Influence of the deposition angle on the magnetic anisotropy in thin Co films on Cu(001)

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Off-normal molecular beam epitaxy of Co on Cu(001) has been found to result in a uniaxial magnetic anisotropy. The easy magnetization axis is perpendicular to the plane of incidence for deposition angles between 10° and 80°. The uniaxial magnetic anisotropy is related to the formation of a uniaxial surface morphology during off-normal growth. Spot profile analysis low-energy electron diffraction measurements reveal the presence of elongated adatom structures in contrast to square ones growing at normal incidence. The long sides of the adatom structures are oriented perpendicular to the plane of incidence, i.e., parallel to the easy magnetization axis. The formation of elongated adatom structures is due to steering. Steering originates from long-range attractive forces between incident atoms and substrate atoms and leads to preferential arrival of atoms on top of adatom structures.

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I. INTRODUCTION

Magnetic anisotropies in ultrathin films are inherently related to the structure and morphology of the films. In thin films of fourfold symmetry, such as Co on Cu(001), the cubic in-plane magnetic anisotropy is a natural consequence of the crystalline symmetry. An additional uniaxial in-plane magnetic anisotropy has been measured in films grown on stepped surfaces.[1–3] The micromagnetic origin of this uniaxial anisotropy is currently believed to arise from missing bonds (Néel-type anisotropy)[3–6] and/or strain (magnetoelastic anisotropy)[4,7,8] at surface steps. Since a long time it is known that off-normal deposition also induces an uniaxial in-plane magnetic anisotropy.[9–11] This anisotropy is connected to an uniaxial surface morphology which develops during off-normal growth. Growth-induced uniaxial magnetic anisotropies in Co films (150–1000 Å) have been studied as a function of the deposition angle recently.[12] Furthermore, an uniaxial easy axis perpendicular to the deposition direction has been measured in ultrathin magnetic films (20 Å).[13]

The purpose of our study was to investigate the influence of the deposition angle on the magnetic anisotropy in ultrathin magnetic films grown by molecular beam epitaxy (MBE). Particularly, the investigations were focused on a few monolayer thick Co films on Cu(001) grown with incident angles up to 80° with respect to the surface normal. Off-normal Co deposition results in an in-plane uniaxial magnetic anisotropy, with the easy axis oriented perpendicular to the plane of incidence. The uniaxial anisotropy in ultrathin Co films is directly related to the formation of elongated adatom structures during growth. The evolution of an uniaxial surface morphology is rationalized by a phenomenon named steering,[14] i.e., the focussing of incident atom flux on top of growing adatom structures. Steering and as a result the uniaxial magnetic anisotropy increase with increasing deposition angle.

II. EXPERIMENTAL

Thin Co films were grown on Cu(001) under ultrahigh vacuum (UHV) conditions (base pressure $<1\times10^{-10}$ mbar) and studied by spot profile analysis low energy electron diffraction (SPA-LEED), Auger spectroscopy (AES) and the magneto-optic Kerr effect (MOKE). The *ex situ* desulfurized Cu substrate was further prepared in UHV by cycles of sputtering with 800 eV Ar^+ ions and prolonged heating at about 800 K. This preparation method resulted in a clean Cu substrate with an average terrace width of 1000 Å .

The Co was deposited by electron beam induced sublimation from a Co wire at various angles of incidence. The growth rate, calibrated by measurements of He diffraction intensity oscillations before growth of the Co film and checked by AES after MOKE measurements, was about 0.1 monolayer (ML) per minute. The azimuthal direction of deposition was along the close packed [110] direction, which is a preferential step edge direction and an easy magnetization direction in Co/Cu(001). During most growth experiments, the temperature of the Cu(001) substrate was kept at 250 K and the pressure in the vacuum chamber did never exceed 2×10^{-10} mbar. Immediately after deposition the temperature of the Cu substrate was quenched rapidly in order to suppress undesired diffusion. The MOKE and SPA-LEED measurements on the as grown Co film were performed at 175 and 100 K, respectively.

For the MOKE measurements the sample was transferred into a small protrusion of the main vacuum chamber. Outside the vacuum chamber four current driven coreless coils were used to create magnetic fields perpendicular (polar geometry) and parallel (longitudinal geometry) to the surface plane. In both Kerr geometries, magnetic fields up to 400 Oe could be applied. The sample was illuminated at 45° by polarized HeNe-laser light. The Kerr ellipticity was measured by placing a quarter wave plate and a second polarizer in front of a photodiode. Azimuthal rotation of the sample made it possible to apply magnetic fields at angles varying from −45° to 35° with respect to the deposition direction.

III. RESULTS AND DISCUSSION

A. Magnetic anisotropy

As a first result we demonstrate that Co films grown with an off-normal angle of incidence exhibit a large in-plane

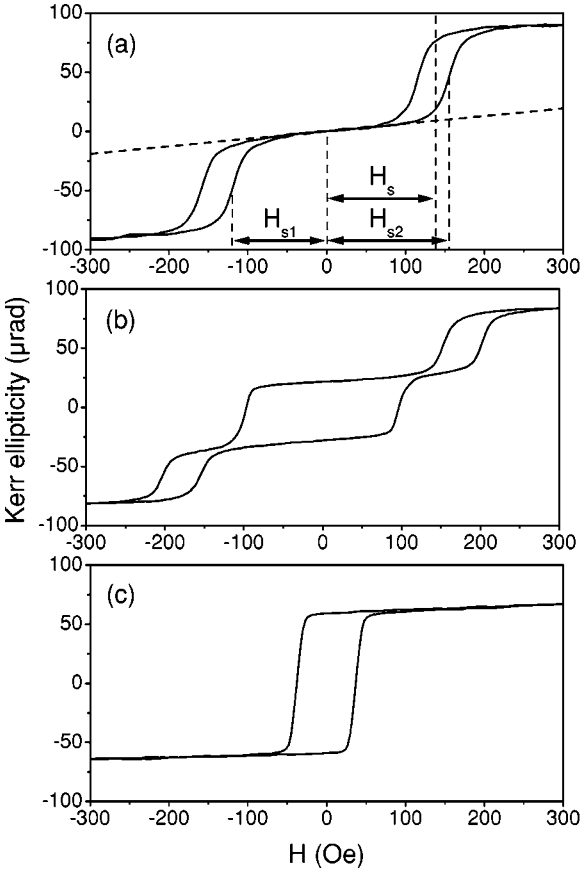

FIG. 1. In-plane hysteresis curves for a 4 ML thick Co film on Cu(001), grown at 250 K with an angle of 80° with respect to the surface normal. The magnetization curves were obtained at 175 K. The applied magnetic field is 0°, 20°, and 45° away from [110], i.e., away from the deposition direction, in (a), (b), and (c) respectively.

