

## Evidence for strong surface magnetoelastic anisotropy in epitaxial Cu/Ni/Cu(001) sandwiches

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Measurements of effective magnetic anisotropy energy of epitaxial Cu/Ni/Cu(001) sandwiches are analyzed as a function of Ni film thickness  $h$ . The magnetization easy axis is perpendicular to the films for  $20 < h < 140$  Å. The magnetic anisotropy is best described by inclusion in the effective anisotropy energy of the strain-dependent magnetic surface anisotropy term, predicted by the strain-dependent Néel model, along with the usual magnetostatic, magnetocrystalline, and bulk magnetoelastic energies. The *surface magnetocrystalline* and *surface magnetoelastic* anisotropy energies of the Ni/Cu(001) interface are determined to be  $+0.9$  erg/cm<sup>2</sup> and  $-52$  erg/cm<sup>2</sup>, respectively. The effective magnetoelastic coupling coefficient (bulk plus surface) of Cu/Ni/Cu(001) is predicted to change sign at  $h = 80$  Å. The two observed magnetization easy-axis reversals are also well described by this model.

The total anisotropy energy density of a uniformly magnetized material can be represented as

$$K^{\text{eff}} \sin^2 \theta, \quad (1)$$

where  $\theta$  is the angle between the magnetization and the film normal and  $K^{\text{eff}}$  includes all relevant anisotropy energy density contributions:  $K^{\text{eff}} = -2\pi M_s^2 + K_{\text{MC}}^b + B^b e$ , magnetostatic, magnetocrystalline, and magnetoelastic terms, respectively. Here  $M_s$  is the saturation magnetization,  $K^b$  is the bulk magnetocrystalline anisotropy,  $B^b$  is the bulk magnetoelastic coupling coefficient, and  $e$  is the strain tensor. (The magnetoelastic coupling coefficient  $B^b$  is proportional to the negative of the more familiar magnetostriction coefficient  $\lambda_s$ .) The reduced symmetry in the atomic coordination at the interfaces of magnetic thin films generally introduces a significant contribution to the effective magnetic anisotropy:

$$K^{\text{eff}} = -2\pi M_s^2 + K_{\text{MC}}^b + B^b e + (K^s + B^s e)/h. \quad (2)$$

The existence of the magnetic surface anisotropy is often justified in terms of a phenomenological model first proposed by Néel.<sup>1</sup> It is not widely recognized that the strain-dependent part of the magnetic surface anisotropy,  $B^s e/h$ , i.e., *surface magnetoelastic* (ME) anisotropy,<sup>2</sup> comes as naturally from the pair-interaction model<sup>3</sup> as does the strain-independent *surface magnetocrystalline* anisotropy,  $K^s/h$ .

Few studies of magnetic anisotropy in thin films consider strain effects due to lattice misfit and its partial accommodation by misfit dislocations,<sup>4-10</sup> surface relaxation, and/or surface terraces.<sup>3,11</sup> However, in Refs. 4-11 only *bulk* ME interactions have been taken into account in the analysis of the behavior of the magnetic anisotropy.

In the past few years, several experimental results have unambiguously demonstrated that ME interactions at surfaces and in thin films are significantly different than in the bulk. Zuberek *et al.*<sup>12</sup> found that the effective magnetostriction constant of Ni/Ag multilayers goes from negative to positive values as the Ni film thickness goes to zero. Sun and O'Handley<sup>13</sup> found that the surface ME coupling coefficient in Co-rich and Fe-rich amorphous alloys can differ sharply from the bulk value. More recently, Song *et al.*<sup>14</sup> measured

