Magnetic Anisotropies of Ultrathin Co(001) Films on Cu(001)

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We present experimental evidence that ferromagnetic order at room temperature in ultrathin epitaxial Co(001) layers on Cu(001) substrates is stabilized by in-plane magnetic anisotropies. All relevant anisotropy contributions have been determined as a function of Co layer thickness with and without an additional Cu overlayer.

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The stabilization of long-range ferromagnetic order in two-dimensional systems at finite temperatures, which is experimentally found in epitaxial films of very few atomic layers, is presently discussed very controversially. According to the theorem of Mermin and Wagner [1] a ferromagnetic ground state cannot exist at finite temperatures in an isotropic two-dimensional Heisenberg system with short-range interactions. Several mechanisms have been proposed to account for the experimentally observed existence of ferromagnetic order in two-dimensional systems [2-6]. Among the mechanisms the stabilization of ferromagnetic order due to dipolar interactions or due to magnetic anisotropies is currently discussed most.

In this Letter we present room-temperature data of all relevant magnetic volume and surface anisotropy contributions in epitaxial (001)-oriented films of fcc Co on Cu(001) substrates. The data have been determined by means of Brillouin light scattering from spin-wave excitations. The spin-wave data together with measurements of the magneto-optic Kerr effect (MOKE) show that at room temperature ferromagnetic order exists for Co film thicknesses d larger than $d_c = 1.6 \pm 0.3$ monolayers (ML) in agreement with results of other research groups [7,8]. With decreasing Co film thickness we find that the disappearance of ferromagnetic order at d_c coincides with the in-plane magnetic anisotropy approaching zero. We therefore suggest that in Co(001) films ferromagnetic order at room temperature is stabilized by magnetic in-plane anisotropy. The stabilization mechanism is further corroborated by additional measurements on Co(001) films covered with a Cu overlayer. We note that the mechanism might be system dependent and thus not be applicable to other systems.

The samples were prepared in an ultrahigh-vacuum chamber of base pressure better than 5×10^{-11} mbar by e^--beam evaporation at a pressure of 2×10^{-10} mbar. The deposition rate was 1 ML = 1.77 Å per 6 min. Prior to film deposition the Cu substrate was cleaned by sputter and annealing cycles with annealing temperatures increasing up to 600 °C. The crystallographic quality of the substrate and of the films was monitored by LEED and the surface cleanliness was checked by Auger spectroscopy. The film thickness was monitored by a quartz microbalance calibrated by means of *ex situ* x-ray interference measurements. The error in absolute thickness is smaller than 5%. Special emphasis was placed on the preparation of smooth films with minimum interdiffusion at the Co/Cu interface: Films of 3 ML thickness were prepared at different deposition temperatures T_{dep} between 20 and 140 °C, yielding the best results at $T_{dep} = 85$ °C. For the latter deposition temperature the Auger-intensity ratio of the signals from the Co 656 and 716 eV lines to those of the Cu 849 and 920 eV lines has a maximum, indicating a uniform film with smooth interfaces and low interdiffusion. From the spin-wave spectra we find that the "magnetic quality" of the films is also best at $T_{dep} = 85$ °C. For this deposition temperature the magnetization is found to rotate as a function of field strength in the most uniform way from the easy [110] direction into the hard [100] direction along which the external field is applied (see below).

The Brillouin light scattering measurements of the spin-wave excitations were performed *in situ* in UHV at room temperature by focusing the $\lambda = 5145$ Å light of an Ar^+-ion laser of up to 120 mW intensity through a viewport onto the sample. The inelastically scattered light was collected in backscattering geometry through the same viewport. The frequency analysis was performed using a high-contrast (3+3)-pass tandem Fabry-Pérot interferometer [9]. The measurement time per spectrum was typically 30–120 min. The wave vector $q_\parallel$ of spin waves tested in the experiment was 1.73×10^5 cm^{-1}. In addition magnetic hysteresis loops were measured *in situ* in UHV by the transverse magneto-optic Kerr effect using a He-Ne laser.

Figure 1 shows the geometry used in the light scattering experiments on the Co(001) films. The external magnetic field of up to 1.8 kOe is applied in the plane along the magnetic hard [100] axis. The wave vector $q_\parallel$ tested in the scattering experiment lies in the [010] direction. The angles θ and ϕ denote the direction of the saturation magnetization with respect to the surface normal **n** and to the in-plane [100] direction, respectively.

