Morphology-Induced Oscillations of the Magnetic Anisotropy in Ultrathin Co Films

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The magnetic anisotropy in Co films epitaxially grown on Cu(100) is found to oscillate as a function of the Co thickness. The oscillation period corresponds to one monolayer, as revealed by measuring magneto-optical Kerr hysteresis loops during film growth. These oscillations are attributed to the periodic variations of the film morphology alternating between filled and incompletely filled atomic layers.

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Magnetic anisotropies of ultrathin films are inherently connected to the structure and morphology of the films. This has been shown, for example, for Ni/Cu(100) [1] and Co/Cu(110) [2]. In these systems strong relaxations of the lattice constants upon growth are found, giving rise to drastically altered magnetic anisotropies. In principle the change of the morphology when going from a filled to an incompletely filled layer could also cause the magnetic anisotropy to change. Thus, provided the film grows layer by layer, one might expect that the variations of the film roughness could lead to oscillations of the magnetic anisotropy with a period of one monolayer—quite analogously to the intensity variations in a reflection high-energy electron diffraction (RHEED) experiment.

In this paper we present direct experimental evidence that the magnetic anisotropy in Co films grown on a Cu(100) single crystal oscillates as a function of the film thickness. In order to obtain easy access to the magnetic anisotropy of our films, the Cu crystal was slightly miscut by 0.1°. We have used the magneto-optical Kerr effect to measure magnetic hysteresis loops along an axis not coinciding with the easy magnetization direction. From an analysis of these loops we find oscillations in the uniaxial magnetic anisotropy with a period of 1 monolayer (1 ML = 0.18 nm). We relate this behavior to the anisotropy at the step edges [3].

Our findings on the stepped Cu crystal are of general importance for macroscopically flat surfaces: Any "flat" crystal surface considered to be fourfold symmetric consists of a random arrangement of steps, breaking the macroscopic fourfold symmetry locally to a uniaxial symmetry.

The Co films were evaporated at room temperature in an ultrahigh vacuum system by molecular beam epitaxy onto a stepped Cu(100) single crystal at an evaporation rate of 0.05 ML/min. The Cu crystal has a preferential step direction along [110] with a mean distance of $\approx$100 nm between adjacent steps. Prior to film deposition, the Cu substrate was cleaned by sputtering and annealing up to 800 K. The film thickness was determined by Auger electron spectroscopy and calibrated with a stylus profilometer on thick films. The error of absolute thickness is smaller than 10%.

The magnetic hysteresis loops were recorded *in situ* during the growth of the Co film using the magneto-optical Kerr effect. For these experiments the Co evaporator was at an angle of 70° to the surface normal in the $(1\bar{1}0)$ azimuth of the Cu crystal.

Figure 1 shows typical hysteresis loops of a Co film having a thickness of 2.3 ML, the magnetic field being applied either parallel (along $[1\bar{1}0]$) or perpendicular (along [110]) to the step edges of the Cu substrate. The easy magnetization axis runs parallel to the step edges along the $[1\bar{1}0]$ direction, as shown by the rectangular hysteresis loop shape. The loop taken along the [110] direction is more complicated in appearance and comprises two shifted single loops. This difference of the magnetic response in the $[1\bar{1}0]$ and [110] directions, which are magnetically equivalent on a perfect fourfold Co(100) film, is due to the presence of a uniaxial anisotropy [3–5]. Whereas the $[1\bar{1}0]$ direction is the easy magnetization axis, the [110] direction is the intermediate axis because it combines the easy character of the fourfold cubic anisotropy with the hard character of the uniaxial anisotropy.