uniaxial magnetic anisotropy. Magnetization loops were measured at 175 K with the external magnetic field applied along different azimuthal directions. Figure 1 shows the result for a 4 ML thick Co film, grown with a deposition angle of 80° with respect to the surface normal. Parallel to the deposition direction a hard axis magnetization curve with two loops at large external field is measured [Fig. 1(a)]. At the fields where the loops appear the magnetization switches from the uniaxial easy direction (perpendicular to the deposition direction) into the uniaxial hard direction (parallel to the deposition direction) or vice versa. Around zero field a linear behavior is found, indicating coherent magnetization rotation away from the uniaxial easy direction. Comparable hard axis hysteresis curves have been obtained for Co films grown on stepped Cu(001).[5–7,16–19] When the external field is applied at an angle of 45° with respect to the deposition direction, i.e., along a cubic hard magnetization axis and in between the uniaxial hard and easy magnetization axes, the hysteresis loop exhibits a high squareness with an extremely sharp switching behavior [Fig. 1(c)]. The measured curve is, however, not saturated indicating magnetization reversal along the uniaxial easy axis. The small slope beyond the switching fields shows that a large magnetic anisotropy hinders magnetization rotation in the direction parallel to the applied field. This magnetic anisotropy consists mainly of a component with fourfold symmetry, i.e., the cubic magnetic anisotropy in off–normal deposited Co films is still larger than the uniaxial magnetic anisotropy (see below).

Hysteresis loops with three irreversible transitions are measured on off-normal deposited Co films when the external field is applied at an angle between 10° and 30° from the deposition direction [see Fig. 1(b)]. The remarkable hysteresis loop reveals that the magnetization reversal process is mediated by the nucleation and propagation of domain walls and not by coherent rotation. In systems with a cubic anisotropy and an uniaxial anisotropy that is aligned along one of the cubic easy axes, one and two jump hysteresis curves can be measured when magnetization reversal proceeds via coherent rotation. The observed switching behavior in these systems depends on the direction of the applied field and the ratio of the uniaxial to cubic anisotropy constants. When magnetic switching proceeds via the nucleation and propagation of domain walls, one, two, and three jump hysteresis curves can be obtained.[20] Three jump switching between four stable domain configurations, each aligned close to one of the cubic easy axes, involves one jump of 180°. In case of domain wall propagation this jump can be made when the total energy gain is comparable to the energy cost in propagating domain walls. Magnetic domain structures, several hundred microns large, have been measured in ultrathin Co films on Cu(001).[21,22]

We will use a phenomenological energy model to explain the results in more detail and to determine the anisotropy constants for Co films grown with different deposition angles. In this model it is assumed that magnetic switching proceeds via the nucleation and propagation of domain walls, such that switching occurs at applied fields smaller than the anisotropy field. Furthermore, it is assumed that domain wall propagation as opposed to domain wall nucleation is the limiting factor in the magnetic switching process.

Figure 2 illustrates the deposition, anisotropy and applied field geometry used in the model. In the growth experiments, the azimuthal direction of deposition was along the close packed [110] direction. The MOKE measurements in Fig. 1 show that in addition to a cubic anisotropy (K_4) an uniaxial anisotropy (K_u) has to be considered. The orientation of the uniaxial easy axis is perpendicular to the deposition direction. Hence, the free energy density $E(\phi)$ for off-normal deposited Co films on Cu(001) can be written as

$$E(\phi)=K_u\sin^2(\phi)+\frac{K_4}{4}\cos^2(2\phi)-MH\cos(\phi-\theta), \quad (1)$$

where ϕ is the angle of magnetization with respect to the $[\bar{1}10]$ direction and θ is the angle between the applied field and the $[\bar{1}10]$ direction. The cubic anisotropy K_4 has been found to be negative for Co films on Cu(001),[21–24] i.e., the cubic easy directions are parallel and perpendicular to the deposition direction.

First, the magnetic switching behavior with the applied field parallel to the deposition direction is discussed. Figure

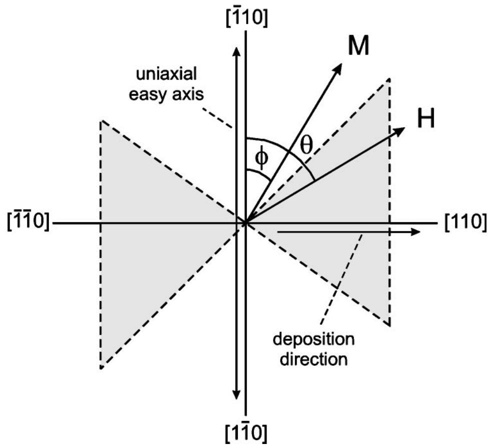

FIG. 2. The deposition, anisotropy and applied field geometry used in this paper. Off-normal deposition in the [110] direction results in an uniaxial magnetic anisotropy with the easy axis perpendicular to the plane of incidence. Longitudinal MOKE measurements are possible with the applied field at an angle between 45° and 125° with respect to the [-110] direction (shaded region).

1(a) shows that a transition from a single domain state in the $[\overline{1}\overline{1}0]$ direction to a state in the direction perpendicular to the deposition direction occurs at a large negative field. The exact domain structure after switching cannot be extracted from the measured hysteresis curve. It is possible that a small deviation in the applied field direction causes a single domain structure with the magnetization aligned along the $[\overline{1}10]$ direction when θ is a little less than 90° or with the magnetization aligned along the $[1\overline{1}0]$ direction when θ is a little more than 90°. However, the error in the applied field direction is small ($\pm 3°$). It is therefore more probable that both magnetization orientations are present after the first magnetic switching. Such coexistence of two domain orientations perpendicular to the applied field has been observed in Fe films on Ag(001).[25] After switching an almost linear increase of the Kerr ellipticity is observed. The external field which rotates the magnetization in the domains slightly towards the [110] direction causes this behavior. The magnetization rotation can be described by the equation of motion $dE(\phi)/d\phi=0$. For small rotation angles the slope of the hysteresis curve depends linearly on $1/(K_u-K_4)$:[26,27]

$$\frac{I_k}{H}=\frac{M_s I_s}{2(K_u-K_4)}, \tag{2}$$

where I_k/H is the slope of the hysteresis curve around zero field, M_s is the saturation magnetization, and I_s is the saturation Kerr ellipticity. In the case of larger rotation angles a slight deviation from the linear behavior can be observed. This makes a separation of K_u and K_4 possible when (K_u-K_4) is small.[27] In the off-normal deposited Co films separation was not possible and only the quantity (K_u-K_4) could be determined from the slope around zero field.