the effective ME coupling coefficient in polycrystalline NiFe/Ag/Si, NiFe/Cu/Si, and Ni/SiO<sub>2</sub>/Si thin films by a direct *in situ* method.<sup>2</sup>  $B^{\text{eff}}$  was shown to diverge from the bulk value to more positive values for film thicknesses below 150 Å and to take giant positive values in films thinner than 40-60 Å due to a significant surface contribution. In particular, they found that  $B^s \approx +20$  ergs/cm<sup>2</sup> in polycrystalline Ni thin films. The magnetostriction constant has also been measured *in situ* for polycrystalline Fe thin films using a cantilever beam technique.<sup>15</sup> Significant deviations from the bulk value of the magnetostriction constant were observed for Fe film thicknesses below approximately 100 Å. The effective magnetostriction constant was shown to change sign when the Fe film thickness was between 80 and 30 Å. It has recently been shown that, for vacuum/Ni/Cu/Si(001),  $B^{\text{eff}} = B^b + B^s/h$  changes sign at a Ni thickness of 28 Å.<sup>16</sup> Bochi, Song, and O'Handley<sup>17</sup> have shown that published data on the magnetic anisotropy in epitaxial fcc (111) Co/Cu superlattices<sup>6</sup> can be more effectively interpreted by inclusion of a surface ME term. It was found that  $B^s$  (Co/Cu)(111)  $\approx -24$  ergs/cm<sup>2</sup>, implying that the effective ME coupling coefficient of fcc Co/Cu(111) multilayers changes sign around a Co thickness of 9 Å.

In the present work, we report the behavior of both the magnetic anisotropy and the ME coupling in epitaxial Cu/Ni/Cu/Si(001) sandwiches. This system is remarkable for the broad Ni thickness range over which the easy axis of magnetization is perpendicular to the film plane. This strong perpendicular magnetization has recently been vividly confirmed by extensive magnetic force microscopy (MFM) studies.<sup>18</sup> We show that the interpretation of the dependence of the magnetic anisotropy energy on Ni film thickness is severely wanting without the inclusion of the surface ME anisotropy,  $B^s e/h$ , of Eq. (2). Using a phenomenological model developed recently<sup>17</sup> and based on the strain-dependent pair-interaction model of magnetic surface anisotropy,<sup>3</sup> we are able to determine the surface magnetocrystalline anisotropy  $K^s$ , and surface ME coupling coefficient  $B^s$ , corresponding to the Ni/Cu(001) interface. The variation of the effective ME coupling coefficient  $B^{\text{eff}} = B^b + B^s/h$  with Ni film thickness is also determined. The in-

plane magnetization at the smallest Ni thickness is found to be due to the negative surface ME anisotropy energy,  $2B^s e(h)/h$ . The origin of the strong perpendicular magnetic anisotropy in this system, evident over the Ni thickness range  $25 < h < 140$  Å, is found to arise from the surface magnetocrystalline anisotropy energy  $K^s$  and the bulk ME anisotropy energy  $2B^s e(h)$ . The return to in-plane magnetization above 140 Å is a magnetostatic effect.

