

Thickness and temperature dependence of magnetic anisotropies in ultrathin fcc Co(001) structures

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Metastable fcc Co(001) structures were grown on fcc Cu(001) using molecular beam epitaxy. Magnetic properties of these structures were studied using the ferromagnetic resonance technique. The uniaxial perpendicular and fourfold in-plane anisotropies are presented for different thicknesses. Both anisotropies are shown to be well described by a sum of contributions from the constant volume term and the $1/d$ -dependent surface term. The effect of tetragonal distortion on magnetic anisotropies is presented. The temperature dependence of anisotropies is discussed.

I. INTRODUCTION

Magnetic properties of ultrathin layers of fcc Co(001) structures grown on a Cu(001) substrate had been extensively investigated by our group previously.¹ Due to the sudden deterioration of the Cu substrate, we had been able to grow only a limited number of single-layered cobalt samples, which did not allow us to determine quantitatively the thickness and temperature dependence of magnetic anisotropies. In order to supplement our previous studies, additional single-layered Co structures were epitaxially grown after a successful revival of the Cu(001) substrate. A good Cu(001) surface with large extended terraces up to 300 nm was recovered when the following procedure was performed: after sputtering the surface of the substrate at room temperature ($V_{\text{ion}}=600$ eV, $I_{\text{ion}}=4$ $\mu\text{A}/\text{cm}^2$) to remove contaminants, the temperature was increased to 900 K while sputtering was continued for 10 h with voltage and current reduced to 500 eV and 0.1 $\mu\text{A}/\text{cm}^2$, respectively.² A detailed description of epitaxial growths and their monitoring had been presented earlier.¹ The magnetic properties of all our samples were studied by the ferromagnetic resonance (FMR) technique at both room and liquid-nitrogen temperatures.¹ The present paper is a direct extension of our earlier paper;¹ it gives a more detailed analysis of measured samples and compares our results with other recently studied fcc Co(001) ultrathin structures.

Thermal-energy atomic scattering (TEAS) and low-energy electron diffraction (LEED) studies^{3,4} showed that for the first 10 monolayers (ML) the fcc Co structures grown on Cu(001) substrates have a $p(1\times 1)$ symmetry with cobalt expanded in the plane of the surface. Detailed LEED investigations in combination with dynamical scattering calculations revealed changing tetragonal distortion for films 3 ML and thinner.⁴ The vertical compression of 5 ML films and thicker is more or less constant and amounts to $\sim 3\%$, corresponding to a ratio $c/a \approx 0.96$. Hence, a homogeneous and constant strain due to lattice mismatch is expected for samples between 5–10 ML. The tetragonal compression along the surface normal has a profound effect on the magnetic properties of Co(001) structures.

Such a distorted fcc Co lattice can be viewed as either

a face-centered-tetragonal or a body-centered-tetragonal, depending on the choice of the in-plane unit-cell vectors. In both cases, for the fourfold symmetry of the lattice about the surface normal, the anisotropy energy can be expressed as follows:

$$E_k = -\frac{1}{2}K_{1\parallel}^{\text{eff}}(\alpha_x^4 + \alpha_y^4) - \frac{1}{2}K_{1\perp}^{\text{eff}}\alpha_z^4 - K_u^{\text{eff}}\alpha_z^2, \quad (1)$$

where either $\alpha_x, \alpha_y, \alpha_z$ are direction cosines with respect to $\langle 100 \rangle$ axes for the face-centered-tetragonal (fct), or α_x, α_y are direction cosines with respect to $\langle 110 \rangle$ axes and α_z with respect to the $[001]$ axis for the body-centered-tetragonal (bct). The importance of distinction between the analysis for the fct and for the bct lattices will be shown later. $K_{1\parallel}^{\text{eff}}, K_{1\perp}^{\text{eff}}, K_u^{\text{eff}}$ are effective fourfold in-plane, fourfold perpendicular, and perpendicular uniaxial anisotropy constants. For a saturation magnetization, M_s , in the plane of the sample (parallel configuration), the fourth-order uniaxial anisotropy is negligible in FMR. The effective uniaxial and fourfold in-plane anisotropies include both bulk and surface contributions. For microwave frequencies used in FMR the magnetic moments in ultrathin films are parallel and locked in together by a strong exchange interaction. Therefore, any magnetic torques acting on the surface atomic moments are equally shared by all the atomic moments in the film. Effectively, surface anisotropies divided by the film thickness appear as bulklike anisotropies:

$$K_{1\parallel}^{\text{eff}} = K_{1\parallel}^{\text{bulk}} + \frac{1}{d}K_{1\parallel}^s, \quad (2a)$$

$$K_u^{\text{eff}} = K_u^{\text{bulk}} + \frac{1}{d}K_u^s, \quad (2b)$$

where $K_{1\parallel}^{\text{bulk}}, K_u^{\text{bulk}}, K_{1\parallel}^s, K_u^s$ describe bulk and surface fourfold in-plane and perpendicular uniaxial anisotropies. All relevant interfaces must be considered. The effective anisotropy fields, H^{eff} , are given by

$$H^{\text{eff}} = -\frac{\partial E_k}{\partial \mathbf{M}}. \quad (3)$$

II. EXPERIMENTAL RESULTS AND ANALYSIS

FMR measurements were performed at 36 and 72 GHz at both room and liquid-nitrogen temperatures. As men-

tioned earlier, the distorted fcc Co(001) lattice can be viewed as either a face-centered-tetragonal or a body-centered-tetragonal. Our analysis in Heinrich *et al.*¹ was performed with respect to the body-centered-tetragonal, in which the in-plane unit-cell vectors are along $\langle 110 \rangle$ axes. Hence directional cosines are with respect to $\langle 110 \rangle$ axes, not with respect to $\langle 100 \rangle$ cubic axes as stated in that paper. In the coordinate system along $\langle 110 \rangle$ axes the fourfold in-plane anisotropy effective fields, $2K_{1\parallel}^{\text{eff}}/M_s$, should be positive, not negative as quoted in Ref. 1. For the present paper, the anisotropy energy was taken with respect to $\langle 100 \rangle$ cubic axes, in which the lattice can be viewed as a face-centered-tetragonal. It is interesting to find a relationship between magnetic anisotropies in fct and bct lattices. For the fct film the in-plane four-fold magnetic anisotropy energy is (z direction is along the normal to the surface)

$$E_{k,\text{fct}} = -\frac{1}{2}K_{1\parallel\text{fct}}^{\text{eff}}(\alpha_{x,\text{fct}}^4 + \alpha_{y,\text{fct}}^4), \quad (4)$$

where $\alpha_{x,\text{fct}}, \alpha_{y,\text{fct}}, \alpha_{z,\text{fct}}$ are direction cosines with respect to $\langle 100 \rangle$ axes for the face-centered-tetragonal lattice. For the bct film the in-plane fourfold magnetic anisotropy is

$$E_{k,\text{bct}} = -\frac{1}{2}K_{1\parallel\text{bct}}^{\text{eff}}(\alpha_{x,\text{bct}}^4 + \alpha_{y,\text{bct}}^4). \quad (5)$$

Here $\alpha_{x,\text{bct}}, \alpha_{y,\text{bct}}$ are direction cosines with respect to in-plane $\langle 110 \rangle$ axes and $\alpha_{z,\text{bct}}$ is a directional cosine with respect to the [001] axis for the body-centered-tetragonal lattice. For the fct lattice, the directional cosines are

$$\begin{aligned} \alpha_{x,\text{fct}} &= \sin\theta \cos\phi, \\ \alpha_{y,\text{fct}} &= \sin\theta \sin\phi, \\ \alpha_{z,\text{fct}} &= \cos\theta, \end{aligned} \quad (6)$$

where θ is the angle with respect to the [001] axis and ϕ is the angle with respect to the [100] axis. The bct lattice is rotated by $\pi/4$ with respect to the fct lattice in ϕ :

$$\begin{aligned} \alpha_{x,\text{bct}} &= \sin\theta \cos\left[\phi + \frac{\pi}{4}\right], \\ \alpha_{y,\text{bct}} &= \sin\theta \sin\left[\phi + \frac{\pi}{4}\right], \\ \alpha_{z,\text{bct}} &= \cos\theta. \end{aligned} \quad (7)$$