As outlined earlier, switching of the magnetization between the four cubic easy directions is mediated by domain wall propagation. Therefore, the equation of motion cannot be used to describe the observed irreversible transitions. Domain propagation makes reorientation of the magnetization in the Co films possible when the energy gain is comparable to the energy density cost for propagating domain walls. The magnetic switching behavior in thin films with an in-plane cubic and uniaxial magnetic anisotropy can be described by a simple phenomenological model.[19,20,25] In this model the activation energy involved in establishing a domain wall is ignored and only the energy needed to move a domain wall is considered. This energy, interpreted as the maximum height of the defect energy barriers that the domain walls encounter when they propagate, is indicated by $\epsilon_{90°}$ and $\epsilon_{180°}$ for 90° and 180° domain walls, respectively. The free energy density for single domain states with the magnetization oriented along one of the four cubic easy axes can be found by substituting the relevant values of ϕ into Eq. (1):

$$E_{[\overline{1}\overline{1}0]}=K_u+\frac{K_4}{4}+HM\sin(\theta), \tag{3a}$$

$$E_{[1\overline{1}0]}=\frac{K_4}{4}+HM\cos(\theta), \tag{3b}$$

$$E_{[\overline{1}10]}=\frac{K_4}{4}-HM\cos(\theta), \tag{3c}$$

$$E_{[110]}=K_u+\frac{K_4}{4}-HM\sin(\theta). \tag{3d}$$

Based on the MOKE measurements shown in Fig. 1, in which only small rotations away from the cubic easy axes are observed, we make the good approximation that the irreversible transitions observed in the hysteresis curves are caused by magnetic switching between the $\langle 110\rangle$ directions. Such a transition occurs when the energy density advantage ΔE in doing so is equal to the energy density cost in propagating a domain wall of the relevant type. For an applied field in the deposition direction ([110] direction) two irreversible jumps are measured. First, a transition from $[\overline{1}\overline{1}0]$ to $[1\overline{1}0]$ or $[\overline{1}10]$ occurs when $\Delta E=K_u+HM_s=\epsilon_{90°}$. Therefore, the switching field H_{s1} is given by: $H_{s1}=(-K_u+\epsilon_{90°})/M_s$. In the same way it can be derived that a second transition from $[1\overline{1}0]$ or $[\overline{1}10]$ to [110] occurs at a switching field $H_{s2}=(K_u+\epsilon_{90°})/M_s$. Measuring the two switching fields makes it possible to determine the uniaxial anisotropy: $K_u=(H_{s2}-H_{s1})M_s/2=H_sM_s$. Note that the shift field H_s does not equal the uniaxial anisotropy field H_u, which is usually defined as $H_u=2K_u/M_s$. The uniaxial anisotropy field is given by $H_u=2H_s$ instead. The relation for K_u together with the earlier derived relation for (K_u-K_4) will be used to determine the anisotropy constants for Co films grown with different angles of incidence.

With the help of the phenomenological energy model outlined above, the three jump switching behavior can be explained. For the 4 ML thick Co film deposited at an angle of 80°, three irreversible transitions were observed for applied

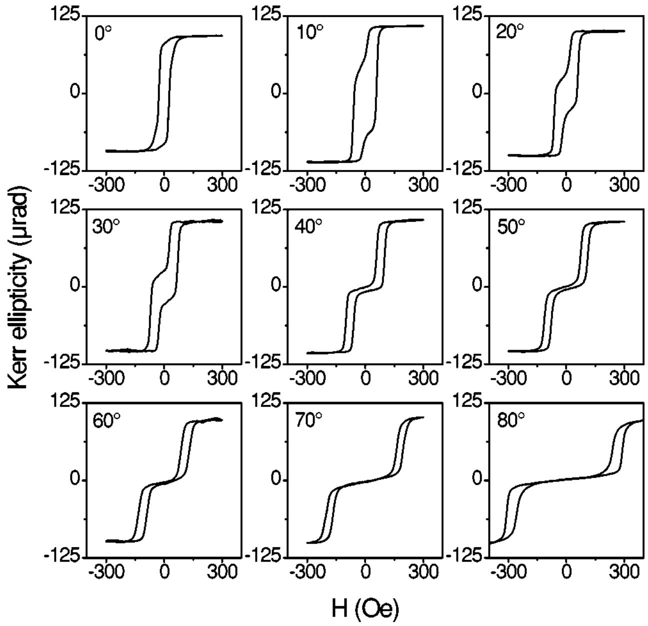

FIG. 3. In-plane hysteresis curves for Co films on Cu(001), grown at 250 K with different deposition angles. The magnetization curves were obtained at 175 K with the applied magnetic parallel to the deposition direction.

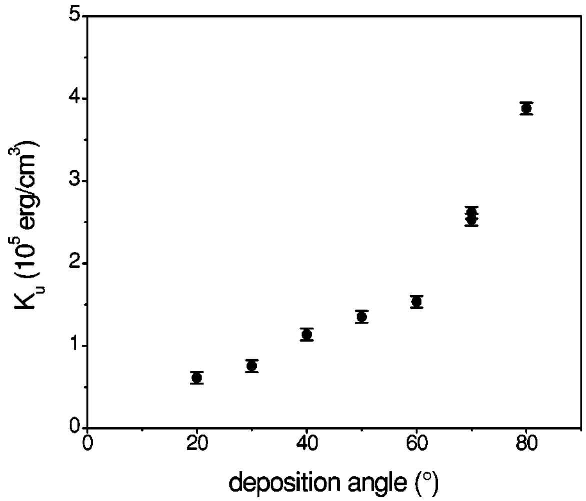

FIG. 4. Uniaxial magnetic anisotropy K_u in 5 ML thick Co films on Cu(001) as a function of the deposition angle.

fields with $60° < \theta < 80°$ and $100° < \theta < 120°$. For applied fields with $60° < \theta < 80°$, the first transition from $[\bar{1}\bar{1}0]$ to $[\bar{1}10]$ is followed by a transition from $[\bar{1}10]$ to $[1\bar{1}0]$. This second transition is mediated by the propagation of 180° domain walls. Finally, a third transition occurs from $[1\bar{1}0]$ to $[110]$. From a comparison of the energy densities along the four cubic easy directions it follows that a three jump switching route can only be observed when $K_u > \epsilon_{90°}$.[20]

The single jump hysteresis curve measured with an applied field 45° away from the deposition direction [Fig. 1(c)], i.e., with the applied field along a cubic hard axis, reveals switching of the magnetization from $[\bar{1}10]$ to $[1\bar{1}0]$ and vice versa. Rotation of the magnetization towards the direction of the applied field after switching is hindered by a large magnetic anisotropy, i.e., 300 Oe is not enough to saturate the magnetization. Theoretically, the remanent Kerr ellipticity should be smaller than the saturation Kerr ellipticity by a factor $1/\sqrt{2}$. From the saturation Kerr intensity measured in Fig. 1(a) and the remanent Kerr intensity measured in Fig. 1(c) it follows that this is indeed the case within experimental error.