Ferromagnetic order is observed for film thickness d larger than $d_c = 1.6 \pm 0.3$ ML for uncovered Co films and $d_c = 1.9 \pm 0.3$ ML for Cu-covered Co films. We identify the onset of ferromagnetic order by the existence of a remanent magnetization resulting in a coercive field,

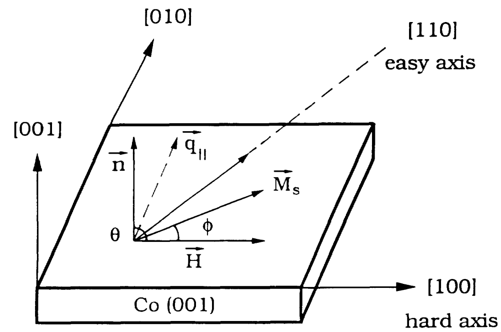

FIG. 1. Geometry for the Brillouin light scattering experiments. The external field H is applied in the magnetically hard [100] direction. The wave vector of the spin waves points into the [010] direction.

which is tested by MOKE, as well as by the existence of spin waves at zero applied fields [10]. The obtained coercive field is 81 ± 14 Oe for the uncovered film and 66 ± 11 Oe for the covered film. The obtained values are larger than values published by de Miguel *et al.* [11], since we measured the coercive field for external fields along the magnetic hard [100] axis.

For ultrathin fcc Co(001) films of fourfold symmetry about the surface normal, the free energy contribution, E_{ani}, due to anisotropies is expressed in lowest nonvanishing order as [5]

$$E_{\text{ani}} = \frac{1}{4}\left(K_p + \frac{2}{d}k_p\right)\sin^2 2\phi \sin^4\theta + \frac{2}{d}k_s \sin^2\theta , \qquad (1)$$

with K_p and k_p the in-plane volume and surface anisotropy constants of fourfold symmetry, respectively, and k_s the out-of-plane surface anisotropy constant. Here k_p and k_s are averaged over both surfaces. From the data analysis described below we did not find any evidence of further anisotropy contributions as might arise from a volume out-of-plane anisotropy K_s, or from a uniaxial in-plane contribution.

In thin ferromagnetic films there exists at room temperature the thermally activated so-called Damon-Eshbach spin-wave excitation. Its energy is primarily determined by the dipolar spin-spin interaction, since the wavelength of the order of 3000 Å is large compared to interatomic distances. If the product of the film thickness d and the wave vector $q_\parallel$ is small compared to unity, the frequency ω of the mode can be expressed as [12]

$$\left(\frac{\omega}{\gamma}\right)^2 = \left(H_0 + \frac{2A}{M_s}q_\parallel^2 + H_\alpha(\phi) + 4\pi M_s f(1 - \tfrac{1}{2}q_\parallel d)\right)\left(H_0 + \frac{2A}{M_s}q_\parallel^2 + H_\beta(\phi) + 2\pi M_s f q_\parallel d \sin^2(\phi - \phi_q)\right), \qquad (2)$$

with γ the gyromagnetic ratio, H_0 the external applied field, A the exchange constant, M_s the saturation magnetization, and ϕ_q the angle of $\mathbf{q}_\parallel$ with the [100] direction. The parameter f is the demagnetization factor of ultrathin films, which for $n > 1$ is approximately $f = 1 - 0.2338/n$ with n the number of monolayers [13]. The anisotropy field components $H_\alpha(\phi)$ and $H_\beta(\phi)$ are obtained from the free energy E_{ani} as defined in Eq. (1) [14]. From Eq. (2) it follows immediately that the spin-wave frequency depends (i) on the angle $\phi - \phi_q$ between the direction of magnetization and the wave vector (determined by the scattering geometry), and (ii) on the angle ϕ between the direction of magnetization and the crystallographic [100] axis due to the anisotropy terms. By applying the external magnetic field along the magnetic hard [100] axis, one can probe the magnetic anisotropies by studying the rotation of the magnetization with increasing field into the direction of the applied field via the corresponding change in the spin-wave frequency: Upon increasing the applied field the spin-wave frequency first decreases due to the change in ϕ, until a critical field strength H_{crit} is reached. For $H > H_{\text{crit}}$ the magnetization and the applied field are collinear and the spin-wave frequency increases nearly linearly with further increasing field. H_{crit} is a measure of the in-plane anisotropy.

