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Magnetic anisotropies of ultrathin Co films on Cu(1 1 13) substrates

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All magnetic anisotropy contributions in single-crystalline (1 1 13)-oriented Co films on Cu were determined at room temperature using Brillouin light scattering. An in-plane uniaxial anisotropy contribution, not found in the reference system Co/Cu(001), is identified as magnetoelastic in origin. From the thickness dependence of the in-plane anisotropies of both twofold and fourfold symmetry about the film normal it is concluded that elastic strain fields are very likely to provide the driving mechanism for stabilizing ferromagnetic order in ultrathin Co(1 1 13) films.

Magnetic properties of ultrathin films of 3*d* transition metals are inherently determined by magnetic anisotropies which are found to be up to 3 orders of magnitude larger than in the bulk materials. Apart from the shape anisotropy Néel-type surface anisotropies,[1] magnetoelastic anisotropies due to elastic strains,[2] and magnetocrystalline anisotropy contributions are discussed. Although the experimental database on anisotropy constants in thin films is steadily increasing little is known up to now about the origin of the observed large anisotropy values.

Recently, all relevant anisotropies were determined at room temperature in epitaxial fcc Co(001) films deposited on Cu(001) single crystals, both uncovered and covered with 2 monolayers (ML) of Cu, using Brillouin light scattering.[3,4] Apart from a perpendicular surface anisotropy contribution, an in-plane anisotropy of fourfold symmetry about the film normal was found. This anisotropy $K^{(4)}_{\text{in-plane}}$ could be separated into a thickness independent volume term $K_p^{(4)}$ and a surface term $k_p^{(4)}$:

$$K^{(4)}_{\text{in-plane}} = K_p^{(4)} + \frac{2}{d} k_p^{(4)} \tag{1}$$

with d the film thickness and the factor of 2 counting the two surfaces. It was found[3,4] that the two right-hand side terms of Eq. (1) cancel each other at a critical thickness d_c^*, which coincides with the minimum thickness d_c for the onset of ferromagnetic order at room temperature, which is obtained independently from magneto-optic Kerr effect (MOKE) measurements and from the observation of spin waves. This result indicates that the mechanism for stabilization of ferromagnetic order is closely connected to the properties of magnetic anisotropies acting in these films.

However, the origins of these anisotropy contributions are largely unknown up to now. Experiments on Co films covered with 2-ML Cu show that the out-of-plane anisotropy is drastically changed (even the signs are different for the Co/Cu and the Co/vacuum interfaces) ruling out a standard Néel-type surface anisotropy mechanism which is caused by the broken translational symmetry at the surface along its normal. Unfortunately, the in-plane anisotropy of fourfold symmetry is not easily accessible by standard theoretical concepts due to its higher-order nature.

To achieve a better understanding of the nature of the involved anisotropies we have performed experiments on Co films grown on Cu surfaces with a slightly differing orientation as shown in Fig. 1. As substrates we used Cu(1 1 13) which mainly consists of terraces separated by monoatomic steps. The terrace width is 6.5 atomic distances in average.[5] The step edges are aligned with the $[1\bar{1}0]$ direction. Films grown on such templates exhibit a twofold in-plane symmetry. The films were grown at room temperature after carefully cleaning the substrate by sputter and annealing cycles as described elsewhere.[5,6] The formation of terraces and the narrow distribution of the terrace widths were observed by a characteristic splitting of the spots of the (001) low-energy electron-diffraction (LEED) patterns. After deposition of the Co films they were covered by a 20-ML thick protective Cu layer for the *ex situ* Brillouin light scattering measurements.

Due to the reduced surface symmetry a new, lower-order anisotropy contribution is likely to occur. Its existence, but not its strength, has been previously found by scanning electron microscopy with spin-polarization-analysis measurements[6] and by MOKE measurements.[7] On our set of samples transverse MOKE measurements

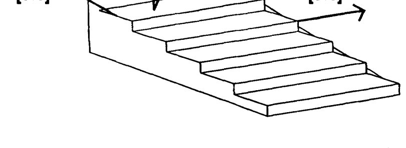

FIG. 1. Schematic representation of the Cu(1 1 13) surface, consisting of (001) terraces of 6.5 atoms width on average.

