Structural relaxation and magnetic anisotropy in Co/Cu(001) films

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(Received 23 February 1996)

The magnetic anisotropy of Co/Cu(001) films has been investigated by the magneto-optical Kerr effect, both in the pseudomorphic growth regime and above the critical thickness where strain relaxation sets in. A clear correlation between the onset of strain relaxation—as measured by means of reflection high-energy electron diffraction—and changes of the magnetic anisotropy has been found. [S0163-1829(96)02330-2]

Thin films grown on a substrate are generally subject to strain arising from different lattice parameters of the film material and the substrate. In a ferromagnet, the existence of this epitaxial strain gives rise to an additional magnetic anisotropy contribution or, more specifically, to the magnetoelastic energy term. It has been shown that strain causes two relaxation mechanisms in ultrathin films. In a limited thickness range up to a critical thickness, the bulk of the film has the same lattice parameter as the substrate. In this pseudomorphic growth regime, relaxation occurs only at the incomplete surface layer. The surface lattice can relieve the strain by a relaxation of the atomic positions at island edges.[1,2] On the other hand, for film thicknesses above the critical thickness, the misfit-induced strain[3] is relieved by the creation of interfacial dislocations. At these thicknesses the lattice misfit-induced magnetic anisotropy can be regarded as an effective surface anisotropy; i.e., it is inversely proportional to the film thickness.[4] Measurements on Au/Co/Au(111),[4] Cu/Ni/Cu(001),[5] Co/W(110),[6] and Cu/Co/Cu(110) films[7] showed that magnetic anisotropy indeed exhibits such behavior above a critical thickness.

In this study Co films on Cu(001) are investigated both by reflection high-energy electron diffraction (RHEED) and by the magneto-optical Kerr effect in order to correlate the magnetic and structural behavior of this model system. Although Co/Cu(001) is one of the most extensively studied systems, such a correlation has not yet been drawn. In particular the influence of lattice relaxation on magnetic anisotropy has not been determined. Early experiments with transmission electron microscopy[8] showed that Co films grown at room temperature onto a Cu/NaCl(001) substrate have a critical thickness of 11 monolayers (ML, 1 ML = 0.18 nm), above which the Co film begins to relax its misfit-induced strain. In our Co/Cu films, critical thicknesses between 10 and 20 ML were found depending on the substrate temperature during growth. We show that this strain relaxation is accompanied by changes of magnetic anisotropy.

Structural and magnetic investigations were performed in two UHV chambers. For structural measurements the Co films were grown on a Cu(001) single crystal at substrate temperatures between 20 and 90 °C and a deposition rate of 0.2-2 ML/min. Line scans across RHEED images were recorded during growth every 1–3 s. The incident electron beam had an energy of 35 keV and an angle of 3° with respect to the surface plane. The surface projection of the incident electron beam pointed into the [110] direction. The in-plane lattice spacing of the topmost Co layers was determined from the measured separation of the $(\bar{1}0)$ and the (10) RHEED reflections as described in Ref. 2. For the magnetic measurements hysteresis loops were recorded during growth of the film using the magneto-optical Kerr effect. For these experiments a stepped Cu single crystal was used. An average step width of 100 nm is induced by a slight miscut of 0.1° from the (001) orientation, with the preferential step edges running along $[1\bar{1}0]$. This allowed us to determine the magnetic anisotropy of the Co films easily. These films were evaporated at room temperature at a rate of 0.05 ML/min. Prior to film deposition, both Cu substrates were cleaned by sputtering and annealing up to 500–700 °C.

Results of the structural investigations are shown in Fig. 1, where the normalized distances between the $(\bar{1}0)$ and the (10) RHEED reflections are plotted as a function of Co coverage at two different growth temperatures, 20 and 60 °C. Oscillations with a period of 1 ML are observed in the entire Co thickness range up to 40 ML. As discussed in detail elsewhere, these oscillations are caused by an oscillatory relaxation of the lattice constant of the topmost surface layer.[2] The free edges of the Co islands in the incomplete top layer are distorted owing to reduced coordination and thus modify the average lattice constant at the surface. When these islands merge during growth to complete the layer, the average lattice constant changes as well. Therefore, with increasing film thickness the lattice constant varies in an oscillatory manner.

