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Magnetic anisotropy in epitaxial Co superlattices

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A series of epitaxial Co-Au and Co-Cu superlattices has been grown by molecular-beam-epitaxy techniques, with Co thicknesses varying from 5 to 40 Å and constant Au and Cu thicknesses of 16 and 25 Å, respectively. We compare the magnetic properties of both types of superlattice. Crossover to an easy axis normal to the film plane occurs at a thickness of 19 Å of Co in the Co-Au series and 10 Å of Co in the Co-Cu series. We show that the observed magnetic behavior of both of these systems can be accounted for by contributions from demagnetization, magnetocrystalline anisotropy, and magnetoelastic anisotropy. The magnetoelastic term depends on the strain present in Co layers, which in turn is a function of the Co-layer thickness. In our calculation we employ *measured* Co-layer strains, determined by x-ray scattering, in combination with the known elastic and magnetostriction constants of bulk Co.

The magnetic properties of surfaces and thin films have been a topic of interest for several decades. With the advent of new deposition techniques, particularly those carried out in ultrahigh-vacuum conditions such as molecular-beam epitaxy (MBE), studies of novel magnetic structures are now quite feasible down to the atomic scale.[1] The ability to control the local atomic environment has led to extensive work on tailor-made systems, for example, sandwich structures,[2] magnetic multilayers,[3] and superlattices.[4-10] The latter materials are very interesting because they exhibit interfacial magnetic properties on a macroscopic scale. In addition to interest in the fundamental behavior of such systems, an increasing amount of attention is directed towards applications of such materials in magneto-optical recording technology.[11]

Much of the recent work has aimed at a better understanding of magnetic anisotropy, in particular, the tendency of the easy axis of magnetization to be oriented perpendicular to the plane of the layers. In fact, certain calculations of the electronic structure at the interface result in a favored (perpendicular) spin orientation.[12] While such calculations have been successful in certain specific cases, there are at present no first-principles derivations of perpendicular anisotropy for arbitrary layered systems. In the absence of such treatments, experimentalists are guided by phenomenological approaches which separately consider contributions to the magnetic anisotropy such as those from magnetocrystalline, magnetoelastic, and shape (demagnetization) anisotropy energies. These anisotropy terms have been well characterized, for hcp Co, via measurements performed on bulk samples.

The magnetoelastic contribution depends upon the Co-lattice strain present in a given sample. While previous workers such as Chappert and Bruno[13] have recognized the importance of this term, they have been limited to estimates of the level of strain through recourse to critical-thickness models. In this Rapid Communication we use measured values of epitaxial strains, determined by x-ray-scattering methods, in order to estimate the magnetoelastic component of the anisotropy. We will show that a simple calculation involving these measured strains and the bulk properties of hcp Co can account quantitatively for the observed anisotropy in Co-Au superlattices. The Co-Cu superlattices provide an interesting case for comparison, since they present a system with much smaller lattice mismatch at the Co-noble metal interface (2% in this case, in comparison to 14% for Co-Au). In addition, as we shall discuss below, the Co layers occur in the fcc (rather than hcp) stacking symmetry in Co-Cu. We will show that the magnetic anisotropy in Co-Cu superlattices can be accounted for by a reduction of the magnetocrystalline anisotropy relative to that of hcp Co.

Co-Au and Co-Cu superlattices were prepared in ultrahigh vacuum using a Vacuum Generators V-80 MBE system with a background pressure of better than 10^{-10} mbar. In both series Co thicknesses varied from 5 to 40 Å while Au and Cu thicknesses were held fixed at 16 and 25 Å, respectively. The total superlattice thicknesses were 1500 Å in all cases. Growth of the superlattices and their characterization by reflection high-energy electron diffraction, x-ray diffraction, and transmission electron microscopy have been reported elsewhere.[9] X-ray-diffraction studies of these films show hcp (0001) growth of Co in the Co-Au system[9] and fcc (111) growth of Co in the Co-Cu system.[14] Detailed analyses of the x-ray-scattering intensities confirm that the interfaces in these samples are compositionally abrupt with interpenetration limited to a two monolayer region.[9,15]

The magnetic measurements discussed below were carried out in a Quantum Design superconducting quantum interference device (SQUID) magnetometer. Hysteresis loops were measured with applied fields of up to 15 kOe both parallel and perpendicular to the substrate plane at sample temperatures ranging from 5 to 300 K. Although the samples were fully magnetized at a field of less than 10 kOe, the value of the saturation magnetization M_s was determined at a magnetic field of 30 kOe at 5 K in order to ensure complete magnetization. The largest error in the measurement of M_s is due to the uncertainty in the sample volume which is estimated to be ~10%. All of our samples exhibit a saturation moment which is reduced relative to the value for bulk Co (1440 emu/cm^3). In the

Co-Au series the sample with 5-Å Co layers has a saturation moment of 892 emu/cm^3, while the remaining samples have moments which range from 1267 to 1393 emu/cm^3, with an average of 1333 emu/cm^3. In the Co-Cu series, the saturation moments range from 1160 to 1302 emu/cm^3, with an average of 1241 emu/cm^3.

