Transitions in the direction of magnetism in Ni/Cu(001) ultrathin films and the effects of capping layers

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Ultrathin films of nickel exhibit an unusual sequence of transitions from in-plane to perpendicular magnetization as a function of film thickness. A sharp transition from in-plane to perpendicular magnetization is found near 7 ML thickness, followed by a gradual transition back to in-plane magnetization beginning at 37 ML. This sequence of transitions cannot be explained by the surface or shape anisotropies, both of which favor in-plane magnetization in the thickness range where perpendicular anisotropy is found. We have measured the thickness dependence of these transitions for nickel film wedges, and films capped by nonmagnetic and magnetic overlayers, to experimentally determine the surface, interface, and magnetoelastic anisotropies. We find that both the surface and interface anisotropy constants are negative (favoring in-plane magnetization), with the magnitude of the surface term being larger than that of the interface. A correlation is found between the critical thickness for misfit dislocation formation in the nickel film and a sharp transition in the coercive field. This transition is used to accurately determine the onset of a thickness dependence in the bulk magnetoelastic energy, which causes the magnetization to rotate back into the film plane. This model gives a complete description of the mechanism for the easy-axis changes at both the 7 and 37 ML thicknesses. [S0163-1829(96)01937-6]

I. INTRODUCTION

Ultrathin films and multilayers possessing an easy axis of magnetization perpendicular to the surface have recently attracted much interest[1–8] due to their potential advantages in magneto-optical recording. The existence of perpendicular magnetization is determined by the combined effects of all the magnetic anisotropies present. These anisotropies include shape, surface, interface, and crystalline anisotropies, strain-induced magnetoelastic anisotropy, and anisotropies due to roughness and atomic mixing at the interfaces. An important goal in the research on these thin films and multilayers is to understand which anisotropies are important in determining the easy axes of magnetism.

In the past few years a number of studies have been performed on the magnetism of Ni/Cu(001) thin films.[1–3] Ni/Cu(001) films show a transition from in-plane magnetization to magnetization perpendicular to the surface as the film thickness increases. This unusual behavior is opposite to what would be predicted by the effects of a surface anisotropy by itself. Schulz and Baberschke,[2] using ferromagnetic resonance, showed that the unusual magnetic behavior of Ni/Cu(001) could be explained by competition between a magnetoelastic volume anisotropy due to pseudomorophic growth, the shape anisotropy and the sum of the surface plus interface anisotropies which favor in-plane magnetization. Separation of the interface and surface anisotropies is difficult experimentally and the relative importance of their contributions toward determining the direction of magnetization is not presently known. For Ni films less than ~7 ML thick the combination of the surface and interface anisotropies dominate and the films have an in-plane easy axis of magnetization. Films thicker than ~7 ML have an easy axis of magnetization perpendicular to the surface due to the volume effect of the magnetoelastic anisotropy which favors perpendicular magnetization. As the film thickness exceeds the critical thickness for pseudomorphic growth, strain relief diminishes the magnitude of the magnetoelastic anisotropy energy and the magnetization direction changes back to in-plane. The details of the transition back to in-plane magnetization are not as well understood as the transition which takes place near 7 ML.

In this paper we discuss the results of experiments designed to more fully understand the magnetic anisotropies in Ni/Cu(001) thin films. Specifically, we are interested in two details in the magnetism of Ni/Cu(001) which are not well understood. The first is in the region of film growth beyond the pseudomorphic region and the second is the relative importance of the surface and interface anisotropy. Ni wedges grown on Cu(001) are used to measure the direction of magnetization in Ni thin films as a function of film thickness. Between 4 and 7 ML the Ni films magnetize in plane. A sharp transition to perpendicular magnetization occurs at 7 ML, and a gradual transition back to in-plane magnetization begins at 37 ML. By measuring changes in the coercive field with film thickness we determine the critical thickness for epitaxial growth of Ni on Cu(001), d_c, to be 13 ML. Using this value of d_c and the residual strain model developed by Chappert and Bruno[9] we are able to predict the film thickness for the second transition to within 15%. Both transitions in the easy axis of magnetization are now well understood in terms of the magnetoelastic, shape, and surface plus interface anisotropies. Similar measurements, made on a Cu/Ni-wedge/Cu(001) sandwich structure, allows the separa-

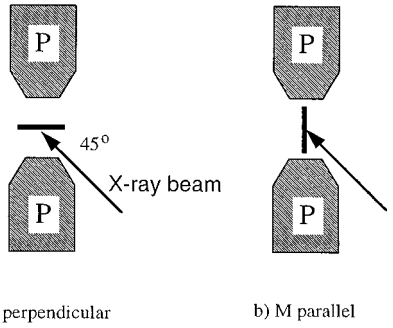

FIG. 1. Experimental geometry for (a) magnetization perpendicular to the sample surface and (b) parallel to the sample surface. The sample is rotated 90° to change the measurement geometry. The electromagnet *P* produces a maximum field of 800 Oe at the sample position. Circularly polarized photons from the synchrotron are incident at a 45° angle for both geometries.

tion of the surface and interface anisotropies. We find that both the interface and surface anisotropies are negative with the magnitude of the surface anisotropy being larger.