In Fig. 2 sample spectra measured at an applied field of 1 kOe are shown. The (a) upper and (b) middle parts show spin-wave spectra of a 2-ML- and a 4-ML-thick Co

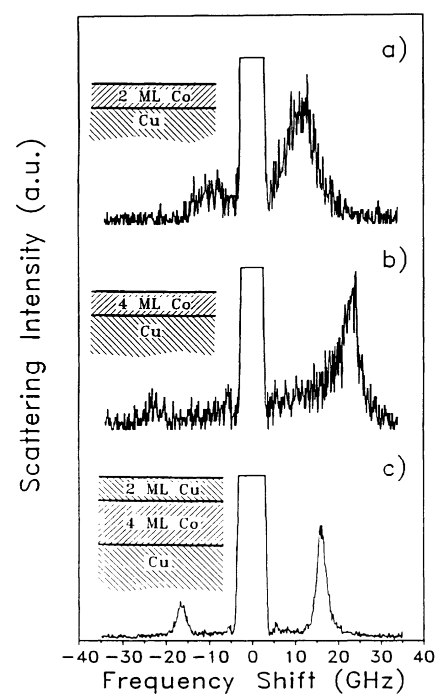

FIG. 2. Brillouin spectra at 300 K for (a) 2 ML Co on Cu(001), uncovered; (b) 4 ML Co on Cu(001), uncovered; and (c) 4 ML Co on Cu(001), covered with 2 ML Cu. The applied external field is 1 kOe.

film, respectively. The Damon-Eshbach mode is clearly seen near (a) 11 GHz and (b) 23 GHz. The large change in its frequency for these two samples already indicates a large contribution from surface anisotropies. The lower spectrum (c) of Fig. 2 is obtained from a 4-ML-thick Co film covered with 2 ML Cu. Here the mode frequency is reduced by 6 GHz compared to the corresponding uncovered film [middle spectrum (b)] indicating different surface anisotropy contributions of the Co/Cu and Co/vacuum interfaces.

From a careful least-squares fit of Eq. (2) to the measured spin-wave frequencies as a function of the applied field and the Co film thickness the saturation magnetization as well as the anisotropy constants K_p, k_p, and k_s were determined. A careful analysis of the validity of the fit results, including an error and correlation analysis of the fit parameters, was performed. For all studied films with $d > d_c$ the saturation magnetization does not deviate from the bulk value of 17.9 kG [15] within the limit of accuracy of 5%. This result is corroborated by finding a linear increase of the Kerr rotation angle with increasing film thickness measured *in situ* on the same films as used for the light scattering studies.

Figure 3 shows the obtained anisotropy values as a function of film thickness for uncovered Co films (open symbols) and Co films covered with 2 ML Cu (solid symbols). In the upper part the total in-plane anisotropy $K_{\text{in-plane}} = K_p + (2/d)k_p$ is shown; the middle part displays the out-of-plane surface anisotropy constant k_s. Also shown, in the lower part, is the coercive field measured *in situ* in UHV on the same samples with MOKE.

The out-of-plane surface anisotropy constant k_s is constant for $d > d_c$ within experimental error indicating no volume out-of-plane anisotropy contribution. The average value of -0.46 ± 0.09 erg/cm^2 for uncovered Co films changes to 0.15 ± 0.04 erg/cm^2 upon covering the Co films by 2 ML Cu. The negative sign indicates that the surface normal is a magnetic hard axis.

Of particular interest are the properties of $K_{\text{in-plane}}$. A separation into the volume (K_p) and the surface (k_p) contribution is possible for different thicknesses d and yields $K_p = (-2.3 \pm 0.15) \times 10^6$ ergs/cm^3 and $k_p = 0.034 \pm 0.004$ erg/cm^2 for the uncovered films, and $K_p = (-2.2 \pm 0.15) \times 10^6$ ergs/cm^3 and $k_p = 0.031 \pm 0.003$ erg/cm^2 for the Co layers covered with 2 ML Cu. Because of their opposite signs, the contributions of K_p and k_p to $K_{\text{in-plane}}$ cancel each other at $d_c^* = 1.7 \pm 0.3$ ML for the uncovered films and at $d_c^* = 1.6 \pm 0.3$ ML for the Cu-covered films. We would like to point out that $d_c^* = d_c$ within the experimental error for both covered and uncovered Co films, although both quantities are determined by independent experiments.

Our anisotropy values of $K_{\text{in-plane}}$ agree with data previously obtained by Heinrich *et al.* [13] for Au/Cu/Co/Cu(001) using ferromagnetic resonance. Our value of k_s is consistent with their data. An analysis of their data,