The deposition angle dependence of the in-plane magnetic anisotropy in Co films on Cu(001) was studied for incident atom beam angles between 0° and 80°. Figure 3 shows an overview of MOKE results on 5 ML thick Co films grown at 250 K. The measurements are performed at 175 K with the applied magnetic field parallel to the deposition direction. After normal-incidence deposition only a small deviation from a square hysteresis loop is measured. The observed deviation is caused by a small uniaxial anisotropy, which probably originates from residual steps on the Cu(001) surface. As mentioned earlier, a magnetic step anisotropy has been measured for thin magnetic layers on stepped surfaces[1–3] and has been considered from a theoretical point of view as well.[3,4,28]

Deposition at an angle of 10° with respect to the surface normal results already in an off-normal growth-induced uniaxial magnetic anisotropy. In this case, two irreversible transitions are measured: first a transition from the uniaxial hard to the uniaxial easy and then from the uniaxial easy to the uniaxial hard magnetization axis occurs. The uniaxial anisotropy of this Co film is too small to align all spins in the direction perpendicular to the deposition direction. The measured remanent Kerr intensity is therefore nonzero. As one can see in Fig. 3, the shift field and thus the uniaxial magnetic anisotropy increases monotonically with increasing deposition angle up to 80°.

Following the phenomenological model the uniaxial anisotropy is given by $K_u = H_s M_s$, where M_s is the saturation magnetization of the Co film. Neutron diffraction experiments have shown that the magnetic moment of Co in Co/Cu(001) is the same as in bulk Co.[29] Therefore, we use the fcc-Co bulk saturation magnetization ($M_s = 1422$ Oe) in this study. From the hysteresis curves in Fig. 3 and from many others, the uniaxial magnetic anisotropy is determined as a function of the deposition angle (see Fig. 4). Each data point in Fig. 4 is the result of an averaging over at least three independently measured hysteresis curves. Obviously, the uniaxial magnetic anisotropy increases with increasing deposition angle. For a 5 ML thick Co film the uniaxial anisotropy is largest when the Co atoms are deposited at an angle of 80° with respect to the surface normal.

Figure 5 shows the quantity $(K_u - K_4)$ as a function of the deposition angle. Qualitatively, the angular dependence of $(K_u - K_4)$ is similar to that of K_u: the magnetic anisotropy $(K_u - K_4)$ increases up to 80°. Even though we averaged over at least three hysteresis curves, the small slope around zero field still results in relatively large error bars. Below a deposition angle of 40° an accurate determination of $(K_u - K_4)$ was not possible. At these deposition angles the remanent Kerr ellipticity is relatively large. This indicates that

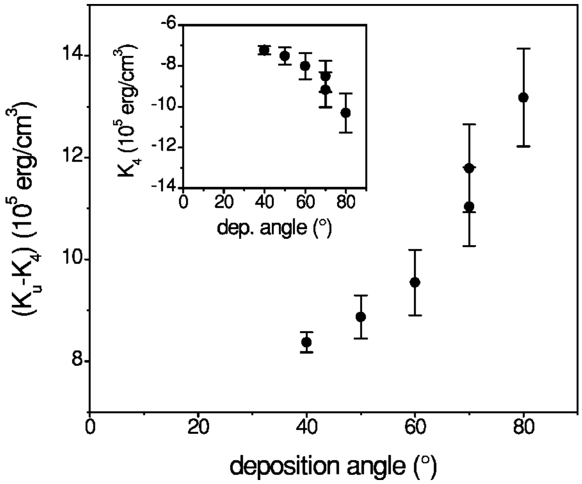

FIG. 5. Total magnetic anisotropy K_u-K_4 in 5 ML thick Co films on Cu(001) as a function of the deposition angle. The inset shows the cubic anisotropy constant K_4.

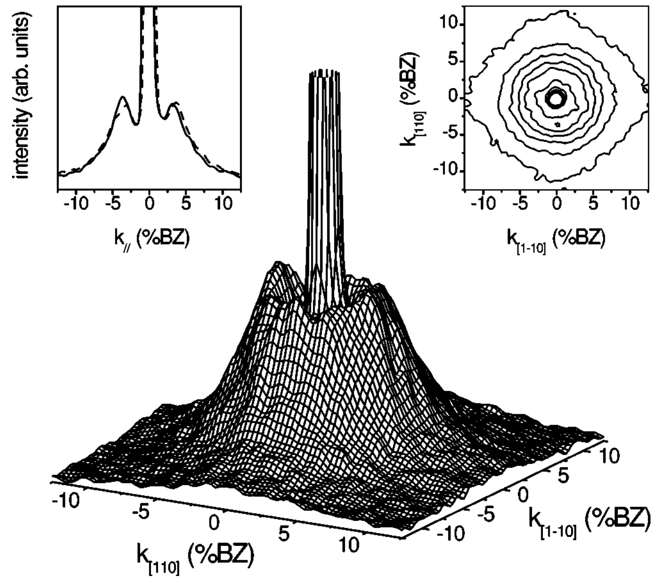

FIG. 6. SPA-LEED peak profile of the specular beam acquired after normal-incidence growth of 5 ML Co on Cu(001) at 250 K. The peak profile was obtained at $E = 290$ eV ($S_z = 4.75$). The left inset shows two line scans through the specular beam in the [110] (solid line) and [1-10] direction (dashed line).

domains parallel and perpendicular to the deposition direction are both present after the first magnetic switching. The slope of the hysteresis curve around zero field can therefore be the result of two effects: coherent magnetization rotation and a change of the population of domains by the propagation of domain walls.

In the inset of Fig. 5 the cubic magnetic anisotropy K_4 is plotted. Despite relatively large error bars the conclusion can be drawn that the absolute value of the cubic anisotropy increases slightly with increasing deposition angle. Extrapolation leads to a cubic anisotropy constant of $-7.0 \pm 1.5 \times 10^5$ erg/cm^3 for a 5 ML thick Co film grown at normal incidence. This anisotropy constant is close to the values for 4 ML and 10 ML thick Co films on Cu(001) found by Heinrich *et al.*[24] (-7.2×10^5 erg/cm^3 and -9.2×10^5 erg/cm^3, respectively). The result is, however, inconsistent with the total cubic anisotropy constant for 5 ML Co/Cu(001) reported in Ref. 23 (-2.1×10^5 erg/cm^3). Since magnetic anisotropy in Co/Cu(001) changes rapidly around a film thickness of 4 ML,[27] the discrepancy with the latter experimental result might be due to small differences in thickness calibration. To get an idea of the strength of the uniaxial anisotropy in off-normal deposited Co films, we compare the values of K_u with the uniaxial anisotropy found in Co/Cu(1 1 13).[8] In a Brillouin light scattering study the total uniaxial magnetic anisotropy constant for a 5 ML thick Co film on Cu(1 1 13) was determined to be 4.0×10^5 erg/cm^3 (interpolation of Fig. 3 in Ref. 8). We measured a comparable uniaxial magnetic anisotropy strength in 5 ML thick Co films on Cu(001), deposited at 80° and with the substrate at 250 K (see Fig. 4). The growth-induced uniaxial anisotropy in Co/Cu(001) is smaller after deposition at less grazing incidence.