It is the observation of these composite loops along the intermediate axis that made our experiment possible. We

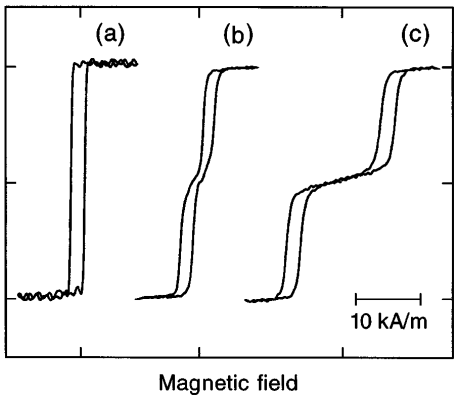

FIG. 1. Hysteresis loops $M(H)$ obtained using the magneto-optical Kerr effect on a 2.3-ML Co film. (a) H along the $[1\bar{1}0]$ direction, (b) H along the [110] direction, and (c) same as (b) but having a bias field of $H_{\text{bias}} = 5$ kA/m along the $[1\bar{1}0]$ direction.

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define the shift field H_s as the field difference between zero field and the center of the single shifted loops. The value of H_s can be determined with high accuracy and is directly proportional to the uniaxial magnetic anisotropy, as shown below.

The in-plane free energy of a Co film on a stepped Cu(100) surface having the external magnetic field H applied along the [110] direction is described by $K_u \sin^2(\phi) + (K_1/4)\sin^2(2\phi) - HM_s \sin(\phi)$, where K_u is the uniaxial anisotropy constant, K_1 the cubic anisotropy constant, M_s the saturation magnetization, and ϕ the angle between the magnetization and the $[1\bar{1}0]$ direction [6]. As the easy axis is along $[1\bar{1}0]$, both K_u and K_1 are positive. Minimizing the energy with respect to ϕ and assuming that $K_u \ll K_1$, one finds that the uniaxial and the cubic anisotropies are directly given by the shift field H_s and the linear initial slope s of the loop, respectively: $K_u = H_s M_s$ and $K_1 = M_s^2/2s$. The assumption that K_u is small compared to K_1 is justified because the miscut of the Cu crystal—which induces the uniaxial anisotropy—is extremely small. Moreover, the hysteresis loops confirm the validity of the approximation because $H_s \approx 1$ kA/m is much smaller than $M_s/2s \approx 50$ kA/m for $d > 2$ ML.

In our films the slope s cannot be determined directly from hysteresis loops such as the one presented in Fig. 1(b) because the width of the single loops is comparable to H_s. Therefore no linear region between the two shifted loops can be identified. We can realize loops with an extended linear slope between the shifted loops by applying a constant bias field H_{bias} along the easy axis while sweeping the loop along the intermediate axis. The bias field introduces an additional uniaxial anisotropy, which results in a shift field of the two loops increased by H_{bias}. From the wide hysteresis-free field region between the loops we can now determine the initial slope s; see Fig. 1(c).

Figure 2 presents the analysis of transverse Kerr hysteresis loops taken during film growth. The saturation magnetization M_s and the shift field H_s are shown as a function of the Co thickness d. Moreover, the half-width of the single shifted loop is shown. Since it corresponds to the coercive field of the easy axis loop, we denote it as H_c. At $d \approx 1.5$ ML the onset of ferromagnetism at room temperature is observed. For larger coverages a monotonous increase of M_s is found. The $H_s(d)$ curve, on the other hand, is nonmonotonous. We identify oscillations having a period of 1 ML and an amplitude that decreases with increasing Co coverage. The H_c data also reveal oscillations, though they are much weaker in amplitude than the H_s oscillations. Except for the first two H_c minima, which coincide with H_s maxima, all further minima of H_c and H_s match. We note that a change of slope in H_c at $d \approx 2$ ML has been measured previously [7]. Moreover, H_c oscillations with a period of 1 ML have already been observed in Fe/Pd(100) superlattices with varying Pd thickness [8] and have been attributed to a possible roughness modulation during the growth of successive Pd layers. But, because the coercive field is determined by several physical mechanisms such as domain wall nucleation and pinning, its relation to the magnetic anisotropy is not straightforward. Now, taking the evidence of anisotropy oscillations, the tiny oscillations in H_c can be understood as an indirect consequence of the anisotropy changes. Obviously, the latter are very weakly reflected in H_c. However, one has to be cautious when evaluating only H_c: The phase jump between the second and the third H_c minima remains unexplained, but must be taken as an indication that other parameters determine H_c at small Co thickness.