Upon substituting the above equations into Eq. (5), and simplifying, we obtain

$$\begin{aligned} E_{k,\text{bct}} &= -\frac{3}{4}K_{1\parallel\text{bct}}^{\text{eff}} + \frac{3}{2}K_{1\parallel\text{bct}}^{\text{eff}}\alpha_{z,\text{fct}}^2 - \frac{3}{2}K_{1\parallel\text{bct}}^{\text{eff}}\alpha_{z,\text{fct}}^4 \\ &\quad + \frac{1}{2}K_{1\parallel\text{bct}}^{\text{eff}}(\alpha_{x,\text{fct}}^4 + \alpha_{y,\text{fct}}^4). \end{aligned} \quad (8)$$

By comparing the above equation with Eqs. (1) and (5), we can conclude that simple rotation of the coordinate system from the fct to the bct lattice affects not only the sign of the in-plane fourfold anisotropy, but also the value of perpendicular anisotropies. From the last term in Eq. (8), one can see that

$$K_{1\parallel\text{bct}}^{\text{eff}} = -K_{1\parallel\text{fct}}^{\text{eff}}. \quad (9)$$

In addition, the second term in Eq. (8) contributes to the perpendicular uniaxial anisotropy and the third to the fourfold perpendicular anisotropy. The first term is just a constant that does not affect effective anisotropy fields. In our studies the fourfold perpendicular anisotropy can be neglected. However, the uniaxial anisotropy change due to rotation of the axis is important. By using Eqs. (3) and (8), one can compare the uniaxial anisotropies, $H_{u,\text{bct}}^{\text{eff}}$ and $H_{u,\text{fct}}^{\text{eff}}$, for bct and fct lattice coordinate systems, respectively:

$$H_{u,\text{bct}}^{\text{eff}} = H_{u,\text{fct}}^{\text{eff}} - \frac{3K_{1\parallel\text{fct}}^{\text{eff}}}{M_s}. \quad (10)$$

In Ref. 1, we calculated the effective anisotropy fields for the bct lattice coordinate system, but quoted the results for the fct lattice coordinate system changing the sign of in-plane fourfold anisotropies only. In the fct lattice coordinate system, the in-plane fourfold anisotropy energies $2K_{1\parallel}^{\text{eff}}/M_s$ are large negative numbers (see Table I of this paper), whose absolute values are equal to those of corresponding samples in Table I of Heinrich *et al.*¹ However, the effective perpendicular demagnetizing fields $4\pi M_{\text{eff}}$ and the perpendicular uniaxial anisotropy field $2K_u^{\text{eff}}/M_s$ in fct are larger in absolute value by $|3K_{1\parallel}^{\text{eff}}/M_s|$ (compare Table I of this paper with Table I of Ref. 1). Both analyses are valid and it is difficult to say which one is more appropriate.

A. Magnetic anisotropies

1. Perpendicular uniaxial anisotropies

The single-layered Co samples show large negative uniaxial anisotropy fields, indicating that the film surface is an easy plane (or, in other words, the surface normal is the hard axis) (see Table I). The uniaxial anisotropy is the result of magnetocrystalline and magnetoelastic energies. The shape anisotropy due to demagnetizing fields is eliminated from the uniaxial anisotropy by using the formula for a finite fcc lattice:¹

$$H_u = \frac{2K_u^{\text{eff}}}{M_s} = 4\pi \left[1 - \frac{0.2338}{d} \right] M_s - 4\pi M_{\text{eff}}, \quad (11)$$

where d is the film thickness in monolayers (ML). The thickness dependence of $2K_u^{\text{eff}}/M_s$ is shown in Fig 1(a) for liquid-nitrogen temperature and in Fig. 1(b) for room temperature. At both temperatures, uniaxial anisotropy field can be fit with a constant plus a term dependent on $1/d$:

$$\frac{2K_u^{\text{eff}}}{M_s} = \left[-13.6 - 82.2 \frac{1}{d} \right] \text{ kOe at LN}_2 \text{ T}, \quad (12a)$$

$$\frac{2K_u^{\text{eff}}}{M_s} = \left[-15.2 - 42.5 \frac{1}{d} \right] \text{ kOe at room temperature}, \quad (12b)$$

where d is in monolayers (ML). The sample with 1.7 ML of Co from Table I is omitted from the fitting procedure at 295 K, because its Curie temperature is below room