B. Surface morphology

The measured uniaxial magnetic anisotropy in Co films is related to the formation of an uniaxial surface morphology during off-normal growth. SPA-LEED peak profiles measured after Co growth and magnetic characterization reveal distinct differences between normal and off-normal deposited Co films. Figure 6 shows a profile of the specular beam obtained after normal-incidence deposition of 5 ML Co on Cu(001) at 250 K. With increasing wave vector k parallel to the surface, two different patterns evolve. Close to the central (00) beam a circular ring is observed, whereas at larger k the pattern clearly shows a fourfold symmetry. The change from circular to fourfold symmetry indicates that the diffraction pattern consists of two different contributions, one from a quite narrow structure separation distribution, the other from a structure size distribution. The adatom structure separation contribution shows up as a circular first order diffraction ring at low k. The homogeneous ring intensity measured after normal-incidence Co growth reflects an isotropic radial distribution of square adatom structures. From the position of the ring in reciprocal space, the average adatom structure separation L is estimated to be $L \approx 80$ Å . As adatom structure sizes are necessarily smaller than their separation, the island size contribution to the diffraction pattern shows up at larger k. Consequently, the fourfold symmetry at larger wave vector in Fig. 6 is due to Fraunhofer diffraction and reflects the square shape of adatom structures. The square adatom structures are distributed with their edges oriented along the close-packed ⟨110⟩ directions. The formation of square adatom structures during normal-incidence Co growth on Cu(001) is in accordance with scanning tunneling measurements.[30–32] The near-equilibrium structure shape is due to a sufficiently large atom mobility along step edges.

In contrast to normal-incidence deposition, off-normal MBE destroys the fourfold symmetry of the film morphology. Instead, a twofold symmetry emerges with the plane of incidence acting as a mirror plane. Figure 7 shows a profile of the specular beam obtained after deposition of 5 ML Co on Cu(001) at an angle of 80° with the substrate at 250 K. In

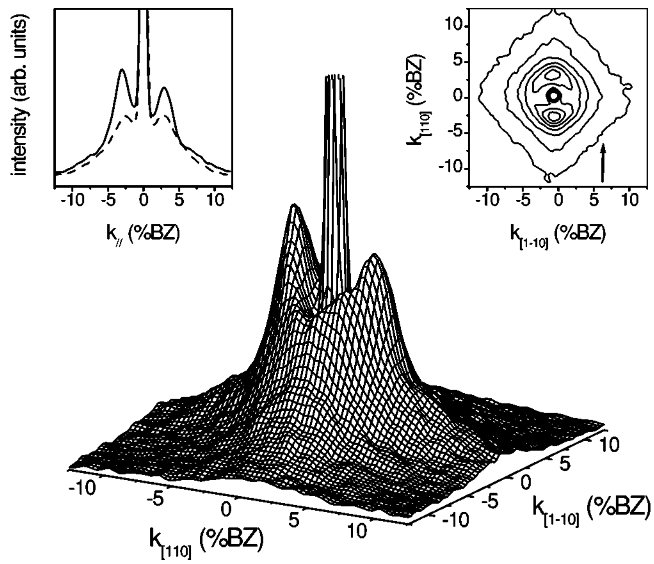

FIG. 7. SPA-LEED peak profile of the specular beam acquired after deposition of 5 ML Co at 80° with the Cu(001) substrate at 250 K. The peak profile was obtained at $E=290$ eV ($S_z=4.75$). The left inset shows two line scans through the specular spot in the [110] (solid line) and [1-10] direction (dashed line). The arrow in the contour plot indicates the deposition direction.

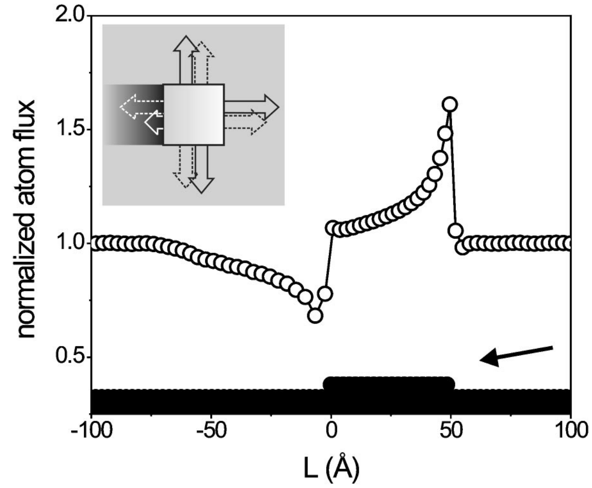

FIG. 8. Calculated incident atom flux at the surface, normalized to a homogeneous atom flux far above the surface for a surface with a monolayer high island on top of it and a deposition angle of 80°. The direction of the incident molecular beam is indicated by an arrow. The inset illustrates the steering-induced elongation of adatom structures. The grey scale is a measure for the incident atom flux during off-normal deposition. The solid and dashed arrows indicate the step edge growth rate during off-normal and normal deposition, respectively.

this case, the ring around the central (00) beam is not homogeneous but exhibits a clearly developed twofold symmetry. The remarkable beam profile with maxima in the deposition direction is interpreted as resulting from an isotropic radial distribution of *elongated* adatom structures in contrast to *square* ones growing at normal incidence. The elongated adatom structures are oriented with their long sides perpendicular to the plane of incidence of the Co atom beam, i.e., parallel to the easy magnetization axis. The diffraction intensity of the two maxima in the diffraction ring clearly differ from each other in Fig. 7. This asymmetry is not observed after deposition of 0.5 ML Co on Cu(001) at an angle of 80°. The asymmetry in the plane of incidence reveals the evolution of a different facet orientation at the illuminated and shadow side of adatom structures. The difference in facet orientation makes a straightforward quantification of the adatom structure aspect ratio difficult. However, from the 1.05 aspect ratio found after deposition of 0.5 ML Co/Cu(001) (fully two-dimensional system) and the substantial increase of the aspect ratio with film thickness (ripple structures are measured after deposition of about 20 ML Co at 80°) the aspect ratio for 5 ML Co/Cu(001) grown at 80° is roughly estimated to be 1.3.