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of the remanent field as a function of the in-plane direction of the externally applied field show clearly that the symmetry of the free anisotropy energy is reduced to twofold.[8]

In order to quantitatively determine both the in-plane anisotropy contributions of twofold and fourfold symmetry about the film normal Brillouin light scattering experiments were carried out. The details of the experimental procedure and the underlying theory are described elsewhere.[3,9,10] Here we need to know that the spin-wave frequencies depend on the magnitude and symmetry of the involved anisotropies, the magnitude and direction of the saturation magnetization, the wave vector of the spin waves determined by the scattering geometry, and the magnitude and direction of the applied external field.[10] Figure 2 shows the measured spin-wave frequencies for a 9-ML thick Co film as a function of the direction of the in-plane applied external field, **H**. The direction of **H** is measured by the angle ϕ_H with respect to the in-plane approximate [100] direction, i.e., the direction which corresponds to the [100] direction of the (001) surface (see inset of Fig. 2). Maxima in the spin-wave frequency indicate an easy direction. A persisting in-plane anisotropy of fourfold symmetry is identified by the relative maxima at −45° and +45°, i.e., with the easy axes along the in-plane approximate ⟨110⟩ directions. A superimposed twofold anisotropy is evident from the different heights of the maxima at −45° and +45° with the $[1\bar{1}0]$ direction as the easy axis.

We have analyzed a whole set of data, equivalent to the data shown in Fig. 2, obtained for films of different thicknesses ($d=4$, 6, 8.2, 9, 10, 12, and 14 ML) by using the following expression for the free anisotropy energy F_{ani}, entering the spin-wave dispersion relation:[10]

$$F_{\text{ani}}=\left[K_p^{(2)}+\frac{2}{d}k_p^{(2)}\right]\cos^2\left[\phi-\frac{\pi}{4}\right]\sin^2\theta+\frac{1}{4}\left[K_p^{(4)}+\frac{2}{d}k_p^{(4)}\right]\sin^2 2\phi\sin^4\theta-\left[K_s+\frac{2}{d}k_s\right]\cos^2\theta\ . \tag{2}$$

θ and ϕ are the polar and azimuthal angle of the direction of magnetization, **M**. Here ϕ is measured with respect to the in-plane approximate [100] direction. The first term on the right-hand side is the in-plane anisotropy of twofold symmetry about the film normal with the angle $\pi/4$ measuring the angle between the $[1\bar{1}0]$ direction and the [100] direction. The second term corresponds to a fourfold symmetry and the last term is the out-of-plane anisotropy. All terms are further split into volume and surface contributions as in Eq. (1).

By use of Eq. (2) the measured spin-wave frequencies were fitted using an appropriate model described elsewhere[3,10] with the anisotropy constants as fit parameters. In the least-squares fit much care has been taken to control the errors and correlations between the parameters. The obtained values for the two in-plane anisotropy constants of twofold and fourfold symmetry, $K_{\text{in-plane}}^{(2)}$ and $K_{\text{in-plane}}^{(4)}$, multiplied by the film thickness d are displayed as a function of d in Fig. 3. From Eq. (1) it follows that the data should lie on a straight line with the intercept equal to twice the surface anisotropy constant and the slope equal to the bulk anisotropy constant. The experimental data fulfills this condition within the experimental error. From the data critical thicknesses are deduced at which the corresponding volume and surface anisotropies cancel to zero: For the twofold uniaxial anisotropy this thickness is $d_c^{(2)}=(2.9\pm0.6)$ ML and for the fourfold anisotropy $d_c^{(4)}=(2.2\pm0.4)$ ML. We find within the experimental error that $d_c^{(2)}=d_c^{(4)}$. The critical thicknesses are compatible with the onset of ferromagnetic order at 1.8 ML observed *in situ* for uncovered films by MOKE.[11] We would like to point out that although the film growth mode is very different for the (1 1 13) orientation compared to the (001) orientation,[3,4,11] the critical thickness for the onset of ferromagnetic order is the same within the error margins of both orientations.

From the data analysis we find for the twofold in-plane anisotropy $K_p^{(2)}=(6.0\pm0.7)\times10^5$ erg/cm³ and $k_p^{(2)}=(-0.009\pm0.002)$ erg/cm², and for the fourfold contribution $K_p^{(4)}=(-6.5\pm0.2)\times10^5$ erg/cm³ and $k_p^{(4)}=(0.012\pm0.002)$ erg/cm².