Our (20-Å Cu)/Ni/(2000-Å Cu) sandwiches were deposited at room temperature on Si(001) substrates by MBE. Base pressure was in the  $10^{-10}$ -Torr range and rose to the  $10^{-9}$ -Torr range during the depositions. The Cu substrate layer was deposited at 3.0 Å/s and the Ni film and Cu capping layer were deposited at 0.5 Å/s. Thicknesses were determined by a quartz crystal oscillator calibrated by repeated profilometer measurements on thicker films. Thickness error is less than  $\pm 6\%$ . Further details of the experimental procedures are given elsewhere.<sup>8,19,20</sup> The crystallographic quality of the sandwiches was studied *in situ* by reflection high-energy electron diffraction (RHEED) and *ex situ* by x-ray diffraction and transmission electron microscopy (TEM). RHEED confirmed Ni epitaxy on the Cu(001) surface by pattern continuity. X-ray diffraction pole figures showed the fourfold symmetry expected of epitaxial Ni/Cu rotated 45° from the fourfold pattern of Si(001). High-resolution cross-sectional TEM confirmed Ni and Cu epitaxy on Si(001) and suggested roughness of order  $\pm 20$  Å about the mean. This roughness was reflected in the spot-plus-streak pattern observed by RHEED. Atomic force microscopy has also confirmed the 20-Å surface roughness. Using plan-view TEM, we observed both 60° and 90° misfit dislocations (MD's) running along the  $\langle 110 \rangle$  directions of the Ni/Cu(001) interface and we measured their densities as a function of Ni film thickness.<sup>19,20</sup> MD's were present in Ni/Cu(001) thin films with Ni thicknesses of 25 Å and greater but not in the 15-Å-thick films, which indicates that the critical thickness  $h_c$  for the onset of MD's is between 15 and 25 Å. A TEM image of the MD's appears in Ref. 19. For Cu/Ni/Cu(001) sandwiches, we observed dislocations in Ni films as thin as 30 Å, indicating that  $h_c < 30$  Å.<sup>21</sup> We also measured the average in-plane biaxial tensile misfit strain  $e_0(h)$  in the Ni films as a function of the film thickness in Ni/Cu/Si(001) epitaxial thin films by measuring the change in substrate curvature using an optical interferometry technique.<sup>20</sup> The magnetic properties of our Cu/Ni/Cu(001) sandwiches were studied *ex situ* using a vibrating sample magnetometer where magnetic fields up to 10 kOe were available to saturate the films in plane or out of plane and hence to measure the effective magnetic anisotropy energy density,  $K^{\text{eff}}$ , as a function of Ni film thickness  $h$ .

The saturation magnetization of our Cu/Ni/Cu/Si(001) films was also determined and found to be within  $\pm 10\%$  of the bulk value for Ni except for films of thickness 20 Å or less, which were lower. The present paper describes results on films of 35-Å Ni thickness and greater. Further, a careful study by Huang *et al.*<sup>22</sup> has shown that the Curie temperature of bulk Ni is valid for Ni films as thin as 35 Å.

Our measurements of  $K^{\text{eff}}h$  versus  $h$  are shown in Fig. 1. The first important result is that perpendicular magnetic anisotropy ( $K^{\text{eff}} > 0$ ) dominates up to Ni thicknesses of approximately 135 Å in Cu/Ni/Cu(001) sandwiches. This is in agree-

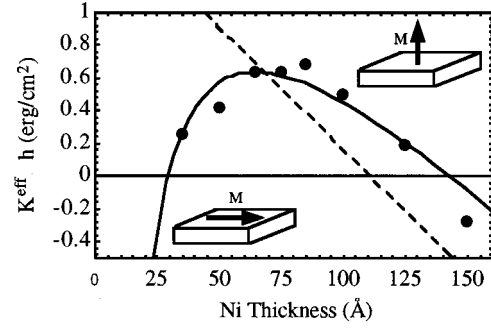


FIG. 1.  $K^{\text{eff}}h$  versus Ni film thickness for our Cu/Ni/Cu(001) sandwiches. The solid curve is a plot of  $K^{\text{eff}}(h)$  versus  $h$  using Eq. (4) and the magnetic surface energies in the first row of Table I. The dashed straight line is the best fit to the data with  $B^s = 0$ .

ment with the measurements of Naik *et al.*<sup>23</sup> on 500-Å Cu/Ni/500-Å Cu/Si(001) and the measurements of Jungblut *et al.*<sup>9</sup> on 25-Å Au/10-Å Cu/Ni wedge/Cu(001), both of which showed that the magnetization easy axis is perpendicular to the sandwiches up to a Ni thickness of approximately 100 Å. The region of perpendicular magnetization in Cu/Ni/Cu(001) shown in Fig. 1 is exceptionally broad, even when compared to the remarkable width of the perpendicular region discovered in Ni/Cu(001) thin films.<sup>8,16</sup> MFM images on our films as thin as 20 Å confirm the strong perpendicular magnetization.<sup>18</sup> The second important result of Fig. 1 is that the magnetization easy axis shows *two* switching thicknesses: one near 135 Å, which our measurements confirm, and another one near 20 Å. Two switching thicknesses for the magnetization easy axis have also been reported in Ni/Cu(001) thin films,<sup>8,16</sup> Fe/Ag(001),<sup>24</sup> and Fe/Cu(001).<sup>25</sup> We now interpret the Ni thickness dependence of  $K^{\text{eff}}$ , the large range of perpendicular magnetization observed, and the two average spin reorientation transitions.