TABLE I. Single-layered Co(001) samples grown on Cu(001) substrate. All measurements carried out at 36 GHz. T_s is the temperature of the substrate during the growth of the Co layer. Data are presented in the face-centered-cubic coordinate system where $\alpha_x, \alpha_y, \alpha_z$ are directional cosines with respect to $\langle 100 \rangle$ axes ($g = 2.155$).

Sample	T_s (K)	H_u^* (kOe)	295 K			77 K		
			$4\pi M_{\text{eff}}$ (kOe)	$\frac{2K_{1\parallel}^{\text{eff}}}{M_s}$ (kOe)	H_u^* (kOe)	$4\pi M_{\text{eff}}$ (kOe)	$\frac{2K_{1\parallel}^{\text{eff}}}{M_s}$ (kOe)	
1.7Co/11.5Cu/20Au	330	-15.5	30.9	-0.065	-39.9	55.3	-0.60	
3.3Co/6Cu/20Au	375	-27.1	43.7	-0.85 ^a	-37.9	54.5	-2.09 ^a	
4Co/6Cu/20Au	300	-26.4	43.3	-1.10	-34.4	51.2	-2.21	
6.5/10.5Cu/20Au	300	-22.0	39.2	-1.25	-26.3	43.5	-2.04	
8.6/10.5Cu/20Au	300	-20.4	37.8	-1.27	-23.5	40.9	-1.83	
10Co/10Cu/20Au	355	-19.2	36.7	-1.29	-21.5	39.0	1.81	

*Using $4\pi M_s = 17.87 \text{ kOe}$ for hcp Co in Eq. (11).

^aThis sample had incorrect entry for the in-plane anisotropy in Ref. 1 due to erroneous use of 21.55 G instead of 17.87 G for $4\pi M_s$ in the calculations.

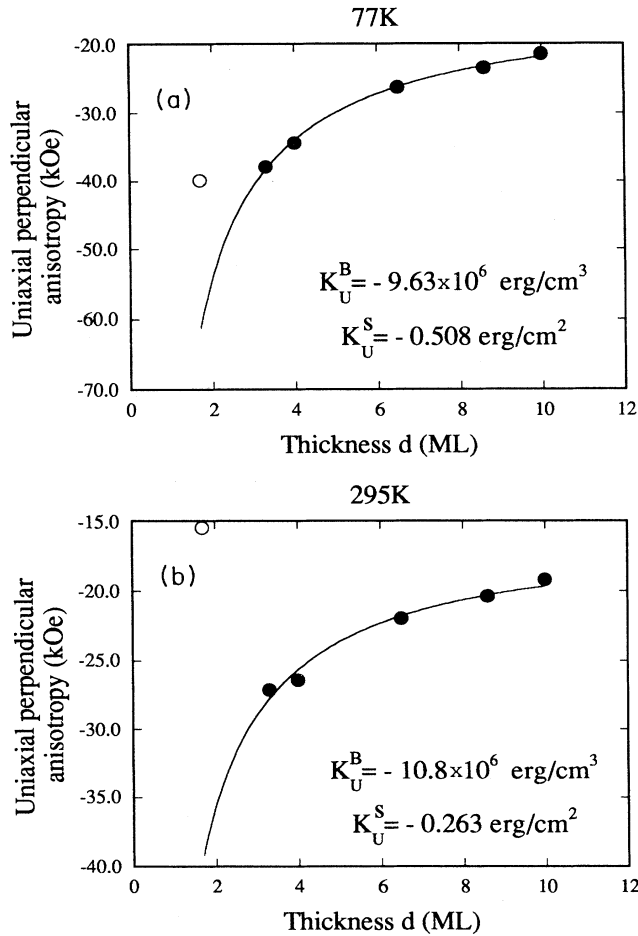


FIG. 1. The perpendicular uniaxial anisotropy as a function of the thickness of a Co(001) film grown on Cu(001) for (a) 77 K and (b) 300 K. The full circles indicate experimental data used to fit bulk and surface contributions to the anisotropy, as described by Eqs. (12) in the text. The empty circle indicates the 1.7-ML Co sample. Note that the anisotropy of this sample is smaller in magnitude at both temperatures compared to a constant plus $1/d$ -dependent term fit through the other points.

