Suppression of the magnetocrystalline bulk anisotropy in thin epitaxial Co(110) films on Cu(110)

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We report on an unexpected suppression of the magnetocrystalline anisotropy contribution in epitaxial fcc Co(110) films on Cu(110) below a thickness of $d_c = (50 \pm 10)$ Å. For film thicknesses larger than d_c the measured anisotropy value agrees with published data. Measurements on films with reduced strain indicate a large strain dependence of d_c. A model calculation based on a crystal-field formalism and discussed within the context of band theory, which explicitly takes tetragonal misfit strains into account, reproduces the experimentally observed anomalies. Our results indicate that the usually applied phenomenological description of anisotropies, assuming additive free energy terms for each anisotropy contribution, fails in this case.

Progress in understanding the physical origins of magnetic anisotropies in thin magnetic films is largely determined by the accuracy and unambiguity of experimental data and by the success of a more sophisticated theoretical modeling. For uniaxial anisotropy contributions a close agreement between measurements and calculations has been achieved using both band structure calculations[1] and phenomenological models.[2] For higher order anisotropies success has been poor.[3,4] This is in part due to the requirements in higher energy resolution, wherein standard theoretical approaches, such as total energy difference calculations based on band structure data, fail.[3,4] On the experimental side these contributions are also harder to determine, in particular since they are often one or two orders of magnitude smaller compared to uniaxial contributions also present in thin films. Here we would like to point out that in particular for films of cubic crystallographic symmetry, the fourth-order cubic anisotropy is of key importance in understanding thin film magnetic anisotropy properties, since in the respective bulk materials this anisotropy contribution is of lowest order.

In this paper we show that the transformation of the fourth-order, magnetocrystalline bulk anisotropy into uniaxial contributions in the thin film regime as a consequence of tetragonal misfit strains is a characteristic feature of thin film anisotropy. This is demonstrated experimentally for the system of Co(110) films on single-crystalline Cu(110) substrates. The (110) orientation allows us to differentiate between cubic and uniaxial anisotropy contributions due to different symmetry axes, even if these contributions differ by more than an order of magnitude. We present model calculations based on a crystal-field formalism with energies and parameters interpreted within the context of a band theory. We obtain a transformation of cubic anisotropy into in-plane and out-of-plane uniaxial contributions as a function of the uniaxial distortion of the unit cell due to misfit strain. Our theoretical results are in good agreement with our experimental findings.

We have investigated the thickness dependence of all contributing anisotropies and the correlation of the set of determined anisotropy constants on strain and strain relaxation processes in the regimes of pseudomorphic growth and of lattice relaxation due to dislocation formation. The detailed results of the correlation between anisotropies and film strains are reported elsewhere.[5,6] Here we only report those relevant to the investigation of the magnetocrystalline anisotropy. In order to distinguish between the magnetocrystalline anisotropy contribution from uniaxial contributions (both Néel and magnetoelastic contributions) we chose the (110) film orientation: It contains both the [001] and [1$\bar{1}$0] axes, which are the symmetry axes for uniaxial anisotropies, as well as the [1$\bar{1}$1] and [1$\bar{1}$ $\bar{1}$] axes, which are easy axes for the magnetocrystalline anisotropy in fcc bulk cobalt.[7] We have measured the spin wave frequencies as a function of the in-plane angle of the applied external field using Brillouin light scattering from thermally excited dipolar spin waves propagating along the film plane (Damon-Eshbach modes).[8–12] An analysis of the spin wave frequencies using a full spin wave model[11,12] yields the anisotropy constants. Within the investigated Co layer thickness regime the saturation magnetization is independent of thickness within an error margin of $\pm 5\%$.

The samples used in the present study were molecular-beam-epitaxy grown onto Cu(110) single-crystal substrates in ultrahigh vacuum (base pressure $< 10^{-10}$ mbar) with deposition rates of 0.3 Å/s for Co and 0.2 Å/s for Cu.[5,6] Both a wedge-shaped Co film (thickness range 0–40 Å) and two staircase-shaped films were prepared by withdrawing a shutter during deposition covered by a 12-Å-thick Cu layer and a 25-Å-thick Au protective layer. From low-energy electron diffraction (LEED) -$I(V)$ measurements a perpendicularly distorted fcc crystal structure is inferred for the investigated thickness range.