With quantitative knowledge of the anisotropy constants it should be possible to alter, in a controlled manner, the magnetization direction in Ni/Cu(001) thin films. In an initial attempt to do so we have studied the effects which ferromagnetic capping layers have on the direction of Ni magnetization. We find that the addition of 2 ML of Co causes the magnetization to lie in-plane for Ni thicknesses up to at least 18 ML. The addition of 2 ML of Fe has no measurable effect on the magnetization direction of Ni. These results are discussed in terms of changes in the surface and shape anisotropy energies due to the capping layers.

II. EXPERIMENT

The experiments were performed on the 10 m TGM beamline located at the Synchrotron Radiation Center, Stoughton, WI. All magnetic measurements were made using x-ray magnetic circular dichroism (XMCD) which gives element specific magnetic information. The XMCD signal, $\sigma_M=\sigma_+-\sigma_-$, is the difference between the x-ray-absorption spectra with the photon spin vector Σ parallel (σ_+) and antiparallel (σ_-) to the sample magnetization. The absolute value of the XMCD intensity at the L_3 edge, $|\sigma_M(L_3)|$, can be used to monitor the degree of long-range magnetic ordering, since it is proportional to the net magnetization along the direction of the photon spin (Poynting vector), $|\sigma_M(L_3)|\propto\mathbf{M}\cdot\mathbf{\Sigma}$. By normalizing $|\sigma_M(L_3)|$ to the total absorption cross section at L_3, $\sigma_0(L_3)=1/2[\sigma_+(L_3)+\sigma_-(L_3)]$, we obtain an important intensive quantity: the magnetization or degree of ferromagnetic ordering on a per atom basis.

XMCD measurements were made on Ni wedges grown at room temperature on a clean Cu(001) single crystal at a base pressure of 2×10^{-10} Torr. The pressure during film growth was less than 8×10^{-10} Torr. Five different wedges, with overlapping thickness ranges were used in this study. Film structure and orientation was observed by low-energy electron diffraction. Two geometries were used for the XMCD measurements, as shown in Fig. 1. To measure magnetization perpendicular to the surface, the sample normal was aligned parallel to the applied magnetic field and the photon angle of incidence on the sample was 45° [Fig. 1(a)]. To measure magnetization parallel to the surface, the sample normal was aligned perpendicular to the magnetic field and the photon angle of incidence on the sample was again 45° [Fig. 1(b)].

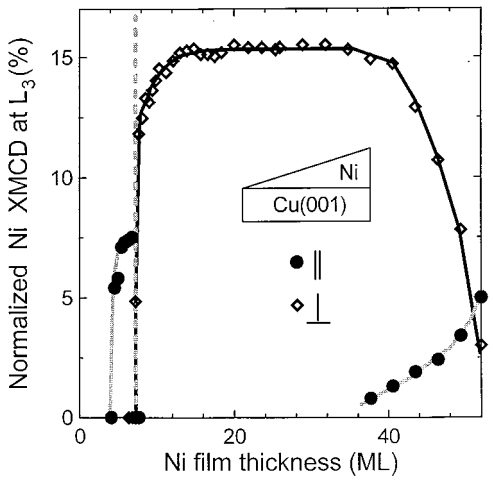

FIG. 2. Normalized remanent dichroism intensity for magnetization both in-plane ∥ and perpendicular ⊥ to the surface for a series of Ni wedges grown on Cu(001). There is sharp transition from an in-plane to a perpendicular easy axis of magnetization at 7 ML and a gradual transition from a perpendicular to an in-plane easy axis of magnetization beginning near 37 ML.

XMCD spectra were taken at room temperature by switching the direction of the magnetic field and measuring the total electron yield while sweeping the incident photon energy at a fixed polarization. Element specific hysteresis measurements were made by varying the magnetic field while measuring the yield at a photon energy fixed at the L_3 maximum. The easy axis of magnetization was determined by measuring hysteresis curves for both geometries of Fig. 1, and determining the remanent magnetization M_R. Square hysteresis curves are obtained when the magnetic field is aligned along the easy axis and zero remanence hysteresis curves are obtained when the easy axis is orthogonal to the applied magnetic field.