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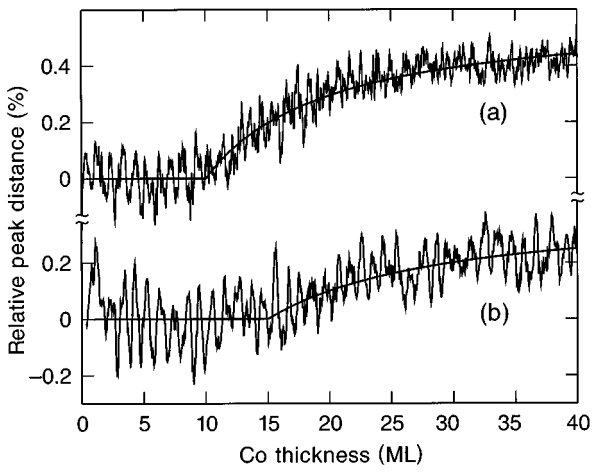

FIG. 1. Relative change of peak separation between the $(\bar{1}0)$ and the (10) RHEED reflections compared to initial peak separation as a function of Co thickness. (a) Substrate temperature 60 °C, (b) substrate temperature 20 °C. The lines through the data points are fits with the function $\eta-\epsilon(d)=\eta-\eta[\alpha+(1-\alpha)d_c/d]$, where $\eta=0.018$, (a) $\alpha=0.67$ and (b) $\alpha=0.78$, respectively.

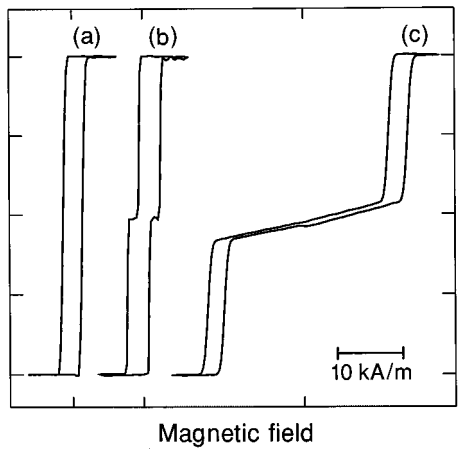

FIG. 2. Hysteresis loops $M(H)$ obtained using the magneto-optical Kerr effect on a 6-ML Co film grown on the stepped Cu(001) substrate. (a) H along the $[1\bar{1}0]$ direction, (b) H along the [110] direction, and (c) same as (b) but having a bias field of $H_{\text{bias}}=13$ kA/m along the $[1\bar{1}0]$ direction.

These oscillations are superimposed on a constant background strain up to a critical thickness d_c and on a monotonically increasing relaxation of the average surface lattice constant above d_c. This relaxation behavior is typical for the appearance of dislocations within the film at d_c.[8] Above d_c, the dislocated film is more stable than the coherently strained film: The energy gained from the relief of misfit strain is greater than the energy expended by the creation of dislocations. We emphasize that all strain measurements are measurements of the surface strain because RHEED is a surface-sensitive technique. Concerning the monotonically increasing relaxation above d_c, however, arguments have been given that the measured surface strain can be identified with the average strain of the film.[6]

The data for 20 °C show only a weak relaxation starting at 15 ML. Even for thicknesses of 40 ML the film is still strongly strained. At higher substrate temperatures the critical thickness where relaxation starts to become smaller is reduced to 10 ML for 60 °C [see Fig. 1(a)] and to 6 ML for 90 °C (not shown). Although relaxation is much stronger at 60 °C, the Co film is still strained within the entire thickness range investigated. The temperature dependence of d_c shows that the strain of the Co film is markedly influenced by thermal energy. We interpret these observations by a reduced mobility of dislocations at lower temperatures.

In the simplest model the thickness dependence of the misfit-induced strain ϵ in equilibrium can be described as $\epsilon(d)=\eta d_c/d$ for $d>d_c$, where η is the misfit between substrate and film.[4,9] However, no agreement of the experimental data with this function can be obtained in Co/Cu(001), even if we assume that the Cu surface lattice constant itself is already relaxed by 1%.[10] This means that even the thickest films do not relax to zero misfit strain. A more appropriate description for the relaxation behavior is given by $\epsilon(d)=\eta[\alpha+(1-\alpha)d_c/d]$, where α describes the residual strain.[6] Such a residual strain has been observed for both the Ni/Cu(001) (Refs. 9, 11) and Co/W(110) systems.[6] For the Co/Cu system, this strain is rather large. For the investigated substrate temperatures shown in Fig. 1 we find $\alpha\approx 0.7$.

Both the lattice relaxation in the pseudomorphic thickness range and the strain relaxation by dislocations above d_c cause appreciable changes in the magnetic anisotropy, as will be shown in the following.

