ and [001] being almost equivalent hard axes. This is the first time to our knowledge that anyone has been able to controllably engineer the direction of the easy axis of magnetization at a constant thickness in an ultrathin film. This is made possible by the unique properties of the twofold symmetry of the Co/Cu(110) surface which exhibits different symmetry axes for the uniaxial and cubic anisotropies, and by the ability to controllably change the sign of the effective in-plane uniaxial anisotropy.

To gain further insight into the changing anisotropy shown in Fig. 3, we can express the equilibrium angle of the magnetization φ_0 in terms of the anisotropy coefficients K_1 and K_U^{eff} by minimizing Eq. (1) with respect to φ,

$$\frac{K_U^{\text{eff}}}{K_1} = \left(1 - \frac{3}{2}\sin^2\varphi_0\right) = r\,. \tag{2}$$

Since we have experimentally observed the equilibrium angle of the magnetization φ_0 as a function of Cu thickness (i.e., at 90°, 60°, 45°, 30°, and 0°), we can calculate the ratio of the uniaxial to cubic anisotropy coefficients from Eq. (2) for each easy axis orientation observed in Fig. 3. This of course assumes that the bold loops in Fig. 3 are perfectly square, with unity remanence indicating that the easy axis is exactly along these directions. Figure 3 shows that this is a reasonable approximation and is sufficient for our rough calculation. Figure 1 (bottom panel) shows the result of this calculation for the 30 ML Co film. Assuming that the Cu overlayer does not influence the cubic anisotropy so that K_1 is always constant, then Fig. 3 clearly shows the change in sign of the effective in-plane uniaxial anisotropy constant as the easy axis is shifted gradually from $[1-10]$ ($r = -0.5$) to [001] ($r = 1$). We

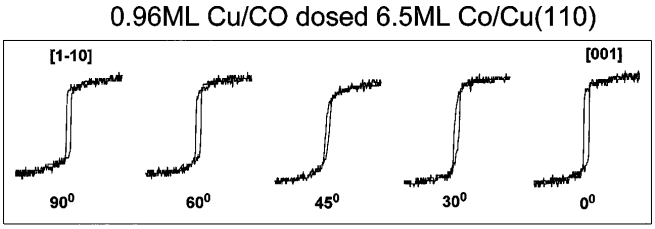

FIG. 2. An angle dependent MOKE study of a 0.96 ML Cu/CO dosed 6.5 ML Co/Cu(110) film. Since K_1 is negligible at this thickness, it is possible to engineer a magnetically isotropic surface with no preferred easy axis direction by reducing K_U^{eff} to zero with an appropriate deposition of Cu. Further Cu deposition creates a negative K_U^{eff}, reestablishing the easy axis along the [001] direction.

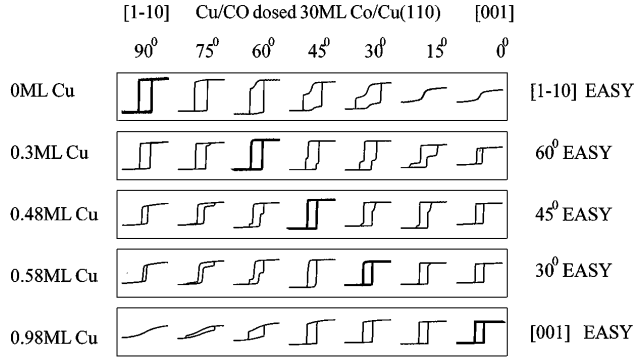

FIG. 3. Angle dependent MOKE studies of Cu/CO dosed 30 ML Co/Cu(110) film as a function of Cu overlayer thickness. The top panel shows that initially the easy axis is almost along the $[1-10]$ direction. As Cu is deposited the easy axis direction (indicated by the square loops with thicker lines) can be seen to shift to 60°, 45°, 30°, and finally back to [001].