III. RESULTS

A. Ni/Cu(001): transitions in the direction of magnetization

In Fig. 2 the normalized dichroism intensity at remanence for both in-plane and perpendicular magnetization is plotted vs film thickness. Below 7 ML the Ni films have in-plane remanent magnetization only, no remanent magnetization is detected for fields applied perpendicular to the surface. At 7 ML there is a sharp transition to magnetization perpendicular to the surface, no in-plane remanent magnetization is found. Beginning at 37 ML the direction of magnetization begins to switch back to in-plane with remanent magnetization found both in-plane and perpendicular to the surface. The results in Fig. 2 are compiled from measurements made on five Ni wedges with overlapping thickness ranges.

The total anisotropy energy density, ignoring crystalline anisotropies, of a uniformly magnetized ultrathin film can be represented as $E=K^{\text{eff}}\sin^2\theta$, with

$$K^{\text{eff}}=-2\pi M^2+K_{\text{ME}}+\frac{K_I+K_S}{d}, \tag{1}$$

where K_{ME}, K_S, and K_I are the magnetoelastic, surface, and interface anisotropy constants respectively, d is the film thickness, and θ is the angle between the magnetization and surface normal. Perpendicular magnetization in the Ni/Cu(001) films will result when

$$K_{\text{ME}}+\frac{K_S+K_I}{d}>2\pi M^2. \tag{2}$$

The surface and interface anisotropy energies per unit volume, K_S/d and K_I/d, respectively, are thickness dependent, and are difficult to separate experimentally. At room temperature, $K_{\text{ME}}=3.95\times10^6$ ergs/cm^3 in the pseudomorphic region and $K_I+K_S=-0.38$ ergs/cm^2.[2]

The use of Eq. (2) requires a knowledge of the dependence of T_C on film thickness, and of M on T/T_C. The magnetization of Ni ultrathin films on Cu(100) exhibits a power-law behavior, $M(T)=M_0(1-T/T_C)^{\beta}$, where both the Curie temperature and critical exponent β are film thickness dependent. A study of Ni films on Cu(100) and Cu(111) has been completed by Huang *et al.*,[3] who have compiled these values for films below 16 ML in thickness. Their results show that the power-law expression is valid down to room temperature. For films thicker than 16 ML, no experimental data is available other than the bulk Ni values, so we use a simple extrapolation to connect the ultrathin film values to the bulk. Using these experimental values to determine M at room temperature we find that the transition from in-plane to perpendicular magnetization should take place near 6 ML at room temperature. This is in excellent agreement with our experimental results shown in Fig. 2 (also see Fig. 6, below) and with the results of Schulz and Baberschke.[2]

The magnetoelastic anisotropy K_{ME} is strain (ε) dependent and can be written as $K_{\text{ME}}=B\varepsilon$, with B being the magnetoelastic constant derived from bulk Ni properties.[2] Both Ni and Cu crystallize in the fcc structure with the Ni lattice constant being 2.5% smaller than the Cu lattice constant. Structural investigations[10,11] of Ni growth on Cu(001) show that the Ni films initially grow pseudomorphically. This growth mode continues up to a critical thickness d_c where the onset of strain relaxation causes the formation of misfit dislocations. Strain relaxation also reduces the magnetoelastic anisotropy energy, causing the magnetization to switch back to in-plane for thicker films. In the pseudomorphic growth region, the strain is a constant $\varepsilon=-\eta$, where η is the lattice misfit. Chappert and Bruno[9] have derived an expression for the residual strain for film thicknesses greater than d_c, $\varepsilon=-\eta\, d_c/d$. Using this and the value of K_{ME} for $d<d_c$ determined by Schulz and Baberschke,[2] we obtain $K_{\text{ME}}=3.95d_c/d\times10^6$ ergs/cm^3 for $d>d_c$.