For $d>d_c^{(2)}$ the $[1\bar{1}0]$ direction (parallel to the steps) is the easy axis for $K_{\text{in-plane}}^{(2)}$, and for $d>d_c^{(4)}$ the in-plane approximate ⟨110⟩ directions are the easy axes for $K_{\text{in-plane}}^{(4)}$. For $K_{\text{out-of-plane}}$ no surface dependent contribution k_s was found, whereas the volume part was found to be $K_s=(-5.0\pm0.6)\times10^6$ erg/cm³. The negative sign indicates that the surface normal is a magnetic hard axis.

We now show that both $K_p^{(2)}$ and K_s are caused by magnetoelastic interaction due to the elastic strain field originating from the lattice mismatch at the Co/Cu inter-

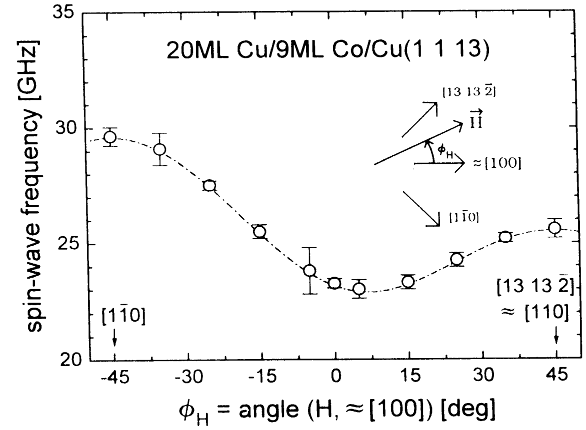

FIG. 2. Measured spin-wave frequencies of a 9-ML Co(1 1 13) film as a function of the in-plane direction of the applied external field measured by the angle ϕ_H with respect to the in-plane approximate [100] direction as defined by the inset. The applied field is 4 kOe. Relative maxima in the spin-wave frequencies indicate easy axes.

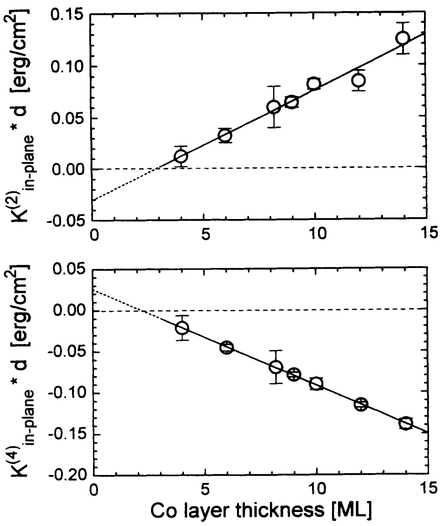

FIG. 3. Experimentally obtained values for the in-plane anisotropy constants of twofold ($K^{(2)}_{\text{in-plane}}$) and fourfold ($K^{(4)}_{\text{in-plane}}$) symmetry multiplied by the Co film thickness d as a function of d. The straight lines are least-squares fits to the data.

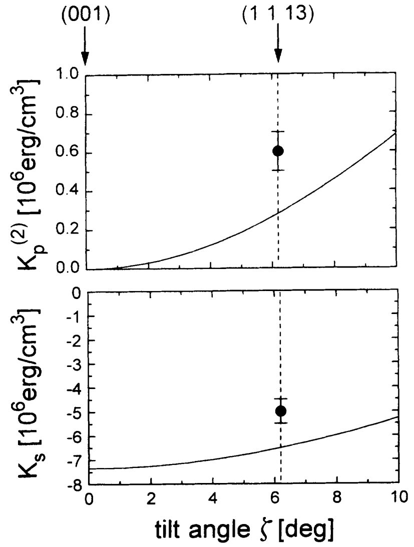

FIG. 4. Calculated volume in-plane ($K_p^{(2)}$) and out-of-plane (K_s) magnetoelastic anisotropy contributions (solid lines) as a function of the tilt angle ζ, by which the surface is rotated about the in-plane $[1\bar{1}0]$ axis with respect to the [001] orientation. The (1 1 13) orientation is marked by a dashed line. The experimental values for the (1 1 13) orientation are shown by full dots.