see that K_U^{eff} needs to be twice as strong to produce an easy axis loop along [001] as it does to produce an easy axis loop along $[1-10]$. Furthermore, Fig. 1 (bottom panel) indicates that $K_U^{\text{eff}} = 0$ at a Cu coverage of 0.37 ML on the CO dosed 30 ML Co film. This contrasts with the data in Fig. 2 which implies $K_U^{\text{eff}} = 0$ for a Cu coverage of 0.96 ML on the CO dosed 6.5 ML Co film, and therefore we need 2.6 times more Cu to reduce K_U^{eff} to zero in the thinner Co film than in the thicker Co film. This can be explained if we assume that the inherent uniaxial anisotropy field before CO gas adsorption is stronger along the [001] direction in the thicker Co film than in the thinner film. Then, for the thicker Co film, it is more difficult for the CO gas to overcome this uniaxial anisotropy field. Hence, when the easy axis is switched in the thick Co film, the new uniaxial anisotropy field is relatively weaker in the $[1-10]$ direction in comparison to the case for the thin Co film. Therefore less Cu overlayer is needed to reverse the sign of K_U^{eff} for thicker Co films, although both Co thicknesses still need the same amount of Cu overlayer to completely displace all of the adsorbed CO gas, as is consistent with the data of Fig. 1. This result agrees well with *ex situ* BLS work by Hillebrands *et al.* which shows a twofold increase in the in-plane uniaxial anisotropy along the [001] direction over the same thickness range [10]. This may also explain why it takes twice as long for the residual CO gas to switch the easy axis in the 30 ML film as it does for the 6 ML Co film.

In summary, we have deposited submonolayer coverages of Cu onto the Co/Cu(110) system which has already undergone a 90° switch of the easy axis from the [001] to the $[1-10]$ direction due to the adsorption of minute quantities of residual CO from the UHV environment. We find that the Cu is able to completely reverse the effect of the CO thereby switching the easy axis back to the [001] direction. Two distinct modes of switching are observed depending on the thickness of the Co film and hence the size of the cubic anisotropy constant K_1. For $d_{\text{Co}} = 6$ ML, $K_1 = 0$ and the switch is found to be abrupt. For this thickness we are able to engineer a magnetically isotropic surface with no preferred easy axis direction by reducing the effective uniaxial anisotropy K_U^{eff} to zero at a certain Cu coverage. For $d_{\text{Co}} = 30$ ML, K_1 becomes comparable to K_U^{eff} and the competition between the two anisotropies as Cu is deposited allows us to controllably engineer the direction of the easy axis from the $[1-10]$ to the [001] direction at a constant Co thickness.

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[1] M. Albrecht *et al.,* Europhys. Lett. **20**, 65–70 (1992).
[2] M. E. Buckley *et al.,* J. Phys. Condens. Matter **8**, L147–L152 (1996).
[3] M. E. Buckley *et al.,* Phys. Rev. B **52**, 6596 (1995).
[4] W. Weber *et al.,* Phys. Rev. B **52**, R14 400 (1995).
[5] W. Weber *et al.,* Phys. Rev. Lett. **76**, 3424 (1996).
[6] W. Weber *et al.,* Nature (London) **374**, 788 (1995).
[7] A. V. Smirnov *et al.,* Phys. Rev. B **54**, R17 371 (1996).
[8] M. T. Kief and W. F. Egelhoff, Jr., Phys. Rev. B **47**, 10 785 (1993).
[9] S. Hope *et al.,* (to be published).
[10] B. Hillebrands *et al.,* Phys. Rev. B **53**, R10 548 (1996).
[11] J. Fassbender *et al.,* J. Appl. Phys. **76**, 6100 (1994).
[12] J. Fassbender *et al.,* J. Magn. Magn. Mater. **148**, 156 (1995).
[13] F. Falo, I. Cano, and M. Sakmeron, Surf. Sci. **143**, 303–313 (1984).