The transition from perpendicular to in-plane magnetization at higher coverages due to strain relaxation can now be determined. In Fig. 3 we compare $K_{\text{ME}}+(K_S+K_I)/d$ to $2\pi M^2$ for different film thicknesses. When $K_{\text{ME}}+(K_S+K_I)/d$ is less than $2\pi M^2$, the gray region in Fig. 3, the magnetization will be in-plane. The pseudomorphic region is represented by the straight line with negative slope and predicts a transition from in-plane to perpendicular magnetization near 6 ML. The abrupt change in the slope of $K_{\text{ME}}+(K_S+K_I)/d$ occurs at d_c and is a result of the change in K_{ME} from being thickness independent, pseudomorphic region, to being thickness dependent, beyond the pseudomorphic region. The generally accepted value of d_c for Ni growth on Cu(001) is 8 ML.[2,3] Using this value of d_c, (2) predicts that the magnetization in the film should be in-plane again above 14 ML, Fig. 3. This is clearly not in agreement with the results in Fig. 2, which show that the perpendicular to in-plane transition occurs at ~37 ML. Either the assumption that $\varepsilon=-\eta\, d_c/d$ or that $d_c=8$ ML must be incorrect. This value of $d_c=8$ ML comes from an *ex situ* TEM study of a Ni/Cu(001) wedge.[10] The Cu(001) substrate was grown on a NaCl surface in high vacuum (10^{-7} Torr). After Ni deposition (2×10^{-8} Torr) the bilayer was floated off the NaCl surface and examined by TEM and diffraction. Due to the greatly different techniques and conditions in film preparation between our experiment and the TEM experiment we suspect that the value of $d_c=8$ ML may not be valid for our experiment. In fact, recent x-ray photoelectron diffraction experiments on 10 ML Ni/Cu(001) (Ref. 11) are well explained by assuming that no strain relaxation has taken place. This sets a lower limit of 10 ML on d_c.

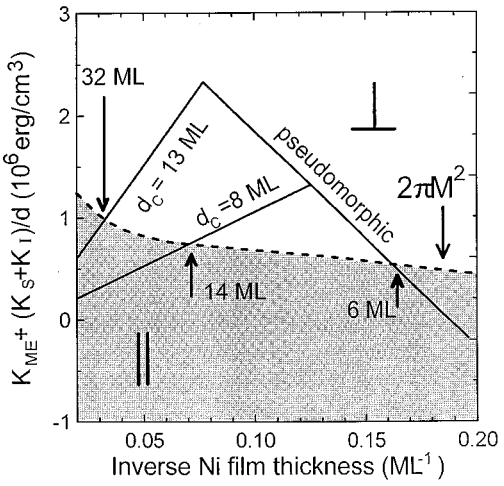

FIG. 3. Magnetoelastic anisotropy K_{ME} plus thickness-dependent anisotropy $(K_S+K_I)/d$ vs inverse Ni film thickness. Values of K_{ME} for the pseudomorphic region and K_S+K_I are from Ref. 2. The dashed line represents the demagnetizing energy for perpendicular magnetization $2\pi M^2$. If the plotted anisotropy constants are less than the demagnetizing energy, gray region, the Ni films will be magnetized in-plane. Otherwise the magnetization will be perpendicular to the surface. Values for K_{ME} beyond the pseudomorphic region are given assuming $d_c=8$ and 13 ML, see text.

The need for an accurate value of d_c is apparent. In Fig. 4(a) we show hysteresis curves obtained for different Ni thicknesses. The hysteresis curves are square up to about 30 ML. In Fig. 4(b) we plot the coercive field, determined from the hysteresis curves vs film thickness. The results in Fig. 4(b) show a gradual rise in the coercive field H_c up to about 12 ML. Near 13 ML there is a rapid rise in H_c, from about 25 Oe to about 190 Oe. It is well known that the strain

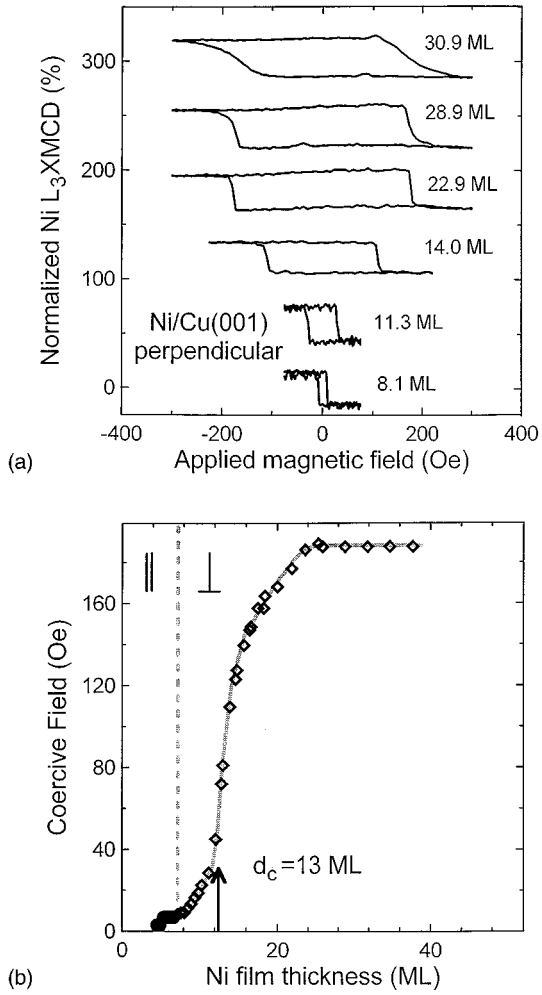

FIG. 4. (a) Hysteresis curves for different Ni film thicknesses from Ni/Cu(001) wedges. The magnetic field was applied perpendicular to the film surface. (b) Coercive field vs thickness across Ni/Cu(001) wedges. The rapid rise in the coercive field beginning near 13 ML is due to the formation of misfit dislocations and the critical thickness for epitaxial growth d_c is determined to be 13 ML.