face. In a continuum approach we assume a smooth film, i.e., we neglect the stepped surface structure. Our approach is valid since the step distance of 6.5 atomic distances is much smaller than the static coherence length over which the magnetic moments might vary in direction. The free magnetoelastic energy F_{me} is given by[12]

$$F_{\text{me}}=\sum_{ijkl} b_{ijkl}\alpha_i\alpha_j\epsilon_{kl} \tag{3}$$

with b_{ijkl} the magnetoelastic tensor, α_i the direction cosines of the magnetization with respect to a carthesian coordinate system aligned along the [1 1 13], $[1\bar{1}0]$, and $[13\,13\,\bar{2}]$ axes (i.e., the approximate [001], $[1\bar{1}0]$, and [110] axes). ϵ_{kl} are the strain components. Please note that in the chosen coordinate system only the diagonal strain components ϵ_{kk} are nonzero. The two in-plane strain components ϵ_{11} and ϵ_{22} are obtained from the lattice mismatch at the interface, and the out-of-plane component ϵ_{33} is calculated by a standard minimization procedure of the elastic energy stored in the film.[12] The tensor components of the magnetoelastic tensor are obtained[12] from the magnetostriction constants λ_{100} and λ_{111} of fcc Co extrapolated from Co-rich CoPd alloys[13] as well as from the elastic constants of bulk fcc Co.[14] For evaluating Eq. (3) the magnetoelastic tensor must be rotated from its crystallographic reference frame into the film coordinate system (i.e., in our case by 6.2° about the $[1\bar{1}0]$ axis). The magnetoelastic anisotropy constant $K_p^{(2)}$ is obtained from the difference in the magnetoelastic free energy for **M** lying in-plane parallel and perpendicular to the steps, and K_s is obtained analogously from **M** lying in the directions of the film normal and the steps.[15]

Figure 4 shows the obtained results. The solid lines show the calculated magnetoelastic anisotropy constants K_s and $K_p^{(2)}$ plotted as a function of the tilt angle ζ, by which the surface is rotated about the $[1\bar{1}0]$ axis. For $\zeta=0$ the in-plane constant $K_p^{(2)}$ is zero for symmetry reasons. With increasing tilt angle $K_p^{(2)}$ increases. The experimental values of K_s and $K_p^{(2)}$ are shown as full dots for $\zeta=6.2°$. They agree with the calculation within a factor of 2, which is a rather good agreement taking into account the uncertainty in the estimates of the magnetostriction constants as well as the approximation of using bulk elastic constants for the thin films. We therefore conclude that both anisotropy contributions are most likely caused by magnetoelastic interaction.

A careful investigation of Eq. (3) shows that the magnitude and sign of $K_p^{(2)}$ depend very sensitively on a possible strain relaxation: A relaxation of the lattice parameter in the in-plane direction perpendicular to the steps, as recently proposed,[7] by 3% with no relaxation parallel to the steps would cancel $K_p^{(2)}$, and a further relaxation would reverse the sign. From the LEED data we estimate that this type of relaxation is not present in our films although a relaxation of 3% is at the limit of resolution. We would also like to emphasize that in addition to the magnetoelastic contribution to the volume in-plane anisotropies, surface contributions might exist due to the large density of aligned steps on the surface.[16] However, a quantitative analysis of this contribution is presently out of reach.

In conclusion we have determined all relevant anisotropies in (1 1 13)-oriented Co films. Due to the induced twofold symmetry a uniaxial in-plane anisotropy contribution is found in addition to the anisotropies found in (001)-oriented films. The origin of this anisotropy was found to be magnetoelastic. In addition the out-of-plane volume anisotropy contributions was found to be consistent with a magnetoelastic origin, although other mechanisms like a magnetocrystalline contribution might contribute as well.

The obtained results offer access to the origin of the stabilization of ferromagnetic order in (001)- and (1 1 13)-oriented films. As first discovered for (001)-oriented Co layers the stabilization mechanism is closely related to the in-plane anisotropy.[3] We find the same result both for the in-plane anisotropy contributions of twofold and fourfold symmetry in (1 1 13)-oriented Co films. Since the former could be identified as being of magnetoelastic origin the elastic strain fields are likely to provide the driving force for the stabilization of ferromagnetic order in ultrathin Co films. Unfortunately the origin of the in-plane anisotropy contribution of fourfold symmetry is inaccessible since higher-order magnetostriction constants of fcc Co are not available for a calculation of this contribution.

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