temperature.^{1,3} Corresponding anisotropy constants at liquid nitrogen temperature,

$$K_u^{\text{bulk}} = -9.63 \times 10^6 \text{ erg/cm}^3 = -0.963 \text{ MJ/m}^3, \quad (13a)$$

$$K_u^S = -0.508 \text{ erg/cm}^2 = -0.508 \text{ mJ/m}^2, \quad (13b)$$

and at room temperature,

$$K_u^{\text{bulk}} = -10.8 \times 10^6 \text{ erg/cm}^3 = -1.08 \text{ MJ/m}^3, \quad (13c)$$

$$K_u^S = -0.263 \text{ erg/cm}^2 = -0.263 \text{ mJ/m}^2. \quad (13d)$$

The shape anisotropy contribution to the bulk term was already subtracted using formula (11). The surface term value is quoted per one interface.

2. Fourfold in-plane anisotropies

The strong negative fourfold in-plane anisotropy fields favor the $\langle 110 \rangle$ as the easy axes (see Table I). The thickness dependence of this anisotropy is shown in Fig. 2(a) for liquid-nitrogen temperature and in Fig. 2(b) for room temperature. Again, a constant plus a term dependent on $1/d$ could describe the behavior:

$$\frac{2K_{1\parallel}^{\text{eff}}}{M_s} = \left[-1.51 - 3.01 \frac{1}{d} \right] \text{ kOe at LN}_2 \text{ T}, \quad (14a)$$

$$\frac{2K_{1\parallel}^{\text{eff}}}{M_s} = \left[-1.41 + 1.17 \frac{1}{d} \right] \text{ kOe} \quad \text{at room temperature,} \quad (14b)$$

where d is in ML. Samples 1.7 and 3.3 ML of Co were omitted from the fit. The values of bulk and surface anisotropy constants are given below for reference: At liquid-nitrogen temperature,

$$K_{1\parallel}^{\text{bulk}} = -1.07 \times 10^6 \text{ erg/cm}^3 = -0.107 \text{ MJ/m}^3, \quad (15a)$$

$$K_{1\parallel}^S = -1.86 \times 10^{-2} \text{ erg/cm}^2 = -1.86 \times 10^{-2} \text{ mJ/m}^2. \quad (15b)$$

At room temperature,

$$K_{\parallel}^{\text{bulk}} = -1.00 \times 10^6 \text{ erg/cm}^3 = -0.100 \text{ MJ/m}^3, \quad (15c)$$

$$K_{\parallel}^{\text{S}} = 7.24 \times 10^{-3} \text{ erg/cm}^2 = 7.24 \times 10^{-3} \text{ mJ/m}^2. \quad (15d)$$

The values of surface terms correspond to one interface only.

III. DISCUSSION

For an ideal fcc lattice there would be no bulk contribution to the perpendicular uniaxial anisotropy. Therefore, large negative constant terms in Eqs. (12) are a

consequence of the tetragonal distortion of Co lattice grown on Cu(001). This contribution favors the surface of a film as an easy plane. The thickness dependent part of $2K_u^{\text{eff}}/M_s$ is well described by the $1/d$ term, and it also favors an easy plane. LEED studies indicate a constant and homogeneous strain for Co/Cu(001) films thicker than 4 ML,⁴ and therefore the measured $1/d$ terms in these films are due to the interface anisotropies, not thickness-dependent magneto-elastic energy. This is in contrast to Co/Cu(111) superlattices where the measured apparent surface anisotropies are a consequence of the magneto-elastic energy with strains inversely proportional to the Co layer thickness.⁵ Our results indicate that a 4 ML Co/Cu(001) sample also follows the constant strain behavior. But thinner samples deviate from the constant plus thickness-dependent term behavior in Eqs. (12) and (14), as discussed below.