in pseudomorphic films is relieved by the formation of misfit dislocations once the critical thickness for pseudomorphic growth has been exceeded. It is equally well known that defects and dislocations in magnetic single crystals increase the coercive field.[12] These two arguments taken together allow us to interpret the rapid rise in the coercive field vs Ni film thickness as being due to the formation of misfit dislocations. We can also take the onset of the rapid rise in H_c as an estimate for d_c. From Fig. 4 $d_c \sim 13$ ML for Ni/Cu(001). Using $d_c = 13$ ML, we determine that the crossover from perpendicular to in-plane magnetization should occur at 32 ML, Fig. 3, in good agreement with our experimental results in Fig. 2.

Both transitions in the easy axis of magnetization are now well described by Eq. (2). Our analysis relies heavily on the ability to determine d_c from changes in H_c. To show that the rapid increase in H_c vs film thickness is a general phenomena which is useful for measuring d_c we have made similar measurements on Co wedges grown on Cu(001). Hysteresis curves for Co/Cu(001) were all square except for the 13–15 ML region. The results are given in Fig. 5 where H_c is plotted vs film thickness. There is a gradual increase in H_c between 2 and 5 ML, due to the Curie temperature changes with film thickness. Beginning at 12 ML there is a rapid increase from $H_c = 20$ Oe to $H_c = 100$ Oe. Co grows pseudomorphically on Cu(001) in a metastable face-centered-tetragonal structure up to a critical thickness of ~11 ML as determined by TEM.[13] Extended x-ray absorption fine-structure (EXAFS) measurements[14] show no strain relaxation for film thicknesses up to 8 ML, supporting the TEM results. For Co/Cu(001), interpreting the onset of the rapid rise in H_c near 12 ML as a measure of d_c is consistent with the TEM and EXAFS measurements.

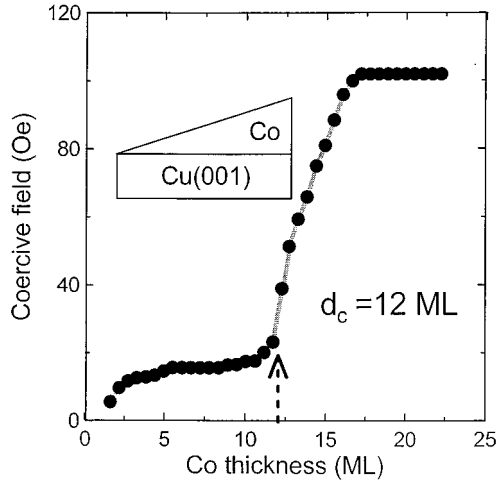

FIG. 5. Coercive field vs Co thickness across Co/Cu(001) wedges, for in-plane magnetization along the ⟨100⟩ direction. The rapid rise in the coercive field beginning at 12 ML suggests that the critical thickness for epitaxial growth d_c is 12 ML. This is consistent with TEM measurements setting $d_c = 11$ ML, Ref. 13, and EXAFS measurements which set $d_c > 8$ ML, Ref. 14.

B. Cu/Ni/Cu(001): separation of surface and interface anisotropy energies

It is of interest to determine the surface and interface anisotropy constants separately. Since the two anisotropies have the same thickness dependence, they are difficult to separate experimentally. One way to achieve the separation is to compare measurements made on thin films to measurements made on sandwich structures. Magnetism in the thin film is influenced by $K_I + K_S$, while the magnetism of the sandwich structure is influenced by $2K_I$. We have grown a 5–11 ML wedge of Ni/Cu(001) and have covered half of the wedge with 10 Å of Cu. The normalized Ni XMCD intensity at L_3 was measured at different positions along the wedge on both the Cu covered and uncovered sections. In this manner, any change in the magnetization direction due to the addition of Cu could be precisely determined.