The magnetic anisotropies can easily be determined on the stepped Co/Cu film by analyzing the hysteresis loops. Figure 2 shows such loops of a 6-ML Co film with the magnetic field H applied either parallel (along $[1\bar{1}0]$) or perpendicular (along [110]) to the step edges of the Cu substrate. For H along the $[1\bar{1}0]$ direction a rectangular loop is found, the signature of an easy magnetization axis. On the other hand, the hysteresis loop for H applied along [110] shows that this direction is an intermediate magnetization axis.[12] It consists of two shifted single loops. These loops are characterized by a shift field H_s, which we define as the magnetic field difference between zero field and the center of these single loops. The different responses of the magneto-optical signal in these two directions are due to the additional uniaxial magnetic anisotropy introduced by the preferential step direction on our Cu substrate.[13–15]

The in-plane free-energy density of a Co film on a stepped Cu(001) surface having the external magnetic field H applied along the [110] direction can be expressed as[16] $K_u\sin^2(\phi)+K_1/4\sin^2(2\phi)-\mu_0 HM_s\sin(\phi)$. Here, ϕ is the angle between the magnetization and the $[1\bar{1}0]$ direction, K_u and K_1 are the uniaxial and the cubic anisotropy constants, respectively, μ_0 is the vacuum permeability, and M_s the saturation magnetization. M_s has been determined from fits to Brillouin light scattering data[17] and agrees within 5 % with the Co bulk value, M_s = 1424 kA/m. Because K_u is small compared to K_1 in our case — the uniaxial anisotropy-inducing miscut is small — K_u is directly given by the shift field H_s of the hysteresis loops, $K_u=\mu_0 H_s M_s$.[18] By determining the linear initial slope s of the shifted loops, on the other hand, the cubic anisotropy constant K_1 can also be evaluated, $K_1=\mu_0 M_s^2/2s$.[18] However, in our films the slope s cannot be deduced directly from hysteresis loops such as

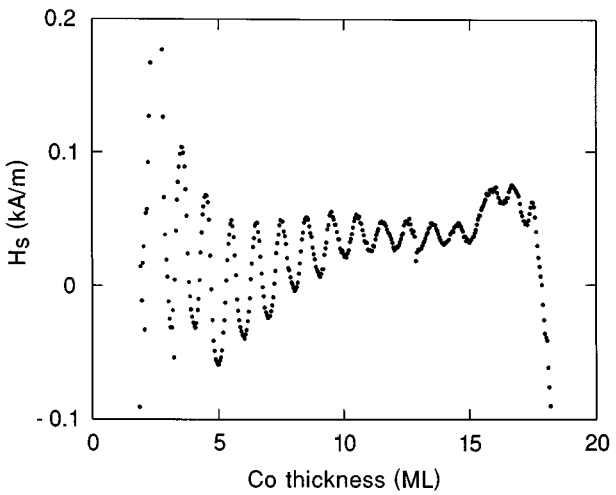

FIG. 3. Shift field H_s vs Co coverage deduced from intermediate axis hysteresis loops. A bias field of 5 kA/m has been applied along the easy [1$\bar{1}$0] direction.

the one presented in Fig. 2(b) because the width of the single loops is comparable to H_s, and therefore no linear segment between the two shifted loops can be identified. Nevertheless, we can experimentally realize loops with an extended linear slope between the shifted loops by applying a constant bias field H_{bias} along the easy axis while recording a hysteresis loop along the intermediate axis. The bias field introduces an additional unidirectional anisotropy contribution, which results in a larger shift field of the two loops given by H_s+H_{bias}. From the wide hysteresis-free field region between the loops we can now determine the initial slope s; see Fig. 2(c). Due to the application of the bias field, it is possible to measure also negative H_s as long as $|H_s|<H_{\text{bias}}$. A sign change of H_s indicates a change of the easy magnetization axis by 90°. Note that the experimental trick to apply a bias field has a profound physical justification. The bias field forces the magnetization into the easy direction as soon as the sweeping field along the intermediate axis is reduced to zero, whereby a single-domain configuration is maintained. The increasing field along the intermediate axis then starts to tilt the magnetization reversibly away from the easy axis. This tilt angle is measured directly by determining the magnetization component along the intermediate axis or, in other words, by determining the initial slope of the hysteresis loops. It signifies the anisotropy barrier against which the magnetization has to be rotated.

Figure 3 presents the shift field H_s deduced from intermediate axis hysteresis loops as a function of Co thickness. The shift field displays pronounced oscillations with a period of 1 ML, with H_s minima at the completion of each full layer. This oscillatory behavior of the uniaxial magnetic anisotropy has recently been reported.[18] Figure 3 shows that the amplitude of the magnetic anisotropy oscillations decays with increasing Co film thickness. In fact, the $1/d$-like decrease indicates that the mechanism leading to the anisotropy oscillations is a surface-related relaxation process. Thus it is tempting to relate these oscillations of the magnetic anisotropy to the 1-ML oscillations observed in the RHEED experiment. A distortion of the surface lattice is likely to modify the magnetoelastic anisotropy and, hence, the shift field.