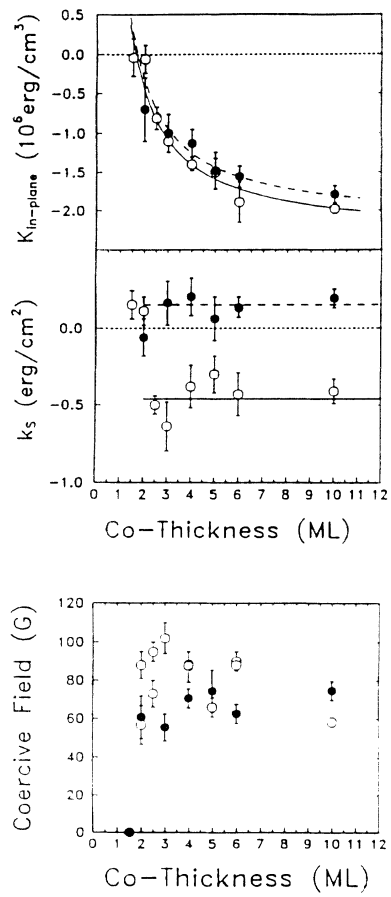

FIG. 3. Obtained in-plane (upper part) and out-of-plane (middle part) anisotropy constants as a function of the Co layer thickness (in ML) for Co/Cu(001) without (open circles) and with (solid circles) a 2-ML-Cu overlayer. A fit of the theory to the data is shown for Co/Cu(001) (solid line) and for Cu/Co/Cu(001) (dashed line). In the lower part the values of the coercive field measured with MOKE are shown for Co/Cu without (open circles) and with (solid circles) a Cu overlayer. Here the external field is applied in the hard [100] direction.

although in number rather limited, indicates a nonzero out-of-plane volume anisotropy (K_s) contribution possibly caused by their different sample preparation conditions and cover layers, in contrast to our results.

Comparing the data of the uncovered Co films with the films covered by 2 ML Cu (symmetric interfaces), the surface anisotropy constants can be separated into the contributions of each surface or interface of the Co film. Since $K_{\text{in-plane}}$ does not change upon covering the Co layer by Cu, both the Co/Cu and Co/vacuum interface have the same value of $k_p = 0.032 \pm 0.003$ erg/cm^2. The out-of-plane anisotropy constant, k_s, was found to be $k_s = -1.06 \pm 0.17$ erg/cm^2 for the Co/vacuum interface and $k_s = 0.15 \pm 0.04$ erg/cm^2 for the Co/Cu interface.

From the observed agreement between the critical thickness for ferromagnetic order, d_c, with the thickness

d_c^* at which the contributions to the in-plane anisotropy cancel, we conclude that the symmetry breaking interaction for stabilizing ferromagnetic order in Co(001) films at room temperature is indeed given by the magnetic in-plane anisotropy contribution. We would like to point out that this argument is backed by the fact that d_c^* may also be obtained from an extrapolation of $K_{\text{in-plane}}(d)$ from data with film thicknesses significantly larger than d_c, thus ruling out structural and/or magnetic percolation effects near d_c^*. The out-of-plane anisotropy, described by the thickness-independent k_s, does not support the stabilization: Our data clearly indicate that in contrast to the critical thickness d_c, and to the in-plane anisotropy constant $K_{\text{in-plane}}$, k_s is very sensitive to the presence of a Cu overlayer.

The origin of k_s and $K_{\text{in-plane}}$, in particular their different sensitivities to the presence of a Cu overlayer, is presently not well understood. The thickness-independent part of $K_{\text{in-plane}}$, described by K_p, can likely be identified as being magnetocrystalline in origin due to its sign and magnitude. However, in order to compare K_p to an anisotropy of cubic symmetry appropriate for (100)-oriented Co films of larger thickness, a thickness-independent uniaxial perpendicular anisotropy contribution of appropriate size must be added in Eq. (1) in order to make the volume anisotropy invariant against cubic symmetry transformations. We do not find any evidence for such a contribution from our data for the investigated film thickness regime of $1\ \text{ML} \le d \le 10\ \text{ML}$.

It might be speculated that the values of k_s and $K_{\text{in-plane}}$ are due to the elastic strain fields parallel to the film induced by the lattice mismatch of 1.9% at the Co/Cu interface. Hence the values of k_s and $K_{\text{in-plane}}$ would be proportional to the interface strain. An estimate for the out-of-plane magnetoelastic anisotropy yields values of the same order of magnitude, in particular the same sign as measured for k_s. For this estimate we used fcc Co magnetostriction constants extrapolated for Co rich fcc Pd-Co alloys [16]. However, since the Co strains are not expected to largely change upon coverage with a Cu overlayer our finding of a very sensitive dependence of k_s on a Cu overlayer is in contradiction to the assumption of magnetoelastic contributions to k_s. In order to estimate a magnetoelastic contribution to $K_{\text{in-plane}}$, higher-order magnetostriction constants would have to be included which are not available and which would need to be very large, which is unlikely. On the other hand, k_s may be related to the spacing between the surface Co layer and the underlying atomic Co layer which is known to be very sensitive to the Cu overlayer. Detailed total-energy calculations are highly desirable to clarify the origin of these anisotropies in this two-dimensional model system.

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