Figure 1 shows hysteresis loops measured at 300 K with applied fields both parallel and perpendicular to the film plane. It is clear from Fig. 1 that the easy axis of magnetization lies out of the film plane at a Co-layer thickness of 5 Å. The effective anisotropy K_{eff} is defined as the area between the perpendicular and the parallel magnetization curves, $M^{\perp}$ and $M^{\parallel}$, divided by the total Co volume:

$$K_{\text{eff}} \equiv \frac{1}{V}\int_0^{H_{\text{sat}}} (M^{\perp} - M^{\parallel})\,dH \,. \qquad (1)$$

K_{eff} is positive when the magnetization is preferentially oriented perpendicular to the film. Using a phenomenological approach, one may express the effective anisotropy as

$$K_{\text{eff}} = 2K_S/t_{\text{Co}} + K_V \,, \qquad (2)$$

where t_{Co} is the Co-layer thickness, K_S is an interface anisotropy, K_V is the volume anisotropy, and the factor of 2 arises from the two interfaces of each Co layer. If K_S and K_V are constants, Eq. (2) leads to a linear plot of $K_{\text{eff}} t_{\text{Co}}$ against t_{Co}, with an intercept of $2K_S$ at $t_{\text{Co}} = 0$ and a crossover to perpendicular anisotropy ($K_{\text{eff}} = 0$) at the critical Co-layer thickness of $-2K_S/K_V$. This treatment is not entirely satisfactory. For example, it says little about the origin of K_S and moreover, does not reproduce the behavior at small thicknesses where a saturation or even a falloff[16] in $K_{\text{eff}} t$ is often observed.

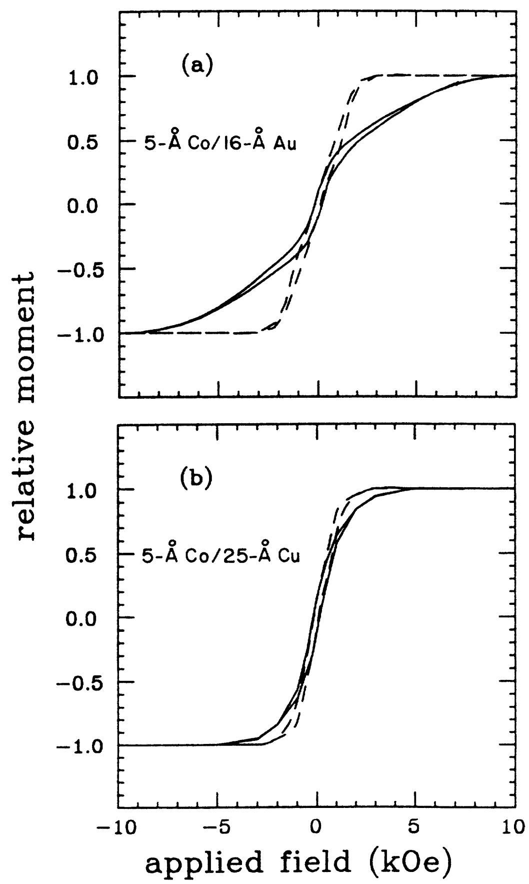

FIG. 1. Hysteresis loops of (a) 5-Å Co–16-Å Au and (b) 5-Å Co–25-Å Cu superlattices measured at 300 K with the applied magnetic field parallel (solid curves) and perpendicular (dashed curves) to the film plane.

We show here that it is not necessary to invoke a surface-anisotropy term when the magnetoelastic anisotropy is a function of the Co-layer thickness. That is, one can express the total anisotropy as

$$K_{\text{eff}} = K_{\text{MC}} + K_D + K_{\text{ME}}(t_{\text{Co}}) \,. \qquad (3)$$