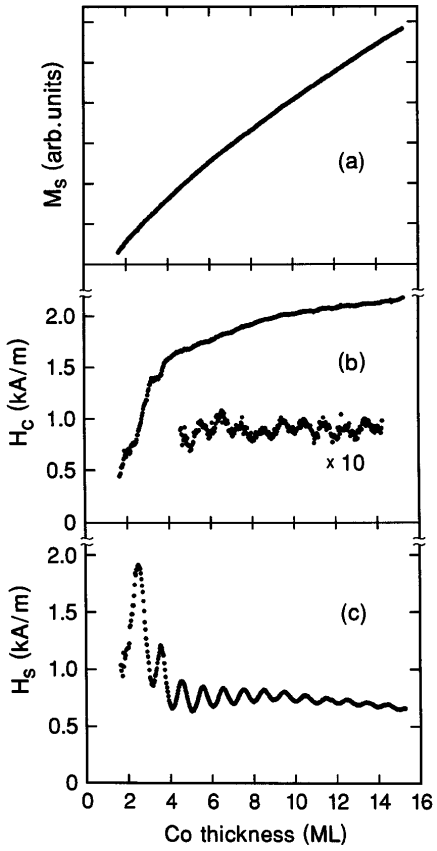

FIG. 2. (a) Saturation magnetization M_s, (b) half-width of the single shifted loops H_c, and (c) shift field H_s as a function of the Co thickness. The inset in the middle panel shows H_c after subtraction of a smooth background.

To compare the uniaxial to the fourfold anisotropy directly, we perform a growth experiment with a bias field H_{bias} applied. Figure 3 shows the result for $H_{\text{bias}} = 5$ kA/m. Again H_s oscillations with a 1-ML period are observed. The overall shape of H_s deviates from that in Fig. 2, which may indicate that the application of a bias field influences the uniaxial anisotropy. The cubic anisotropy is more difficult to determine experimentally because of the relatively large uncertainty of fitting the initial slope of the loops. From Fig. 3 we can exclude oscillations having an amplitude larger than 5% of the signal at large Co coverages. Thus oscillations having a relative amplitude comparable to the H_s oscillations are

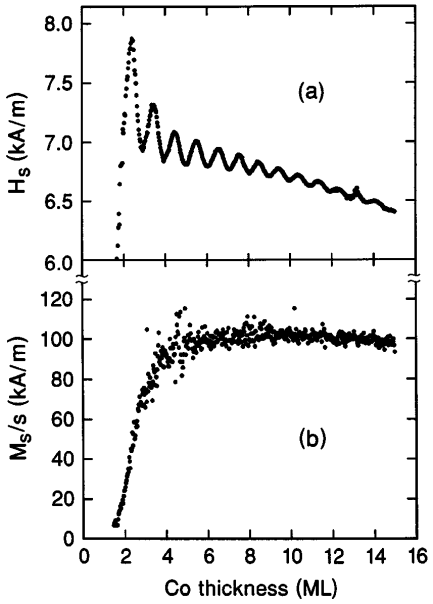

FIG. 3. (a) Shift field H_s and (b) inverse linear initial slope M_s/s as a function of the Co coverage; a bias field of $H_{\text{bias}} = 5$ kA/m is applied along the easy direction. The bias field has not been subtracted from the H_s curve.

not present in M_s/s nor, hence, in K_1. From additional experiments to determine K_1 more precisely, we can set an upper limit for the oscillation amplitude in the entire thickness range investigated, $\Delta K_1 < 1 \times 10^3$ J/m^3.