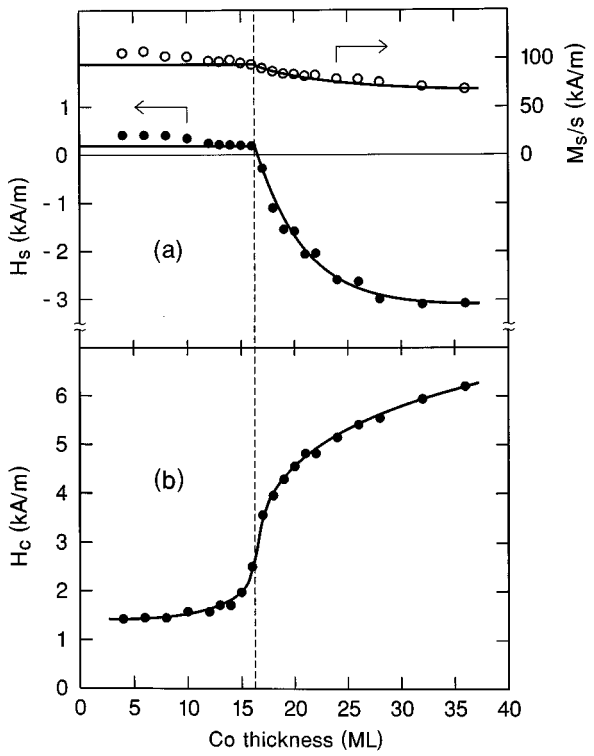

FIG. 4. (a) M_s/s (open circles, right scale) and shift field H_s (solid circles, left scale) as a function of Co thickness deduced from intermediate axis hysteresis loops. A bias field of 13 kA/m has been applied along the easy [1$\bar{1}$0] direction. (b) Coercive field H_c of the easy-axis hysteresis loops vs the Co thickness. Lines to guide the eye have been drawn through the data points. With $M_s=1424$ kA/m the field values can be converted to the anisotropy constants K_u and K_1.

At 17 ML, H_s drops markedly. The uniaxial anisotropy changes sign, and thus the easy magnetization axis flips from the [1$\bar{1}$0] to the [110] direction. Note that for growth at room temperature the pseudomorphic growth regime breaks down approximately at this thickness; see Fig. 1. To relate structural and magnetic information above the critical thickness more directly, we performed an additional experiment in which we also determined the cubic anisotropy.

Figure 4 shows both the shift field H_s and M_s/s as a function of Co thickness. Moreover, the coercive field H_c determined from the easy-axis hysteresis loops is shown. For these measurements a strong bias field of 13 kA/m has been applied in order to obtain reliable values of the slope in the entire thickness range investigated, in particular in the region where the coercive field H_c increases drastically. For Co coverages below 16 ML the easy magnetization axis is along [1$\bar{1}$0] and thus the H_s values are positive. Above 16 ML the uniaxial anisotropy drops. At the thickness where the uniaxial magnetic anisotropy crosses zero, the anisotropy of the Co film is purely fourfold symmetric, and correspondingly the hysteresis loops exhibit a rectangular shape in both the [1$\bar{1}$0] and the [110] directions. For larger thicknesses H_s assumes negative values, indicating that the easy magnetization axis has switched by 90° within the plane into the [110] direction perpendicular to the preferential step edges.

A switching of the easy magnetization axis within the

plane has already been observed in various films grown on twofold-symmetric substrates, such as Fe/GaAs(110),[19] Fe/W(110),[20] and Co/Cu(110).[7] For stepped Co/Cu(001) films, on the other hand, such a switching has not been published before.[21] Indeed it contradicts the earlier belief that the uniaxial anisotropy on stepped Co films will asymptotically vanish for large thicknesses and hence approach the cubic fourfold-symmetric in-plane anisotropy.[22] Instead, for thicknesses $d>16$ ML, the easy axis is perpendicular to the substrate step edges.

The switching thickness is rather insensitive to the average terrace width. We have performed additional experiments on Cu(001) surfaces with a different miscut orientation and hence a different terrace width. For miscuts angles of 1.6°, 1.9°, and 3.4° we find the same behavior as for the 0.1° miscut surface: The easy axis is parallel to the Cu step edges for thin Co films, but perpendicular for Co thicknesses above 15–20 ML. Note that these films remain uniaxial with the easy magnetization direction perpendicular to the substrate step edges up to the largest film thicknesses investigated, e.g., for the 1.6° miscut substrate up to 300 ML. In particular the strength of uniaxial anisotropy at these large Co thicknesses, which have reduced strain, is still much larger than at Co coverages below d_c, where strain is the highest. This is surprising because one would expect a decreasing strength of the uniaxial anisotropy for large Co thicknesses. In order to explain this we have to assume that relaxation preferentially takes place in a uniaxial manner. This can for instance be caused by an anisotropic dislocation network,[23] which is likely to occur on a stepped surface.