The 1.7-ML sample for both the uniaxial and the in-plane anisotropies and the 3.3-ML samples for the fourfold in-plane anisotropy were excluded from the fitting procedure. The reason for this is twofold. First, as mentioned in the Introduction, detailed LEED investigations revealed that the strain in Co films thinner than 4 ML is thickness dependent. For Co coverages of less than 4-ML tetragonal distortions are smaller,⁴ thus resulting in the reduction of the magneto-elastic volume contribution to magnetic anisotropies. Second, scanning tunneling microscopy (STM) studies of Co grown on Cu(001),⁷ and our reflection high-energy electron-diffraction (RHEED) oscillations,¹ indicate that the first two layers of Co exhibit deviation from a layer-by-layer growth. The islands belonging to the second atomic layer are created before the first layer is completed. Such roughness causes local demagnetizing fields at the surface which tend to decrease the surface anisotropy (make it less negative). Hence, for samples thinner than 4 ML, the deviation from the simple constant volume and $1/d$ dependent surface term behavior is due to both smaller strains and increased roughness.

The role of surface roughness and changing lattice strains discussed above is visible in the measured uniaxial and in-plane anisotropies plotted in Figs. 1 and 2. While the uniaxial anisotropy of the 1.7-ML sample at 295 K is strongly decreased [by $\sim 60\%$, see Fig. 1(b)] mainly because the Curie point is just below room temperature, the observed lower value [by $\sim 20\%$, see Fig. 1(a)] of the uniaxial anisotropy at 77 K (well below T_c) is caused mostly by the reduced strain and the increased surface roughness. Those effects are even more pronounced in the fourfold in-plane anisotropy. At both 77 and 295 K temperatures the fourfold in-plane anisotropy of the 1.7-ML Co sample is decreased by $\sim 80\%$ and $\sim 90\%$, respectively, see Fig. 1. The 3.3-ML sample showed an appreciably smaller decrease in the in-plane anisotropy; 12% at LN₂ temperature, and 19% at room temperature. Considering the fact that the Co film surface topology does not change above 3 ML, this reduction could be attributed mainly to the reduced strain in the 3.3-ML thick Co.

The value of magnetoelastic anisotropy can be estimated using equations derived along the lines of Neel's theory:⁶

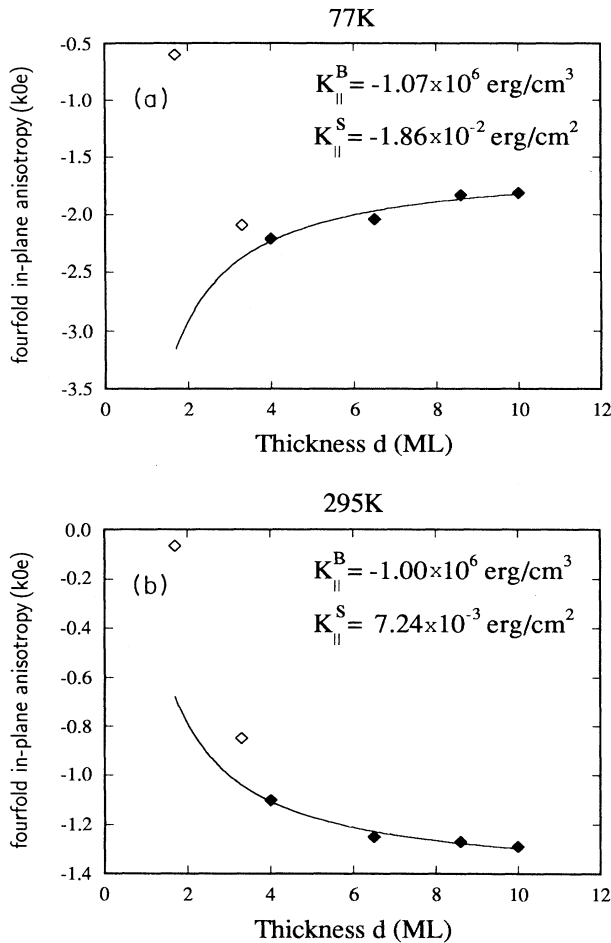


FIG. 2. The fourfold in-plane anisotropy as a function of the thickness of a Co(001) film grown on Cu(001) for (a) 77 K and (b) 300 K. The full diamonds indicate experimental data used to fit bulk and surface contributions to the anisotropy, as described by Eqs. (14) in the text. Note the reversal in thickness dependence upon cooling from room to LN₂ temperature. The empty diamonds indicate 1.7- and 3.3-ML Co samples which were not used in the fit (see text).