The results of these measurements are shown in Fig. 6. The transition from in-plane magnetization to perpendicular magnetization occurs near 6 ML for the Cu/Ni/Cu(001) sandwich structure, 1 ML thinner than for the uncovered Ni wedge. This shows that both K_I and K_S are less than zero and that the magnitude of K_S is greater than the magnitude of K_I. Values of both K_I and K_S can be determined using $K_I + K_S = -0.38$ ergs/cm^2,[2] $K_{ME} = 3.95 \times 10^6$ ergs/cm^3,[2] and

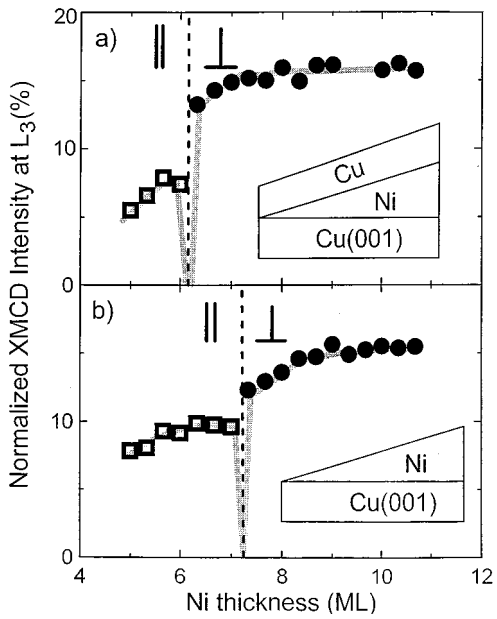

FIG. 6. Normalized Ni XMCD intensity at L_3 measured from a 5–11 ML wedge of Ni grown on Cu(001). Half of the wedge was covered with 10 Å of Cu to form a Cu/Ni-wedge/Cu(001) sandwich structure. The transition from in-plane to perpendicular magnetization occurs at 6 ML for the sandwich structure and at 7 ML for the uncovered Ni wedge. This shows that both the surface and interface anisotropy constants are negative.

values of M vs d.[3] In an analysis similar to that presented in Fig. 3 we find that $2K_I = -0.32$ ergs/cm^2 in order for the in-plane to perpendicular transition to take place at 1 ML thinner coverage for the sandwich structure. This gives the final result that $K_I = -0.16$ ergs/cm^2 and $K_S = -0.22$ ergs/cm^2. Both the interface and surface anisotropies favor in-plane magnetization with the magnitude of the surface anisotropy being larger.

This unusual interplay between a magnetoelastic anisotropy which favors a perpendicular easy axis, and an interface term favoring parallel alignment, was found in Cu/Ni/Cu(111) structures by Gradmann,[15] who determined interface and surface anisotropy constants of $K_I = -0.22$ ergs/cm^2 and $K_S = -0.48$ ergs/cm^2 for the (111) orientation.

Our experiments on Cu/Ni/Cu concern the spin-reorientation transition below the critical layer thickness. The other transition, from perpendicular to in-plane above d_c, has recently been investigated by Bochi *et al.*[16] They found that the transition from perpendicular to in-plane magnetization for Cu/Ni/Cu takes place at ~75 ML Ni. In their analysis Bochi *et al.* included the effects of a strain-dependent magnetoelastic interface anisotropy and determined that the interface anisotropy constant was positive, in direct opposition to our result [and indirectly to that of Gradmann for Ni(111) (Ref. 15)]. The model of Bochi *et al.* predicts that the transition from in-plane to perpendicular magnetization occurs at 17 ML for Cu/Ni/Cu(001), a factor of ~3 larger than the experimental value of 6 ML (see Fig. 6). Our model, based on Eq. (2), which we have used to explain the Ni/Cu(001) results, can easily be extended to the Cu/Ni/Cu(001) system. To do this we assume that the critical thickness for epitaxial growth is the purely thermodynamic result for a capped system.[9] In an analysis similar to Fig. 3 we predict a transition from perpendicular to in-plane at 66 ML for Cu/Ni/Cu(001), very close to the experimental value of 75 ML.[16] Our model based on Eq. (2) explains both transitions in the easy axis direction for the two systems Ni/Cu(001) and Cu/Ni/Cu(001), without having to invoke a strain-dependent surface anisotropy.

C. Fe/Ni/Cu(001) and Co/Ni/Cu(001): Effects of ferromagnetic capping layers

Knowledge of the different anisotropy energies responsible for determining the direction of magnetization in Ni thin films should allow us to switch the magnetization direction simply by changing the magnitude of one of the anisotropy constants in Eq. (2). For example, lowering the sample temperature will increase the demagnetizing energy, reducing the thickness range for perpendicular magnetization. Growing Ni films on other substrates, with different lateral lattice constants, would change both K_{ME} and d_c and therefore the range of perpendicular magnetization. A third way of controlling the magnetization direction would be to cap the Ni films, effectively altering the surface anisotropy.