K_{MC} is the magnetocrystalline anisotropy. For an hcp-Co film at 300 K, with the c axis normal to the film, $K_{\text{MC}} = K_1 + K_2 = 5.56\times10^6$ (erg/cm^3). K_D is the demagnetization term and, in the thin-film geometry, is related to the saturation magnetization by $K_D = -2\pi M_s^2$. In our calculations of K_D we will use the average (measured) saturation moments which were given above. The magnetoelastic anisotropy $K_{\text{ME}}(t_{\text{Co}})$ is obtained from the magnetoelastic energy $E_{\text{ME}} = -(\tilde{c}\boldsymbol{\epsilon})\cdot\boldsymbol{\lambda}$, where $\tilde{c}$ is the elasticity tensor, $\boldsymbol{\epsilon} = (\epsilon_1, \epsilon_2, \epsilon_3)$ is the epitaxial strain, and $\boldsymbol{\lambda}$ is the magnetostriction strain. The measured in-plane strain ϵ is set equal to the in-plane components ϵ_1 and ϵ_2. Then the strain energy $\frac{1}{2}(\tilde{c}\boldsymbol{\epsilon})\cdot\boldsymbol{\epsilon}$ is minimized to obtain $\epsilon_3 = -2\epsilon c_{13}/c_{33}$. The resultant anisotropy for a (0001) hcp film is

$$K_{\text{ME}} = -\left(c_{11} + c_{12} - \frac{2c_{13}^2}{c_{33}}\right)(\lambda_A + \lambda_B)\,\epsilon(t_{\text{Co}}) \,. \qquad (4)$$

Using elastic constants[17] c_{ij} and magnetostriction constants[18] ($\lambda_A = -50\times10^6$ and $\lambda_B = -107\times10^6$) which have been experimentally determined for bulk hcp Co, we obtain $K_{\text{ME}} = 6.484\times10^8\epsilon$ (erg/cm^3). The total effective anisotropy is then

$$K_{\text{eff}} = 5.56\times10^6 - 2\pi M_s^2 + 6.484\times10^8\epsilon(t_{\text{Co}})\ (\text{erg/cm}^3) \,. \qquad (5)$$

From the signs of the three anisotropy contributions we see that K_D favors an in-plane spin orientation, while K_{MC} and K_{ME} (for tensile strain) favor an out-of-plane alignment. If one assumes that K_D and K_{MC} are constant, regardless of the Co-layer thickness, then their contribution to a plot of $K_{\text{eff}} t_{\text{Co}}$ vs t_{Co} will generate a line with a slope given by $K_{\text{MC}} + K_D = 5.56\times10^6 - 2\pi M_s^2$ (erg/cm^3). On the other hand, K_{ME} will have a thickness dependence through the strain $\epsilon(t_{\text{Co}})$, which will in general decrease as the Co-layer thickness t_{Co} increases. The *critical strain* ϵ_c, at which a crossover to perpendicular anisotropy occurs, is determined by the condition $K_{\text{eff}} = 0$. For an hcp Co film oriented along [0001], Eq. (5) yields $\epsilon_c = 0.0115$, using the saturation magnetization of bulk Co (1440 emu/cm^3).

We can apply Eq. (5) to the case of Co-Au superlattices, using measured values of the Co-layer strain. We have obtained these strain values from x-ray scattering measurements with the scattering vector lying in the plane of the film, specifically from scans through the Co $(11\bar{2}0)$ peak.[15,19] The strain values for superlattices with Co-layer thicknesses of 10, 20, and 30 Å were fitted with a straight line (Fig. 2, inset) which we use to parametrize the Co-layer strain over the thickness range of 5–35 Å. The use of a *linear* fit has no particular physical meaning; we use it since the available data are limited to three points with significant error bars. The calculated value of $K_{\rm eff}$ in Fig. 2 (solid curve) was obtained by substituting the strain values $\epsilon(t_{\rm Co})$ from the linear fit, along with the average measured value of M_s (1333 emu/cm^3), into Eq. (5). We find that this calculated anisotropy agrees quite well with the measured anisotropies, given that we have included only bulk constants, measured strains, and measured saturation moments, with no adjustable parameters.

The measured anisotropies indicate that the critical thickness for the Co-Au system is ~ 19 Å. This value is similar to that reported for a Au-Co-Au sandwich,[20] and is somewhat higher than the value of 14 Å reported for sputtered and subsequently annealed Co-Au superlattices.[16] We note that our magnetization results were obtained without any annealing treatment. In fact, we have investigated the effect of annealing on superlattice x-ray-diffraction peaks and found that any annealing in these MBE-grown samples only led to a decrease of both superlattice peak intensities and perpendicular anisotropy.