The formation of elongated adatom structures during off-normal growth is explained by a phenomenon we introduced recently: *steering-enhanced roughening.*[14] Steering originates from long-range attractive forces between incident atoms and substrate atoms and leads to preferential arrival of atoms on top of adatom structures. Thermal Co atoms, approaching the surface at energies of about 0.15–0.20 eV, experience a long-range several-eV-deep attractive well. The attraction of incident atoms gives rise to substantial deflection of off-normal deposited atoms toward the surface. Initially, this has no consequences: the deflection is the same for all atoms and therefore the incident atom flux remains homogeneously distributed. However, as soon as adatom structures start to form, the redistribution of incident atom flux becomes progressively more important. Surface roughness causes a distortion in the attractive potential, and therefore atom trajectories are influenced by the local surface morphology. The result is a redistribution of incident atom flux in such a way that atoms arrive preferentially on top of adatom structures. To substantiate this phenomenon we performed a number of atom trajectory calculations. In these calculations we adopted a Lennard-Jones (12,6) pairwise potential[33] which has been found to describe the attractive forces between incident atoms and substrate atoms reasonably well.[34] From the atom trajectory calculations we derived the inhomogeneous incident atom flux at the surface normalized to the homogeneous atom flux far above the surface. Figure 8 shows the result for deposition at 80° on a surface with a monolayer high adatom structure on top. The atom flux enhancement factor amounts about 1.6 at the front of the adatom structure and decreases to one going further downstream. As should be the case for particle conservation reasons, the enhancement of incident atom flux on top of the adatom structure is exactly compensated by an incident atom flux reduction behind the adatom structure. Note that, in contrast to geometric shadowing, the incident atom flux behind the adatom structure is never zero due to the deflection of atoms toward the surface. A direct consequence of incident atom flux redistribution is a change in the adatom structure growth rate. At off-normal angles of incidence less atoms are deposited close to the shadow side of adatom structures. As a result, the growth rate of the shadow side of adatom structures is reduced. The reduction of the adatom growth rate in the deposition direction is only compensated when all additional atoms arriving on top of adatom structures contribute to the growth of the illuminated and/or shadow side. A part of the additional atom flux, however, leaks away to the perpendicular sides. Therefore, the in-plane adatom structure growth rate is smaller in the deposition direction than in the perpendicular direction (this is illustrated in the inset of Fig. 8). As a consequence, elongated adatom structures evolve with the long sides perpendicular to the plane of incidence of the Co atom beam.

We have calculated numerous atom trajectories for different surface morphologies. These calculations show that the amount of steering depends critically on two parameters: the surface roughness and the deposition angle. With increasing surface roughness the distortions in the attractive potential become more pronounced. Therefore, the atom trajectories differ more from each other with a more inhomogeneous incident atom flux as result: steering-enhanced roughening is autocatalyzed. For normal-incidence deposition only a small local enhancement of the incident atom flux close to step edges is calculated. However, as the deposition angle increases the lateral range as well as the total amount of incident atom flux on top of adatom structures increase, with a larger adatom structure aspect ratio as a result. This is in agreement with the SPA-LEED measurements. The monotonic increase of the in-plane uniaxial magnetic anisotropy (see Fig. 4) is fully attributed to a steering-induced elongation of adatom structures in the direction perpendicular to the plane of incidence.

An in-plane uniaxial magnetic anisotropy has been observed after off-normal deposition of Co on Cu(001) before.[35] Remarkably, though rectangular adatom structures were suggested as a possible explanation for the uniaxial anisotropy, the formation of these elongated structures and thus the uniaxial magnetic anisotropy was not ascribed to off-normal growth. In that study, the small miscut of the Cu crystal (only 0.1°) was used to explain the possible formation of rectangular adatom structures. Obviously this is not in agreement with our off-normal growth experiments and calculations of the steering phenomenon: rectangular adatom structures develop because of considerable steering effects at off-normal deposition.

For relatively thick off-normal deposited Co films on glass substrates (15–100 nm) a critical deposition angle at which the easy magnetization axis rotates by 90° has been found[12]: below and above this critical angle the easy magnetization axis is perpendicular and parallel to the deposition direction, respectively. Such a rotation is not observed in ultrathin Co films on Cu(001). In 5 ML thick Co films (≈ 1 nm) the easy magnetization axis is always oriented perpendicular to the deposition direction (see Fig. 4). We attribute this different magnetic behavior to a phenomenon that gains importance with increasing film thickness/roughness: *shadowing*. As the surface roughens, the redistribution of incident atom flux and thus shadowing become progressively more important. As long as the shadow regions are small, coalescence of adatom structures is reduced in the deposition direction only. In that case, rectangular adatom structures coalesce preferentially in the direction perpendicular to the plane of incidence with even more elongated adatom structures as result. With increasing surface roughness, however, the shadow length increases and coalescence of adatom structures is reduced in the direction perpendicular to the plane of incidence as well. Now, isolated and thus less elongated adatom structures grow. Such a change from an elongated to an isolated surface morphology with increasing deposition angle has been observed in homoepitaxial growth experiments on Cu(001).[15] For example, deposition of 40 ML Cu (≈ 7 nm) at 80° with the substrate at 250 K results in well defined ripple structures, whereas deposition at 85° results in isolated mound structures. From the considerations above it is expected that the critical angle decreases with increasing surface roughness (film thickness).

The micromagnetic origin of the uniaxial anisotropy may arise from missing bonds at Co step edges (Néel-type anisotropy), uniaxial strain (magnetoelastic anisotropy), and shape effects (shape or magnetostatic anisotropy). For elongated adatom structures the number of step edge atoms and thus missing bonds is different in the two high symmetry directions. The uniaxial magnetic anisotropy may therefore arise from this symmetry breaking. This is plausible since recent Co growth experiments on stepped Cu(001) have shown that the step-induced uniaxial anisotropy in this system arises from the missing bonds at step edges.[5,6] The strain in elongated adatom structures, which originates from a 1.9% lattice mismatch, may be relaxed anisotropically. Magnetoelastic effects may therefore also contribute to the uniaxial magnetic anisotropy.[36] The magnetostatic contribution to the total anisotropy will align the easy magnetization axis parallel to the long sides of the elongated adatom structures, i.e., perpendicular to the deposition direction.