In order to investigate the possibilities of controlling the spin direction in Ni thin films we have studied the magnetization of Ni/Cu(001), wedges, 0–18 ML, capped with a 2 ML film of either Co or Fe. For the Co capped Ni wedge we find that the magnetization lies in-plane for Ni thicknesses between 0 and 18 ML. The Co capping layer forces the magnetization of the Ni film to lie in-plane. At each position along the wedge the hysteresis curves of Ni and Co, which can be obtained separately by XMCD, were identical in shape showing that the capping layer and Ni wedge were ferromagnetically coupled. For the Fe capped wedge the magnetization direction is in-plane for Ni thicknesses below ~7 ML and perpendicular to the surface for Ni thicknesses between ~7 and 18 ML. The Fe capping layer has no measurable effect on the Ni magnetization direction. At each position along the wedge the Fe capping layer was ferromagnetically coupled to the Ni.

The coercive fields of the Co capped wedge and Fe capped wedge are plotted as a function of Ni film thickness in Figs. 7(a) and 7(b), respectively. In both cases a rapid rise in the coercive field begins near 11 ML of Ni. The critical thickness for epitaxial growth is reduced by about 2 ML after including the capping layers. This is a result of the additional strain included in the capping layer. What is remarkable about the results presented in Fig. 7 is the nondependence in the changes in the coercive field vs thickness due to domain-wall type. Thin films with in-plane magnetic orientation rotate through the motion of Neél domain walls, while thin films with magnetization perpendicular to the surface rotate through Bloch wall motion. The correlation between d_c and the onset of the rapid rise in H_c appears to be independent of domain-wall type.

In an initial attempt to describe the effects of capping layers on the magnetization direction we consider two changes to Eq. (2). The first change we consider is that the surface anisotropy is now determined by the capping layer, either Co or Fe. To estimate the new surface anisotropy we use the results of experiments on thin films of Co/Cu(001) (Ref. 17) and Fe/Ag(001),[18] where the surface anisotropy of

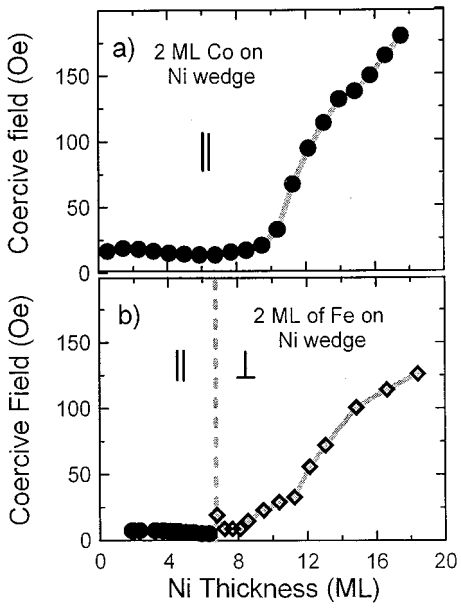

FIG. 7. Coercive fields of (a) 2 ML Co/Ni/Cu(001) and (b) 2 ML Fe/Ni/Cu(001) vs Ni film thickness. The Co capping layer forces the Ni magnetization to lie in-plane, consistent with the prediction of Fig. 8. The Fe capping layer has no affect on the magnetization direction in the Ni films, in disagreement with the prediction given in Fig. 8. For both capped Ni wedges there is a rapid rise in the coercive field which begins near 11 ML.

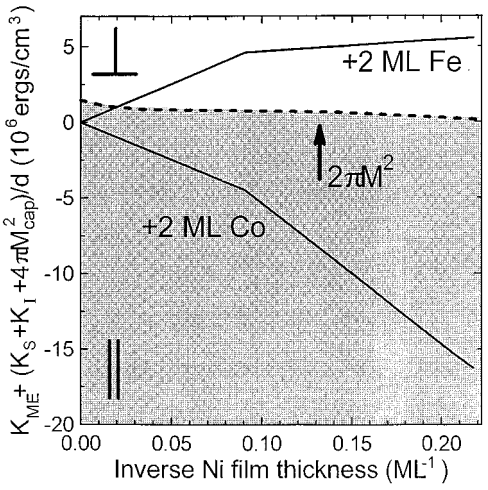

FIG. 8. Predicted effects of ferromagnetic capping layers on the easy axis of magnetization for Ni/Cu(001). The plotted anisotropy energies are similar to those in Fig. 3 except that the surface anisotropy is now that of the capping layer, and the demagnetizing energy of the capping layer has been added. Addition of 2 ML Co is predicted to force the Ni magnetization in-plane for all Ni thicknesses. Addition of 2 ML Fe is predicted to force the Ni magnetization to be perpendicular for all Ni thicknesses less than 50 ML.