The effective magnetic anisotropy energy of an ultrathin strained epitaxial (001) film sandwiched between *two* identical nonmagnetic layers can be described from Eq. (2) by the following general phenomenological equation:<sup>17</sup>

$$K^{\text{eff}} = -2\pi M_s^2 + 2\left(B_1 + \frac{B^s}{h}\right) e_0(h) + \left(K_1 + \frac{2K^s}{h}\right). \quad (3)$$

$B_1$  is the first-order cubic bulk ME coupling coefficient and  $e_0(h)$  is the average in-plane biaxial misfit strain. The bulk magnetocrystalline anisotropy,  $K_1$ , is negligible for Ni/Cu(001) thin films. As explained above, all of the Ni films in this study have a thickness  $h > 30$  Å  $\gg h_c$ . For  $h > h_c$ ,  $e_0(h)$  decreases with increasing film thickness as misfit strain is accommodated by interfacial MD's. Using the form of the average strain suggested by Chappert and Bruno,<sup>5</sup>  $e_0(h) = \eta h_c/h$  (where  $\eta$  is the film-substrate lattice mismatch), in Eq. (3) we obtain

$$K^{\text{eff}}h = -2\pi M_s^2 h + 2(B_1 \eta h_c + K^s) + \frac{2B^s \eta h_c}{h}. \quad (4)$$

By fitting our data of Fig. 1 with Eq. (4) using  $B_1 = 6.2 \times 10^7$  ergs/cm<sup>3</sup>,  $\eta = 2.6\%$ ,  $h_c = 18$  Å (the thermodynamic critical thickness), and  $2\pi M_s^2 = 1.5 \times 10^6$  ergs/cm<sup>3</sup>, we obtain the solid line shown in Fig. 1. Compar-

TABLE I. Surface ME coupling coefficient and surface magnetocrystalline anisotropy energy determined for the Ni/Cu(001) interface. The first three rows summarize the results obtained by applying the phenomenological model of Eq. (4) to our data and to those of Jungblut *et al.* (Ref. 9) on Cu/Ni/Cu(001) sandwiches. For our data, we have used both a  $1/h$  and a  $1/h^{0.7}$  Ni film thickness dependence of the strain. The results in the last row are the ones reported by Jungblut *et al.* using the model of Eq. (4) with  $B^s=0$ .

Cu/Ni/Cu(001) sandwiches	$B^s$ (Ni/Cu)(001) (ergs/cm <sup>2</sup> )	$K^s$ (Ni/Cu)(001) (ergs/cm <sup>2</sup> )
Our data $e_0(h) = (\eta h_c/h)$	-67	+0.98
Our data $e_0(h) = (0.18/h^{0.7})$	-52	+0.88
Data of Jungblut <i>et al.</i> (1994) $e_0(h) = (\eta h'_c/h)$	-37	+0.73
Results of Jungblut <i>et al.</i> (Ref. 9)		-0.40

ing the equation of this line with Eq. (4), we get the magnetic surface anisotropy coefficients for the Ni/Cu(001) interface:

$$K^s = +0.98 \text{ ergs/cm}^2 \quad \text{and} \quad B^s = -67 \text{ ergs/cm}^2.$$