C. Annealing and Cu adsorption

The MOKE and SPA-LEED measurements reveal that the growth-induced uniaxial magnetic anisotropy is directly related to the formation of elongated adatom structures during off-normal deposition. A decreasing uniaxial magnetic anisotropy can therefore be expected when the fourfold symmetry of the surface is restored. To check this conjecture, we performed annealing experiments on off-normal deposited Co films. Annealing activates adatom diffusion processes which not only tend to smooth the Co film but reshape the adatom structures to their energetically favorable square form as well. The diffusion of step edge atoms around the corners of adatom structures and detachment/attachment processes can for example reduce the aspect ratio of elongated adatom structures. Figure 9 shows a selection of Kerr hysteresis curves measured during annealing of a 4 ML thick Co film which was deposited at an angle of 80° with respect to the surface normal (same film as in Fig. 1). The heating rate was about 0.08 K/s in this experiment. Up to a film temperature of 300 K the measured shift field H_s and thus the in-plane uniaxial magnetic anisotropy is nearly constant. Increasing the temperature further, however, results in a monotonic decrease of the shift field. At these temperatures the adatom diffusion processes responsible for surface smoothing and adatom structure reshaping become active on

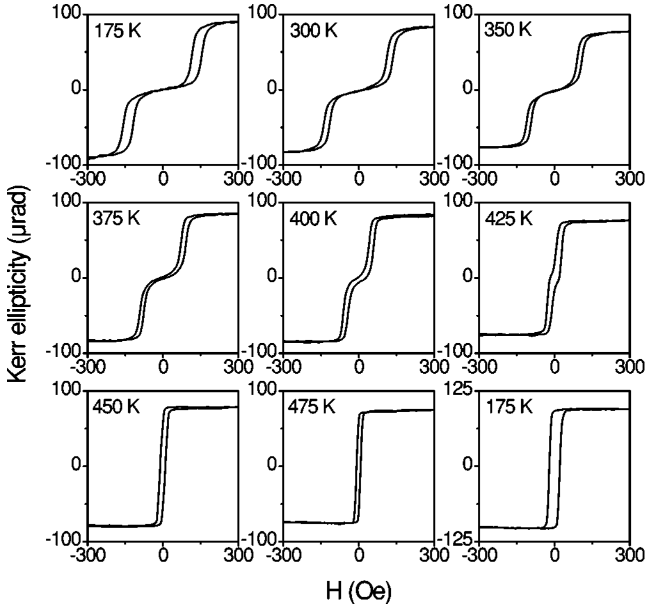

FIG. 9. In-plane hard axis hysteresis curves for a 4 ML thick Co film on Cu(001), grown at an angle of 80° with respect to the surface normal. The magnetization curves were obtained during annealing (heating rate: 0.08 K/s).

the experimental timescale. The square hysteresis curve measured at a film temperature of 450 K indicates that annealing the Co film to this temperature results in a negligible in-plane uniaxial magnetic anisotropy. A square hysteresis curve is measured also when the sample is cooled from 475 back to 175 K. The transition is therefore irreversible: annealing an off-normal deposited Co film destroys the growth induced twofold surface morphology. As a consequence, the in-plane uniaxial magnetic anisotropy, which is directly connected with the surface morphology, disappears. This is also illustrated in Fig. 10, which shows the uniaxial anisotropy constant K_u as a function of film temperature.

In addition to surface smoothing and reshaping of adatom structures, interdiffusion also occurs during annealing of Co

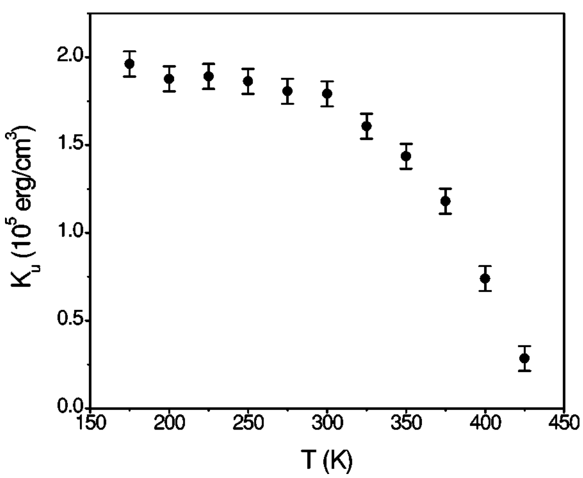

FIG. 10. Uniaxial magnetic anisotropy K_u in a 4 ML thick Co film, grown at 80° with the Cu(001) substrate at 250 K, as a function of annealing temperature.

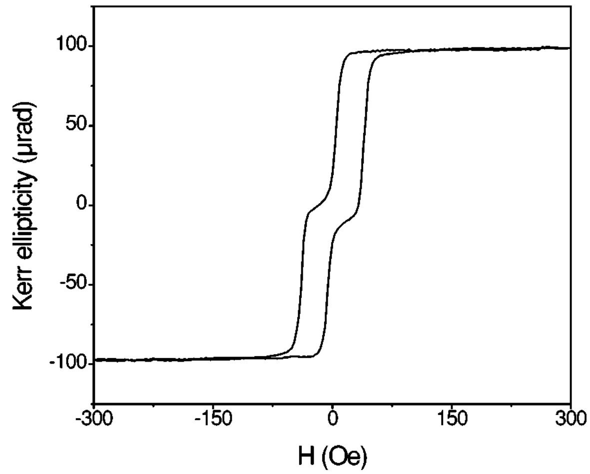

FIG. 11. In-plane hard axis hysteresis curve for a 5 ML thick Co film on Cu(001), grown at 300 K with an angle of 70° with respect to the surface normal. The magnetization curve was obtained at 175 K.

films on Cu(001).[37–40] At elevated temperatures Cu segregates towards the film surface leaving behind deep square pits in the substrate. The driving force for the segregation of Cu substrate atoms is the lower surface free energy of Cu compared to that of Co ($\sigma_{Cu}=1.9$ J/m^2 and $\sigma_{Co}=2.7$ J/m^2 [41,42]). For Co films on Cu(001) interdiffusion has been found at annealing temperatures above 450 K. As a consequence, interdiffusion cannot be excluded in the annealing experiment shown in Fig. 9 and Fig. 10. This is confirmed by annealing experiments on Co films grown on stepped Cu(001) substrates[18]: although surface steps remain, a strong reduction of K_u is observed above 350 K for 2.5 ML thick Co films. This reduction is most probably due to a decrease of the number of missing bonds at Co step edges caused by the segregation and subsequent attachment of Cu atoms.