In the following we discuss anisotropies in a coordinate system oriented such that the $\hat{x}$ and $\hat{y}$ axes are parallel to the

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film plane along the [001] and [1$\bar{1}$0] directions with the $\hat{z}$ axis normal to the film plane; in a second, crystallographic, reference frame $\hat{x}'$, $\hat{y}'$, and $\hat{z}'$ are the unit vectors oriented along the principal crystallographic axes. We describe the magnetic anisotropies by (i) assuming cubic symmetry of the film, represented by a cubic magnetocrystalline bulk anisotropy constant K_1, and (ii) describing the tetragonal distortions from cubic symmetry by two additional uniaxial anisotropy contributions, $K_{\text{in-plane}}$ and $K_{\text{out-of-plane}}$. With $\vec{\alpha}$ the directional unit vector of the magnetization with components α_x, α_y, and α_z expressed in the film coordinate system or $\alpha_{x'}$, $\alpha_{y'}$, and $\alpha_{z'}$ expressed in the crystallographic reference frame, the free anisotropy energy is then expressed as

$$F_{\text{ani}} = K_1(\alpha_x^2\alpha_y^2 + \alpha_y^2\alpha_z^2 + \alpha_z^2\alpha_x^2) + K_{\text{in-plane}}\alpha_x^2 - K_{\text{out-of-plane}}\alpha_z^2. \quad (1)$$

It is observed that the shape anisotropy causes the magnetization to lie in the film plane for the investigated Co thickness range (8 Å–110 Å). Therefore we can set $\alpha_z = 0$ to establish the static in-plane equilibrium direction.

Figure 1 (top, full symbols) shows the obtained results for the magnetocrystalline anisotropy constant K_1 as a function of the film thickness d. For comparison the in-plane and out-of-plane uniaxial anisotropies, $K_{\text{in-plane}}$ and $K_{\text{out-of-plane}}$, are shown as well (Fig. 1, middle and bottom). For d larger than 50 Å a thickness-independent value of $K_1 = (-0.85 \pm 0.05) \times 10^6$ erg/cm^3 is found, which agrees with literature values.[7] For Co film thicknesses smaller than 50 Å we find a sudden breakdown in the magnetocrystalline anisotropy. The breakdown is accompanied by a maximum in the absolute value of the uniaxial in-plane anisotropy (Fig. 1, middle).

To gain further insight, Co(110) films have been grown onto a 60-Å-thick $Cu_{62}Ni_{38}$ buffer layer deposited onto a (110)-oriented Cu single-crystal substrate. The idea was to reduce the film strains by about a factor of 2 due to the smaller lattice mismatch of 1% between the CuNi buffer layer and the Co layer. As displayed in Fig. 1 (top, open symbols) we find that the onset of the suppression of the magnetocrystalline anisotropy is shifted to larger thicknesses, clearly demonstrating that the suppression is strain induced.

The data indicate that the presence of a uniaxial strain strongly suppresses the cubic anisotropy. We now outline a phenomenological approach that provides some insight into the relationship between these second- and fourth-order anisotropies. A preliminary account of the model was given in Ref. 13. Our model gives a natural dependence of the cubic anisotropy on symmetry breaking effects which produce second-order anisotropies.

The existence of a uniaxial surface anisotropy was predicted by Néel as a consequence of the electronic symmetry breaking which occurs at the surface due to lower atomic coordination.[14] Recently band theoretical methods have made considerable progress in the calculation of anisotropies which result from Néel effects.[15–18] Notably *ab initio* calculations for a Co(001) film are reported by Wang *et al.*,[15] and a tight-binding calculation in which only d states are considered for a Co(110) film has been performed by Cinal