We have also fit our experimental data using the thickness dependence of the strain *measured* for Ni/Cu(001) thin films by optical interferometry.<sup>16</sup> In that case we obtained  $K^s(\text{Ni/Cu})(001) = +0.88 \text{ ergs/cm}^2$  and  $B^s(\text{Ni/Cu})(001) = -52 \text{ ergs/cm}^2$ . As evidenced by these numbers, the absolute value of the surface anisotropy energies extracted from the fit are very sensitive to the thickness dependence of the misfit strain. We have also analyzed the data of Jungblut *et al.*<sup>9</sup> using Eq. (4) (with  $h_c = 15 \text{ \AA}$ , as reported in Ref. 9) and found that their data can be very well described by Eq. (4). In this case, the magnetic surface anisotropy energies are determined to be  $K^s(\text{Ni/Cu})(001) = +0.73 \text{ ergs/cm}^2$  and  $B^s(\text{Ni/Cu})(001) = -37 \text{ ergs/cm}^2$ . When they analyzed their own data, Jungblut *et al.* found  $K^s(\text{Ni/Cu})(001) = -0.40 \text{ ergs/cm}^2$ , opposite in sign to our results. They assumed *a priori* that  $B^s = 0$  and  $h_c = 40 \text{ \AA}$  for their Cu/Ni/Cu(001) sandwiches.

Traditionally, the surface ME anisotropy, although arising naturally from the Néel model, has been completely omitted. This is equivalent to setting  $B^s = 0$  in Eq. (4). When  $B^s = 0$ , a plot of  $K^{\text{eff}}h$  versus  $h$  gives a straight line of negative slope equal to  $-2\pi M_s^2$ . We tried to fit our data of Fig. 1 with such a line. The result, displayed in Fig. 1 with the dashed line, clearly shows that the fit is very poor. Such a model ( $B^s = 0$ ) is therefore inadequate for Cu/Ni/Cu(001) sandwiches.

The above results on the surface magnetocrystalline anisotropy energy and the surface ME coupling coefficient of the Ni/Cu(001) interface are summarized in Table I. The surface magnetocrystalline anisotropy energies are all large and positive indicating that  $K^s(\text{Ni/Cu})(001)$  together with the *bulk* ME anisotropy energy,  $2B_1e_0(h)$ , constitute the origin of the remarkable perpendicular magnetic anisotropy in Cu/Ni/Cu(001) sandwiches. On the other hand, the surface

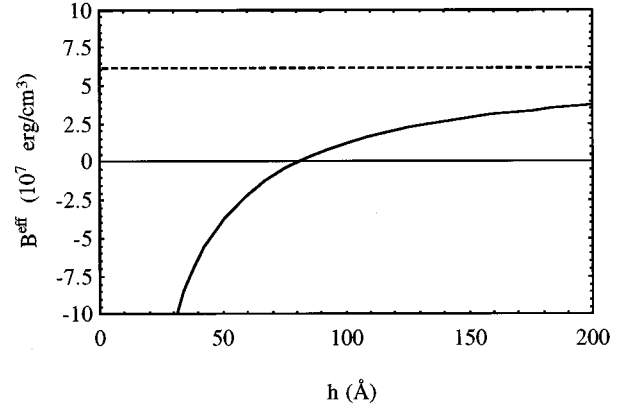


FIG. 2. Dependence of the effective ME coupling coefficient on Ni film thickness in Cu/Ni/Cu(001) sandwiches. We used the average value  $B^s(\text{Ni/Cu})(001) \approx -50 \text{ ergs/cm}^2$  obtained from the data in Table I. The dashed line indicates the bulk ME coupling coefficient of Ni.

ME coupling coefficients in Table I are negative, indicating that the *surface* ME anisotropy energy  $2B^se_0(h)/h$  favors an in-plane magnetization in Ni/Cu(001). At small Ni thicknesses ( $h < 20 \text{ \AA}$ ), this negative term becomes very large and is responsible for keeping the magnetization in-plane there. The bulk magnetostatic energy  $2\pi M_s^2$ , on the other hand, is responsible for bringing the average magnetization back in plane for  $h > 135 \text{ \AA}$ . Our model therefore gives a good explanation as to why the magnetization easy axis changes orientation twice. Double magnetization easy-axis transitions are also present in Fe/Ag(001) (Ref. 24) and Fe/Cu(001).<sup>25</sup> In both of those cases, both transitions occur for  $h < h_c$ , which means that the lower transition is certainly not due to the onset of MD's, contrary to what some groups had speculated. These cases cannot be easily explained by Eq. (4) with  $B^s = 0$ .