What is the mechanism that causes the oscillations in the uniaxial anisotropy? The observation of a 1-ML period indicates a structural origin of the oscillations. In principle, quantum well states could also be responsible for anisotropy oscillations [9] because they periodically change the electronic band structure of the Co layer [10]. However, a period induced by quantum well states is very unlikely to coincide accidentally with the lattice constant. Moreover, in the case of a period slightly different from 1 ML, one would expect to observe a phase slip at some Co thickness to accommodate the discreteness of the lattice, which is not observed in our experiment up to 15 ML. In order to ensure that the oscillations are not induced by the off-normal-incidence growth, we repeated the experiment by evaporating Co in normal incidence. Again, the H_s oscillations are present at the same positions as in oblique incidence and have comparable amplitudes.

Co on Cu(100) grows layer by layer except for the first 2 ML [11]. As soon as a layer is completed, islands form in the next layer until they coalesce to form a complete layer again. This means that the film morphology periodically changes from flat to "rough," corresponding to a complete and incomplete top layer, respectively. The film roughness therefore oscillates with a period of 1 ML [12].

On an atomic scale, evaporation in normal incidence onto a perfectly oriented Cu(100) surface yields, on average, Co patches with fourfold symmetry. The presence of steps, however, locally breaks the symmetry. This can result in rectangular rather than square Co islands owing to an anisotropic step-edge diffusion of the Co atoms [13]. Symmetry breaking could also directly influence the magnetic anisotropy at the step edges which determine the observed macroscopic uniaxial anisotropy [3]. The excess length of one step direction compared to the orthogonal direction determines the anisotropy in the first case, the difference of local step anisotropies in the latter. These quantities may be nonvanishing on our slightly miscut substrate, and thus may vary during growth of the film. As soon as the layer is completed, it is minimized, resulting in an oscillatory variation of the uniaxial anisotropy.

In the following we will present a phenomenological analysis of these growth asymmetries and estimate their influence on the magnetic anisotropy. The model must be able to explain the observed oscillations in the uniaxial anisotropy, and the absence of oscillations in the fourfold anisotropy.

The half-filled top Co layer is approximated by rectangular islands consisting of $N_\parallel \times N_\perp$ atoms distributed over the entire surface. A typical value for $N_\parallel$ is deduced from scanning tunneling microscopy (STM) images at half-filled layers [11], $N_\parallel \approx 30$. Relating our observations to the presence of a strong anisotropy at the step atoms [3,14], we can easily calculate their influence on the uniaxial anisotropy. The additional anisotropy contribution from the edge atoms of Co islands compared to a completely filled layer is equal to the oscillation amplitude ΔK_u:

$$\Delta K_u = (2/N_\parallel N_\perp \sqrt{2}\, ad)\,(N_\parallel K_\parallel^{\text{step}} - N_\perp K_\perp^{\text{step}})\,.$$

The positive constants $K_\parallel^{\text{step}}$ and $K_\perp^{\text{step}}$ are the step anisotropies of the atoms at Co island edges parallel and perpendicular to the preferential Cu step direction, respectively, in units of energy per length. The lattice constant a and the Co film thickness d normalize ΔK_u to an energy density. From the observed overall behavior of the uniaxial anisotropy we can estimate the step anisotropy. It is given by the surface contribution K_u^s of the uniaxial anisotropy K_u [15] multiplied by the average step distance, $K_\parallel^{\text{step}} = K_u^s a/\tan(\alpha)$, where α is the miscut angle of 0.1°. Analyzing the $H_s(d)$ curves in terms of volume and surface contribution yields $K_u^s \approx 5 \times 10^{-7}$ J/m^2.

Quite generally, ΔK_u is nonvanishing if (i) the islands are rectangular, or (ii) the islands are square, but the steps parallel and perpendicular to the substrate step direction are magnetically unequal. We will briefly discuss these two limiting cases.