$$H_u^{\text{mag.el.}} = B_1(e_{\perp} - e_{\parallel})\alpha_z^2, \quad (16)$$

$$B_1 = -\frac{3}{2}\lambda_{100}(c_{11} - c_{12}),$$

where e_{\perp}, e_{\parallel} are an in-plane and a perpendicular strain tensor components, λ_{100} is a magnetostriction constant, and c_{11}, c_{12} are elastic moduli. In order to explain the strength and sign of the volume contribution to the perpendicular uniaxial anisotropy using Néel's linear magnetoelastic theory, one needs to use a positive magnetostriction constant of $\lambda_{100} \approx 1 \times 10^{-4}$ for lattice strains observed in fct Co. This value is close to those found in fcc Pd-Co alloys ($\lambda_{100} \approx 1.3 \times 10^{-4}$)^{8,9}, and has the same sign and order of magnitude as magnetostriction constants of Co-Fe and Co-Ni alloys. In addition, the bulk term for our fcc Co/Cu(001) films, $K_u^{\text{bulk}} = -10.8 \times 10^6 \text{ erg/cm}^3$, and that for fcc Co/Pd(001) films of $K_u^{\text{bulk}} = -3.2 \times 10^7 \text{ erg/cm}^3$ (Refs. 8 and 10) have a ratio that closely corresponds to the ratio of their vertical compressions [3%:9.6% (Ref. 8)]. This fact emphasizes the importance of tetragonal distortion in the strength of uniaxial anisotropies.

In contrast to our Co/Cu(001) films, surface contributions to the uniaxial anisotropy in Co/Pd(001),(110),(111) (Ref. 10) and Co/Pt(001),(110),(111) (Refs. 11 and 12) films are positive favoring the film normal as the easy axis. A negative value of the surface anisotropy in Co/Cu(001) structures is very surprising considering the fact that even the formation of an ordered alloy (CoPt₃) in Co/Pt interfaces did not result in substantial modification of surface anisotropies.^{13,14}

The thickness-dependent part of the uniaxial anisotropy is strongly dependent on temperature, while the thickness-independent part does not change appreciably upon cooling to liquid-nitrogen temperature. A 10% change in the volume term can be attributed to the temperature dependence of the magnetostriction constant in Eq. (16). For example, for fcc Ni, the change in the magnetostriction constant λ_{100} upon cooling from 300 to 77 K is approximately 9%;¹⁵ therefore, λ_{100} for fcc Co might show a similar change. In order to explain the temperature dependence of the $1/d$ term using the renormalized surface anisotropy due to fluctuations caused by two-dimensional (2D) spin-wave theory,¹⁶ one needs a decrease in saturation magnetization of approximately 17%. Unfortunately, the authors are not aware of any published studies related to the dependence of magnetization on temperature for samples 4 ML and thicker of Co grown on Cu(001).

The fourfold in-plane anisotropy has a large constant term due to lattice strains, and a $1/d$ -dependent term most likely due to a surface fourfold in-plane anisotropy. The constant term favors $\langle 110 \rangle$ as easy axes at both liquid-nitrogen (LN)₂ and room temperatures. The thickness-dependent term's behavior is striking: at LN₂ temperature its negative value also prefers $\langle 110 \rangle$ as easy axes, but upon heating to room temperature its sign changes and favors $\langle 100 \rangle$ as easy axes [see Eqs. (14)]. Such behavior is not likely due to thermal fluctuations; for samples 3 ML and thicker, the critical point is above 600 K,³ and hence for room temperature the thermal

fluctuations should be negligible.¹⁷ This reversal in sign of the thickness-dependent term upon heating from 77 to 300 K might be caused by a complex dependence of the spin-orbit contribution to $3d$ valence-band energies on the sample temperature. On the other hand, the volume term is not significantly affected by temperature change. This is in a complete contrast to bcc Fe (Ref. 6) which changes its thickness-independent part by nearly 30% upon cooling from room to liquid-nitrogen temperature, but leaves its $1/d$ term virtually unchanged.