The large values of anisotropy we observe for the Cu/Ni interface may not be typical of ideal, planar interfaces. Our Cu/Ni interfaces showed a roughness of  $\pm 20 \text{ \AA}$  about the mean. We saw no effects of Cu/Ni interdiffusion unless the films were heated to well over  $400 \text{ }^\circ\text{C}$ . If there were significant Cu/Ni interdiffusion it would relieve misfit strain at the Cu/Ni interface and cause MD's to be observed only at thicknesses much greater than the calculated critical thickness,  $h_c = 18 \text{ \AA}$ . As mentioned above, we have observed MD's in Ni/Cu(001) at  $25 \text{ \AA}$  of Ni (Ref. 19) and at  $30 \text{ \AA}$  of Ni in Cu/Ni/Cu(001).<sup>21</sup> Although thin films are not necessarily in thermodynamic equilibrium, the lack of any indication of significant NiCu mixing is consistent with the equilibrium phase diagram, which shows a miscibility gap in this system below  $354 \text{ }^\circ\text{C}$ .

The average values of the surface energies in the first three rows of Table I are  $B^s \approx -50 \text{ ergs/cm}^2$  and  $K^s \approx +0.85 \text{ ergs/cm}^2$ . In agreement with the predictions of the strain-dependent Néel model<sup>3</sup> for fcc (001) interfaces,  $K^s$  and  $B^s$  have opposite signs. Using the average surface ME coupling coefficient corresponding to the Ni/Cu(001) interface, we plot the effective ME coupling coefficient  $B^{\text{eff}} = B_1 + B^s/h$  for Cu/Ni/Cu(001) sandwiches in Fig. 2. The figure indicates that significant deviations from the bulk value  $B_1$  occur for films as thick as  $200 \text{ \AA}$  and that  $B^{\text{eff}}$

changes sign around 80 Å because  $B^s(\text{Ni/Cu})(001) < 0$ . This striking result questions the assumption, often encountered in the literature, that bulk ME coupling coefficients apply to ultrathin films and it is supported by the recent measurements of Song *et al.*<sup>14</sup> and Weber *et al.*<sup>15</sup> on polycrystalline materials. The present results are evidence for surface magnetoelastic effects in epitaxial, single-crystal films. We emphasize that  $B^s$ , just like  $K^s$ , is characteristic of the film-substrate interface and not just of the magnetic film's chemistry. Therefore  $B^{\text{eff}}$  in ultrathin films depends strongly on the symmetry and chemistry of the interfaces, not just on the film thickness  $h$ .

In summary, we have shown that epitaxial Cu/Ni/Cu(001) sandwiches exhibit the largest thickness range of perpendicular magnetization reported for a single epitaxial magnetic film. The perpendicular magnetic anisotropy of this system finds its strength in the surface magnetocrystalline

anisotropy (Néel) term and in the bulk ME anisotropy energy. The behavior of the effective magnetic anisotropy data can only be explained if we include the surface ME anisotropy energy predicted by the strain-dependent Néel model. Our results predict that the effective ME coupling coefficient,  $B^{\text{eff}} = B^b + B^s/h$ , changes sign at  $h = 80$  Å and that the magnetization easy axis exhibits two in-plane to out-of-plane transitions. These magnetization easy-axis transitions at small and large Ni thicknesses are due to the change in sign of  $B^{\text{eff}}$  and to the bulk magnetostatic energy, respectively. The onset of MD's is not found to play any special role in the lower spin reorientation.<sup>16</sup>

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