Magnetometer measurements were also used to determine the effective anisotropies in a series of Co-Cu samples (Fig. 3). The perpendicular crossover thickness in this case (~ 10 Å) is smaller than in the Co-Au series. As above, the in-plane epitaxial strains were determined by x-ray measurements (inset). In this case, however, the Co and Cu layers are coherent in the plane of the superlattice.[14] By coherent we mean that the Co and Cu layers occur with the same in-plane lattice parameter, with no misfit dislocations at the layer interfaces. One may estimate the strains in the Co layers, at a given thickness $t_{\rm Co}$, by minimizing the strain energy, which is proportional to $t_{\rm Co}\epsilon_{\rm Co}^2+t_{\rm Cu}\epsilon_{\rm Cu}^2$, assuming that Co and Cu have similar elastic moduli. This form of the strain energy yields a Co layer strain given by $\epsilon_{\rm Co}=mt_{\rm Cu}/(t_{\rm Co}+t_{\rm Cu})$, where m is the lattice misfit between Co and Cu. We have fit the measured strains with a function of this form (Fig. 3, inset) and used this fit, along with the measured average saturation magnetization (1241 emu/cm^3) in calculating the total anisotropy in the Co-Cu case. However, we note that previous x-ray measurements[14] have shown that the close-packed Co monolayers in the Co-Cu samples are primarily stacked according to the fcc (rather than the hcp) sequence. Thus, in applying Eq. (5) to the Co-Cu case, we must keep in mind that the magnetocrystalline anisotropy and magnetoelastic constants of hcp Co are not expected to be appropriate. (Such constants are not available for fcc Co at room temperature.) Nonetheless the application of Eq. (5) yields interesting results. The dashed curve in Fig. 3 is obtained by using Eq. (5) as written above. The solid curve was obtained by reducing the magnetocrystalline term to 20%, and the magnetoelastic term to 82%, of the values given for hcp Co. That is, Eq. (5) was modified to

$$K_{\rm eff}=1.112\times10^6-2\pi M_s^2+5.312\times10^8\epsilon(t_{\rm Co}) ,$$

where $M_s=1241$ emu/cm^3. The modified terms in Eq. (5), which we have used in fitting the Co-Cu data, indicate that the magnetostrictive properties are more or less unchanged, relative to the hcp-Co case. The magnetocrystalline anisotropy, on the other hand, is reduced significantly. This result is certainly plausible, given pre-

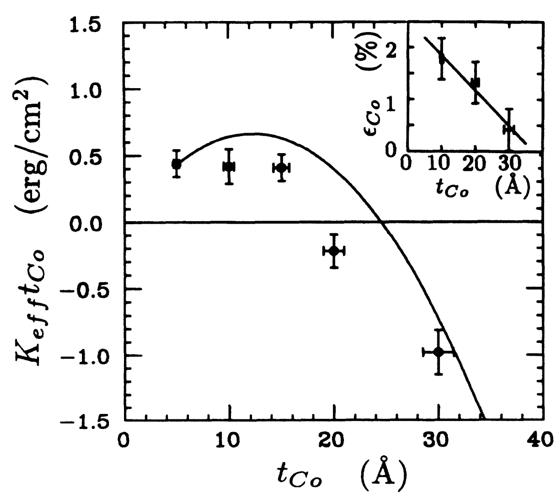

FIG. 2. Measured (points) and calculated anisotropies for a series of Co-Au superlattices. The calculation (solid curve) employed the Co-layer strains given by the linear fit to the measured strains shown in the inset. The magnetocrystalline anisotropy and magnetoelastic constants used here are those of hcp Co.

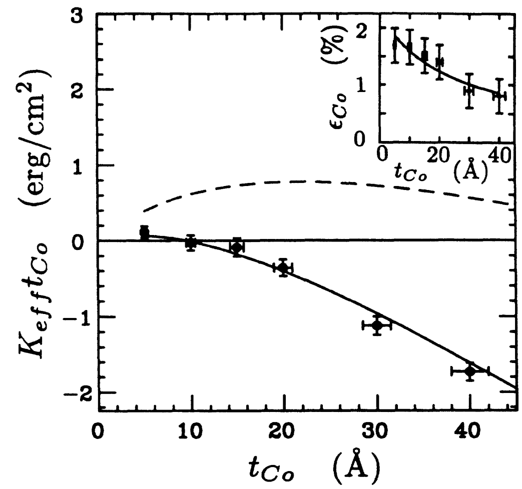

FIG. 3. Measured (points) and calculated anisotropies for a series of Co-Cu superlattices. One calculation (dashed curve) was performed using the magnetocrystalline anisotropy and magnetoelastic constants of hcp Co. The second calculation (solid curve) was performed with modified constants, as described in the text. Both calculations employed the Co-layer strains given by the fit to the measured strains (inset).

vious measurements of the magnetocrystalline anisotropy in bulk fcc Co in its stable form, above 400 °C. In this latter case it is found that a spin alignment along [111] is favored, but the magnitude of the anisotropy energy is substantially reduced, relative to that of hcp Co.[21]

In conclusion, we have shown that the measured strains in two series of Co superlattices can adequately account for their observed magnetic anisotropies through the introduction of a thickness-dependent magnetoelastic anisotropy. In this approach the interface enters only as the source of an epitaxial strain which exists throughout the entire Co layer. Although our measurements do not rule out the existence of a true interface anisotropy, we show that in this case it is not necessary to invoke such a term.

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