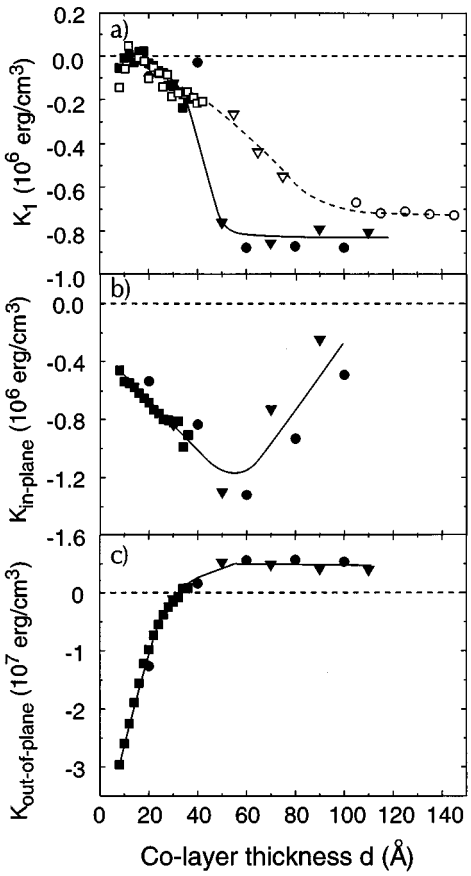

FIG. 1. Magnetocrystalline anisotropy constant K_1 (top), in-plane anisotropy constant $K_{\text{in-plane}}$ (middle), and out-of-plane anisotropy constant $K_{\text{out-of-plane}}$ (bottom) as a function of the film thickness for Co(110) films grown on Cu(110) (full symbols). The different symbols denote different samples (■: wedge-type sample; ● and ▼: staircase-type samples). The open symbols in (a) denote measurements on Co(110) films on a CuNi buffer layer on top of a Cu(110) substrate. The solid and dashed lines are guidelines to the eye.

et al.[16,17] However, the calculation of fourth-order anisotropies requires an energy resolution beyond the scope of current computational methods and consequently they tend to be limited to second-order phenomena. Lorenz and Hafner[18] employ an approach which to some extent avoids these problems. They are able to consider anisotropies which arise to a few μeV. Their methods are promising but have yet to be applied to a situation as complex as ours.

In multilayer structures one also expects the presence of an interface anisotropy. Chappert and Bruno[19] suggest that lattice misfit strains between a single layer and its substrate may significantly contribute to interface anisotropy through magnetostriction. The above treatments focus on the Néel anisotropy, taking the strain to be uniform throughout the system. They have therefore been unable to explicitly consider its effects. In view of this we consider a phenomenological model in which changes to anisotropy energies which arise to both second and fourth order are included. Our approach is founded upon symmetry considerations which explicitly take lattice misfit strains into account.

We use a simple crystal-field Hamiltonian to consider the

changing symmetry of the system. The wave functions relevant to our analysis are $x'y'$, $y'z'$, $x'z'$, x'^2-y'^2, and $3z'^2$-r^2 with x',y',z' the Cartesian coordinates of the electrons in the crystallographic reference frame and $r^2=x'^2+y'^2+z'^2$. The surface normal is along $z=(x'+y')/\sqrt{2}$. We consider a Hamiltonian in terms of Stevens's operators[20] in the form

$$H=A(l_{x'}^4+l_{y'}^4+l_{z'}^4)+X(l_{x'}+l_{y'})^2, \tag{2}$$

where A and X are the cubic and uniaxial energy parameters. From (2) it is apparent that we consider a strain upon cubic symmetry along the growth direction. We calculate anisotropies in the usual way by including the spin-orbit coupling as a perturbation.[21] Assuming that the exchange splitting is very large compared with the spin-orbit coupling, we may write the perturbation for the magnetization along the axis ν as

$$E^{SO}=\xi\vec{l}\cdot\vec{s}\approx\xi l_\nu/2. \tag{3}$$

The anisotropy energy is found by calculating the change in the ground state energy for different directions, ν, as a power series in the spin-orbit coupling constant, ξ. Hence, we obtain expressions for the anisotropy energy to both second and fourth order in ξ.

In the cubic limit there exist the familiar e_g and t_{2g} degeneracies. For increasing coverage the system approaches this cubic limit and the $3z'^2$-r^2 and x'^2-y'^2 states form the e_g orbital while $x'y',y'z',x'z'$ states compose the t_{2g} orbital. Any anisotropy K is then calculated by extrapolating between the two regimes according to

$$K=\frac{\sum_{i=1}^{n} K_i\exp(E_i/\Delta)}{\sum_{i=1}^{n}\exp(E_i/\Delta)}, \tag{4}$$

where K_i and E_i represent the anisotropy and energy, respectively, calculated for the ith crystal-field state. The Δ is a mixing parameter which we take to be $\xi/4$.

The uniaxial energy parameter X is by definition proportional to the misfit strain ϵ. Chappert and Bruno[19] and also den Broeder *et al.*[22] argue that ϵ is inversely proportional to the film thickness and so we plot the anisotropies as functions of A/X which is therefore proportional to the film thickness. The calculated anisotropies are plotted in Fig. 2.

We see that K_1 is suppressed for increasing strain. We can understand this physically from our model. The fourth-order anisotropy energy depends upon $\xi^4/(\text{excitation energy})^3$, where the relevant excitation energy is some combination of X and A: Hence for $X=0$, K_1 is proportional to ξ^4/A^3, but for $X\gg A$ we find K_1 is proportional to ξ^4/X^3. This qualitative behavior of the model is independent of the sign of A or X and hence of our crystal-field ground state that we impose. However, in order for the cubic anisotropy to have the correct sign we take $A>0$. Furthermore, $X>0$ forces the $3z'^2$-r^2 state to lie highest and so to be the (hole) ground state in the noncubic limit. This is in accordance with Wang *et al.*,[15] who have shown the hole state to be predominantly of $3z'^2$-r^2 character.