Co and Fe have been measured. The second change to Eq. (2) which we consider is in the demagnetizing energy. The demagnetizing energy due to the capping layer, per unit volume of the Ni film will depend on $M_{\rm cap}$ and will vary with thickness. The demagnetizing energy of the underlying Ni film is unchanged. Under these assumptions the condition for magnetization perpendicular to the surface becomes

$$K_{\rm ME}+\frac{K_S+K_I-4\pi M^2_{\rm cap}}{d}>2\pi M^2 \qquad (3)$$

for a 2 ML capping layer, where K_S is now the surface anisotropy of the capping layer. In the derivation of Eq. (3) we ignore the anisotropy energies due to the Ni/capping layer interface. The results of Eq. (3) for a 2 ML capping layer of Fe and Co are shown in Fig. 8. Upon addition of 2 ML of Co the Ni magnetization is predicted to lie in plane for all Ni thickness. For 2 ML of Fe the Ni magnetization is predicted to have perpendicular magnetization below ~50 ML and in-plane magnetization above ~50 ML. We have assumed that $K_S=-1.06$ ergs/cm^2 (Ref. 17) for the Co capping layer and 0.96 ergs/cm^2 (Ref. 18) for the Fe capping layer and that $d_c=11$ ML. Room-temperature bulk values were used for $M_{\rm cap}$.

The predictions for a 2 ML capping film of Co are in agreement with experiment. Both the increase in the magnitude of K_S and the increased demagnetizing energy due to the addition of the Co capping layer force the Ni magnetization to lie in-plane. For a Fe capping layer the agreement with experiment is poor. Our analysis shows that the change in surface anisotropy should increase the range of perpendicular magnetization while the increase in demagnetizing energy should decrease the range of perpendicular magnetization. Further analysis shows that including an additional thickness-dependent anisotropy energy term of -0.57 ergs/cm^2 to Eq. (3) is sufficient to achieve agreement with theory. This additional anisotropy term may be due to intermixing, a non-negligible Fe/Ni interface anisotropy, an error in using $K_S=0.96$, or in problems associated with calculating the demagnetizing field for ultrathin films.[19]

IV. SUMMARY AND CONCLUSIONS

Using Ni wedges grown on Cu(001) we have studied the magnetic properties of ultrathin Ni/Cu(001) films. For film thicknesses between 4 and 7 ML the magnetization lies in the surface plane. At 7 ML there is a sharp transition to magnetization perpendicular to the surface. Above 37 ML there is a gradual transition back to in-plane magnetization. Both these transitions in the direction of magnetization can be explained by considering four anisotropy constants: the surface, interface, magnetoelastic, and a shape anisotropy. From similar measurements on a Cu/Ni-wedge/Cu(001) sandwich structure we find that both the surface and interface anisotropy constants are negative, with the magnitude of the surface anisotropy constant being greater.

Below 7 ML the surface and interface anisotropies dominate, causing in-plane magnetization. Above 7 ML the magnetoelastic anisotropy overcomes the thickness-dependent surface and interface anisotropies, leading to magnetization perpendicular to the surface. As the critical thickness for epitaxial growth is exceeded, the strain in the Ni films due to pseudomorphic growth become too large. The film begins to relax to the stable fcc phase as misfit dislocations form. This causes a reduction in the magnetoelastic anisotropy energy causing the magnetization to switch back to in-plane at 37 ML. According to Chappert and Bruno[9] the magnetoelastic anisotropy becomes thickness independent above d_c. The

critical thickness for epitaxial growth can be observed by a rapid rise in the coercive field at 13 ML which is due to the formation of misfit dislocations. Using $d_c = 13$ ML and the theory of Chappert and Bruno we calculate a transition back to in-plane magnetization at 32 ML, within 15% of our experimental value of 37 ML. This analysis gives a complete description of both transitions in the magnetization direction for Ni/Cu(001).

With knowledge of which anisotropies are important we have attempted to change, in a controlled manner, the magnetization direction through the addition of capping layers. In a simple model we assume that the capping layer has two effects on the direction of magnetization. The first is that the surface anisotropy is changed and the second is the addition of a demagnetizing term for the capping layer. The model predicts that a Co capping layer will force the Ni magnetization in-plane for all Ni thicknesses. This prediction is shown to be correct by experiment on a 0–18 ML Ni wedge capped with 2 ML of Co. For a Fe capping layer the model predicts magnetization perpendicular to the surface for Ni films less than 50 ML. Experiments on a Ni wedge capped with 2 ML of Fe show that the Fe capping layer has no effect on the Ni magnetization direction. This discrepancy between model and experiment suggests that either interface anisotropies or intermixing are important for the Fe-capped Ni wedge. Further experiments are presently being planned to study the anisotropy energies important for determining the magnetization direction in ferromagnetically capped ultrathin films and ferromagnetically coupled systems in general.

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