(i) Rectangular islands, $N_\parallel > N_\perp$. In the simplest case the step anisotropies along the two orthogonal directions are equal. With the above estimates for $N_\parallel$ and K_u^s we then arrive at an island shape described by an aspect ratio of $N_\parallel/N_\perp \approx 1.01$. Thus an overall small deviation of 1% from a perfectly fourfold symmetric growth morphology

is sufficient to explain the observed oscillation amplitude in our experiment.

(ii) Square islands, $N_{\parallel} = N_{\perp}$. The observed oscillations are induced by slightly different step anisotropies, $K_{\parallel}^{\mathrm{step}} > K_{\perp}^{\mathrm{step}}$. Analogous to (i) one finds that $K_{\parallel}^{\mathrm{step}}/K_{\perp}^{\mathrm{step}} \approx 1.01$ is sufficient to explain the observed oscillation of the uniaxial anisotropy.

From these rough estimates we conclude that both limiting cases are possible origins of the anisotropy oscillations. In (i), the physical origin leading to rectangular rather than square islands is anisotropic step edge diffusion [13] caused by the presence of the slightly miscut substrate. In (ii), the difference in step anisotropies could be caused by a magnetoelastic anisotropy contribution [16] brought about by a slight distortion of the lattice at steps.

We note that, in principle, the magnetostatic energy could also show an oscillatory behavior. If $N_{\parallel} > N_{\perp}$, the magnetostatic energy is anisotropic because of the elongated shape, and will therefore contribute to the uniaxial anisotropy. However, direct evaluation of the classical formulas given by Stoner [17] for an ellipsoidal shape then requires a ratio of $N_{\parallel}/N_{\perp} \approx 1.3$ to account for the observed oscillation amplitude. Such a strong deviation from isotropic growth, however, is not supported by STM experiments on Cu substrates with different miscuts [11,18]. Shape anisotropy is therefore ruled out as a mechanism for the observed oscillations.

Recent experiments obtained on Co/Cu(100) films by means of RHEED [12] might allow microscopic insight into the origin of the magnetic step anisotropy. These experiments showed that at the island edges of the incompletely filled top layer the in-plane atomic spacing is different from that of completed layers. This results in an oscillatory variation of the average surface in-plane lattice spacing with film thickness. One can expect this change in atomic spacing at the island edges to produce a magnetoelastic anisotropy, i.e., in the phenomenological model discussed above, a contribution to the step anisotropy K^{step}. To account for case (ii) one would require an anisotropic relaxation of the lattice spacing induced by the presence of steps in the substrate. An experiment to determine anisotropic lattice relaxations similar to the one of Ref. [12] has yet to be performed.

Both these limiting cases qualitatively describe the oscillations in the uniaxial anisotropy. Let us take a look at the fourfold anisotropy contribution. In principle, oscillations in the fourfold anisotropy can be expected in both cases. However, contributions to the fourfold anisotropy come only from the corner regions of the islands for symmetry reasons, whereas all other edge sites can only contribute to the uniaxial anisotropy [19]. Taking this into account, a rough estimate of the expected changes of the fourfold anisotropy reveals that they are too small to be detected in our experiment because they are suppressed by a factor proportional to $1/N_{\parallel}$ compared to the uniaxial anisotropy.

In conclusion, oscillations of the uniaxial magnetic anisotropy having a period of 1 ML have been observed in Co films on a stepped Cu(100) surface by means of the magneto-optical Kerr effect. These oscillations are related to the structure and morphology of the Co films. Opposite limits of the likely mechanism have been discussed: an oscillatory variation of the film roughness characteristic for layer-by-layer growth combined with anisotropic growth of the Co islands or, alternatively, with an anisotropic relaxation of the Co in-plane lattice constant. Both of these structural mechanisms result in magnetic anisotropy oscillations via step anisotropies. Further highly precise structural experiments will be necessary to observe directly the anisotropic growth behavior or the tiny distortions of the atomic arrangements at step edges.

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