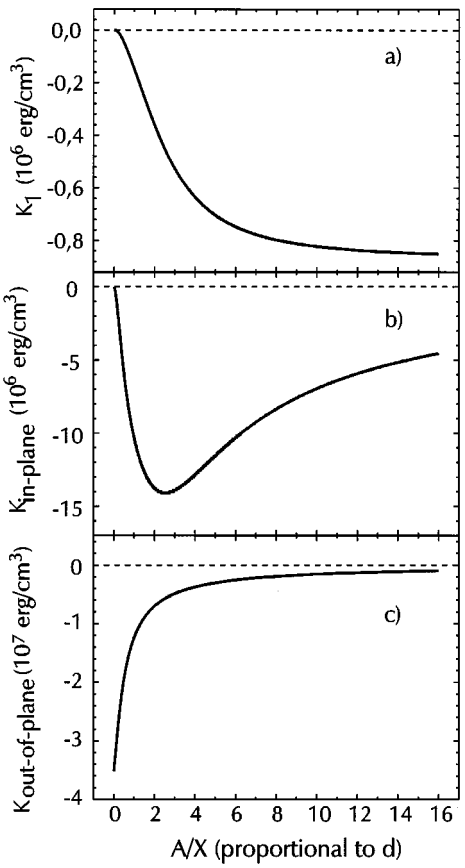

FIG. 2. Calculated cubic anisotropy constant K_1 (top), in-plane anisotropy constant $K_{\text{in-plane}}$ (middle), and out-of-plane anisotropy constant $K_{\text{out-of-plane}}$ (bottom) as a function of the ratio of the cubic and uniaxial energy parameters A/X.

The problem is now open to interpretation within the context of band theory. Each Bloch state at the Fermi surface is a linear combination of d states. When we sum these states over the star of k we generate a combination of Bloch states which reflects the overall crystal symmetry. Thus our crystal-field states can be regarded as the Brillouin zone averaged contribution of each state to the Fermi surface. The crystal-field symmetries reflect those present in full band theory treatment.

We take A and ξ to be fitting parameters such that (i) K_1 approaches its experimental value, and (ii) we obtain the correct value for $K_{\text{out-of-plane}}$ in the limit $X\gg A$. This results in the correct magntitude of $K_{\text{out-of-plane}}$. We have $A/\xi=8.8$ and $\xi=8$ meV/Co. Our values of ξ are approximately 10% of that used by Cinal *et al.*[16,17] Given the simplicity of our model, in which effects of the spin-orbit coupling will be overestimated, we regard this as satisfactory. The agreement of the magnitude of the measured value of $K_{\text{in-plane}}$ is poor. However, the fact that such a simplistic approach yields qualitative agreement we regard in itself as important.

The positive value of $K_{\text{out-of-plane}}$ for $d>32$ Å, not reproduced by the calculations, is likely due to the formations of bulk dislocations. We expect that in this thickness regime the corresponding dislocation diffusion length becomes smaller than the layer thickness. Studies are underway to quantitatively determine these strain components, but this is outside the scope of this paper.

In summary, we have shown that the magnetic anisotropies of Co(110) films change from strongly tetragonal for thin films to dominantly cubic in the thick film limit. The important result of this paper is that *all* the observed anisotropy constants change as the effects of the tetragonality start to dominate. The theoretical model demonstrates that this is to be expected because the dominance of the energies of tetragonal symmetry in the Hamiltonian over those of cubic symmetry changes all the energy levels and hence all the anisotropies. These conclusions hold, irrespective of whether the anisotropy is electronic in origin or due to induced strain. The observed anomaly points to the limitations of the widely used underlying phenomenological approach for describing magnetic anisotropies in thin films: It is often assumed that the various anisotropy contributions, written as free energy density terms, are additive; our data and calculations clearly provide evidence that this is not true in general. On the other hand, such a failure is not so surprising if one takes into account that the magnetoelastic, magnetocrystalline, and the Néel-type surface anisotropy contributions originate from the same microscopic mechanism, which is spin-orbit coupling. As it turns out, the usually applied phenomenological descriptions might well work for uniaxial anisotropy contributions, but they fail in the presence of higher-order contributions.

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