Recently, magnetic anisotropies of Co grown on Cu(001) have been determined by Brillouin light scattering.¹⁸ Studies of the fourfold in-plane anisotropy by Krams *et al.* showed the same sign and order of magnitude for volume and surface contributions as our results. The uniaxial perpendicular anisotropy determined in that paper, however, did not exhibit a volume contribution apparent in our studies, and for Co covered with 2-ML Cu, the thickness-dependent term indicated that the surface normal was an easy axis in disagreement with our result.

Calculations based on Néel's model predicted perpendicular surface anisotropies for Co/Au and Co/Cu textured films in accordance with experiment.¹⁹ It is therefore interesting to compare our results with the value of the surface anisotropy calculated from Néel's model. Using $l = 6.1 \times 10^{-16}$ ergs as calculated for hcp cobalt in Ref. 19 we get

$$K_u^S = \frac{l}{a^2} = 0.47 \frac{\text{ergs}}{\text{cm}^2}, \quad (17)$$

where a is the in-plane lattice constant. The positive K_u^S indicates a perpendicular surface anisotropy for the case of Co(001), in contradiction to our experimental results. This should not be too surprising since Néel's approach assumes a free surface, not a covered one, as investigated in our studies. Calculated $K_u^S = 0.47 \text{ ergs/cm}^2$ also does not agree with Krams *et al.*'s¹⁸ finding of a strong in-plane anisotropy on the uncovered Co(001) surface. These results suggest that predictions of the behavior of the surface anisotropy invoking Néel's model have to be treated with some precaution. To estimate the size or even the sign of the surface anisotropy, detailed *ab initio* band structure calculations have to be performed.

IV. SUMMARY

Ultrathin structures of Co on Cu(001) were grown using molecular-beam epitaxy (MBE). The magnetic properties of such films were investigated using FMR technique. The differences in analysis of magnetic anisotropies in two coordinate systems, fct and bct, were demonstrated.

Co(001) films on the fcc Cu(001) substrate show strong perpendicular uniaxial anisotropy with its hard axis along the film normal. The fourfold in-plane anisotropy is also strong with its easy axes along the $\langle 110 \rangle$ crystallographic directions. Both anisotropies can be represented by bulk and surface contributions. The bulk terms are mainly attributed to tetragonal distortions resulting from lattice mismatch between metastable fcc Co(001) and fcc

Cu(001). The surface terms are most likely due to broken symmetry at the interfaces. The surface uniaxial perpendicular anisotropy in Co/Cu(001) structures is negative in contrast to other Co structures which possess a positive surface anisotropy.

The uniaxial anisotropies in the 1.7-ML sample and the in-plane fourfold anisotropies in the 1.7-ML and 3.3-ML samples exhibit deviation from a simple behavior described by constant plus $1/d$ terms. The uniaxial and the fourfold in-plane anisotropies in the 1.7-ML sample at 77 K are lowered (in absolute value) due to the decreased strain and the increased surface roughness. The magnetic anisotropies in the 1.7-ML sample at 295 K are additionally decreased due to T_c being just below room temperature. The decrease in fourfold in-plane anisotropies in the 3.3-ML sample can be mostly attributed to reduced strain.

The temperature dependence of both anisotropies is very strong. The bulk terms are not appreciably changed upon cooling from room to liquid-nitrogen temperatures, while the surface terms are very strongly affected by the cooling. In the case of uniaxial anisotropy, the bulk term

is most likely affected by the temperature dependence of the magnetostriction constant. The $1/d$ term behavior might be explained using fluctuations due to 2D spin-wave theory if the change in magnetization upon heating from 77 to 300 K is $\sim 17\%$. Temperature behavior of fourfold in-plane anisotropy is intriguing, and cannot be explained by a simple model.

Note added in proof. Recently the authors became aware of an EXAFS study of the Co/Cu(001) system, which confirms presence of the tetragonal distortion in the cobalt films.²⁰

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