From the annealing experiments it can be expected that the uniaxial magnetic anisotropy will be smaller after off-normal Co growth at elevated temperatures. Furthermore, it can be expected that a Cu cap layer affects the magnetic behavior of off-normal deposited Co films. A number of growth experiments were performed to check these conjectures. The first conclusion that can be drawn from these experiments is that the growth-induced uniaxial magnetic anisotropy decreases with increasing growth temperature indeed. Figure 11 shows a hysteresis curve acquired after off-normal deposition of 5 ML Co at 70° with the substrate at 300 K. The shift field in this hard axis magnetization curve is considerably smaller than that measured after off-normal growth with the same deposition angle but with the substrate at 250 K (see Fig. 3). The uniaxial anisotropy constant K_u is 2.5×10^5 erg/cm^3 and 0.3×10^5 erg/cm^3 after growth at 250 and 300 K, respectively, i.e., K_u decreases by about a factor 8. Two effects account for this. First of all, the adatom structure size increases exponentially with increasing growth temperature. For large adatom structures the *average* enhancement of the incident atom flux on top of the structures (flux enhancement per surface area) is small compared to that on

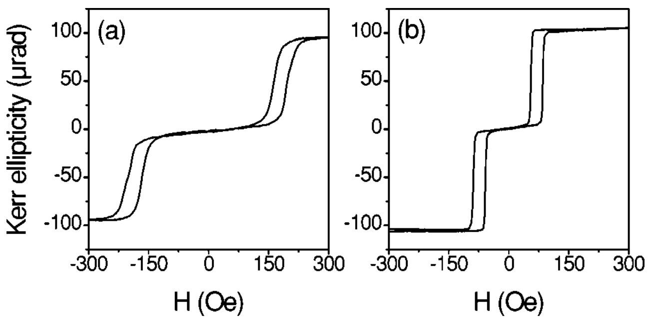

FIG. 12. (a) In-plane hard axis hysteresis curve for a 5 ML thick Co film on Cu(001), grown at 250 K with an angle of 70° with respect to the surface normal. (b) In-plane hard axis hysteresis curve for the same Co film acquired after normal-incidence growth of 0.5 ML Cu at 250 K. Both magnetization curves were obtained at 175 K.

top of smaller structures. The adatom structure growth rate in the deposition and perpendicular direction will therefore differ less at elevated growth temperatures with a smaller aspect ratio (less elongated shape) as result. Second, due to an enhanced surface diffusion at higher growth temperatures the adatom structure shape deviates less from the energetically favorable square shape. In other words, surface diffusion reduces the consequences of steering.

From experiments on stepped surfaces it is well known that the adsorption of Cu atoms onto stepped Co films leads to a strong reduction of the step-induced anisotropy strength.[5,6,16,17] Such a strong reduction of the uniaxial magnetic anisotropy is measured after Cu adsorption on our off-normal deposited Co films as well. Figure 12 shows hysteresis curves acquired before and after growth of 0.5 ML Cu on a 5 ML thick off-normal deposited Co film. The Co film was grown at 70° with the Cu(001) substrate at 250 K, whereas the Cu was deposited at normal incidence and at the same temperature. Obviously, Cu adsorption leads to a reduction of the uniaxial magnetic anisotropy. From the shift field the anisotropy constant is determined to be $K_u = 1.0 \times 10^5$ erg/cm^3 after Cu adsorption, which is about a factor 2.5 smaller than the anisotropy constant for the uncovered Co film. No further decrease of the in-plane uniaxial magnetic anisotropy is measured for larger amounts of Cu deposits. The measurements in Fig. 12 seem to indicate that the micromagnetic origin of the uniaxial anisotropy in off-normal deposited Co films arises mainly from missing bonds at the Co step edges. During Cu growth the number of missing bonds is reduced by the attachment of Cu atoms to Co steps. When all the step edges are fully decorated by Cu atoms, no further decrease of K_u can be expected. This is obviously the case after normal-incidence growth of 0.5 ML Cu.

The adatom structure aspect ratio can be estimated from a comparison between the step-induced uniaxial anisotropy in off-normal deposited Co films [$K_{u(\mathrm{off})} \approx 1.5 \times 10^5$ erg/cm^3 at 70°] and the step-induced uniaxial anisotropy in Co films grown on stepped Cu(1 1 13) surfaces [$K_{u(\mathrm{step})} \approx 4.0 \times 10^5$ erg/cm^3 in Ref. 8]. The aspect ratio (AR) follows from

$$\frac{2N\left(1-\frac{1}{\mathrm{AR}}\right)D}{L^2} = \frac{K_{u(\mathrm{off})}}{dK_{u(\mathrm{step})}}, \tag{4}$$

where N is the average length of the long side of adatom structures, L is the average separation between adatom structures, D is the step-step separation on Cu(1 1 13), and d is the number of exposed surface layers after off-normal deposition. For $N = 25$ atoms, $L = 30$ atoms, $D = 6.5$ atoms, and $d = 5$ ML an aspect ratio of 1.26 is obtained. This is in reasonable agreement with a rough estimation from SPA-LEED measurements.

IV. CONCLUSIONS

The influence of the deposition angle on the magnetic anisotropy in ultrathin Co films on Cu(001) was studied for angles between 0° and 80°. Off-normal molecular beam epitaxy along the [110] azimuth results in an in-plane uniaxial magnetic anisotropy. For deposition angles ⩾10° the easy magnetization axis is oriented perpendicular to the plane of incidence. The strength of the uniaxial magnetic anisotropy increases monotonically upon rotation of the molecular beam from normal to more grazing incidence. SPA-LEED measurements reveal that the observed magnetic behavior in Co/Cu(001) is directly related to the formation of an uniaxial surface morphology during off-normal growth: elongated instead of square adatom structures evolve. The long sides of the elongated adatom structures are oriented perpendicular to the plane of incidence and the structure aspect ratio increases with increasing deposition angle. The influence of the deposition angle on the evolution of the surface morphology is rationalized in terms of steering. Steering is a direct consequence of long-range attractive forces between incident atoms and substrate atoms and leads to a redistribution of incident atom flux: the incident atoms arrive preferentially on top of adatom structures at the cost of flux reduction behind structures. Due to this redistribution of incident atom flux the adatom structure growth rate is larger in the direction perpendicular to the plane of incidence, with the result that elongated structures evolve. While steering increases with the deposition angle, the measured increase of the aspect ratio is explained naturally by this phenomenon as well. The surface morphology and thus the magnetic behavior of an ultrathin Co film are the result of an interplay between steering and surface diffusion processes. Therefore, growth at elevated temperatures or post annealing reduces the adatom structure aspect ratio and as a result the uniaxial magnetic anisotropy drastically. The micromagnetic origin of the uniaxial magnetic anisotropy in off-normal deposited Co films arises mainly from missing bonds at Co step edges. Steering-induced uniaxial magnetic anisotropy should always be anticipated in off-normal MBE growth of magnetic films.

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