The cubic anisotropy displays the same general behavior as the uniaxial anisotropy; see Fig. 4(a). The drop at $d=16$ ML coincides with that of H_s, but the relative change of cubic anisotropy is much weaker. Figure 4(b) shows that the coercive field H_c determined from the easy-axis loops also changes markedly around 16 ML. The onset of the H_c change, however, occurs at a lower thickness than that of the magnetic anisotropies.

From the RHEED data in Fig. 1 we expect a critical thickness of $\approx$15 ML for our Kerr measurements. In fact, the thickness where the anisotropy starts to change is close to this value, 16–17 ML. As the magnetic and structural measurements were performed on different Cu substrates, perfect agreement cannot be expected. From the good agreement we conclude that the observed changes of magnetic anisotropy at 16–17 ML are indeed of structural origin. This also explains the strong increase of the coercive field H_c in the same thickness range. The formation of dislocations above d_c very likely enhances the pinning of magnetic domains, which leads to an increase of H_c. The onset of the change in H_c systematically precedes that of H_s by $\approx$2 ML. This can be taken as an indication that the coercive field is a much more sensitive probe of the onset of strain relaxation than is magnetic anisotropy: The presence of a few dislocations will not yet influence magnetic anisotropy, but it nonetheless governs domain wall pinning and motion.

From our magnetic and structural results we are able to deduce quantitatively the magnetoelastic constant B_1, which describes the extent to which elastic strain contributes to magnetic anisotropy energy. For a cubic system such as Co/Cu(001) the free magnetoelastic energy density F_{me} can be expressed as a function of the direction cosines of the magnetization α_i and of the strain tensor components ϵ_{ij} with respect to the xyz coordinate system ([100], [010], [001]): $F_{\mathrm{me}}=B_1(\epsilon_{11}\alpha_1^2+\epsilon_{22}\alpha_2^2+\epsilon_{33}\alpha_3^2)+B_2(\epsilon_{12}\alpha_1\alpha_2+\epsilon_{23}\alpha_2\alpha_3+\epsilon_{31}\alpha_3\alpha_1)$. The magnetoelastic constants are denoted B_1 and B_2. This general expression can be simplified in our case. For a biaxial strain all nondiagonal strain tensor components vanish. Moreover the easy magnetization axis is either along the $[1\bar{1}0]$ or the [110] direction. Then the magnetoelastic energy reduces to $F_{\mathrm{me}}=B_1(\epsilon_{11}+\epsilon_{22})/2$. The two in-plane strain components ϵ_{11} and ϵ_{22} are obtained from the RHEED study. As the RHEED experiment has been performed on a Cu(001) crystal, considered to be fourfold symmetric, $\epsilon_{11}=\epsilon_{22}$. If we further take into account that the surface projection of the incident electron beam was parallel to the [110] direction, we arrive at $F_{\mathrm{me}}=B_1\epsilon/\sqrt{2}$, where ϵ is the strain along [110]. By taking the room temperature RHEED results in Fig. 1(b) and the change of cubic anisotropy in Fig. 4(a) above the critical thickness, we find a magnetoelastic constant of $B_1=-0.5\times10^7$ J/m^3. No bulk magnetoelastic constants have been measured for fcc-Co. For Co-rich CoPd alloys, $B_1=-1.6\times10^7$ J/m^3 has been found by extrapolation.[24] Although the sign agrees, the magnitude deviates by a factor of 3. This discrepancy cannot be attributed to an additional magnetoelastic surface anisotropy[25] because in our case the film thickness is too large for this term to be significant. Instead, it shows that such an extrapolation is questionable.

In conclusion, changes of the magnetic anisotropy were found in Co films grown on a slightly miscut Cu(001) surface above a critical thickness. In particular an easy-axis switch of 90° within the plane can be identified. By comparing magnetic measurements with RHEED investigations these changes can be attributed to relaxation of the misfit-induced strain in the Co lattice upon growth. A quantitative analysis reveals that the magnetoelastic constant B_1 of Co films on Cu(001) differs appreciably from extrapolated literature bulk values.

Financial support by the the European Community and the BBW within the HCM program "Magnetic properties of novel ultrathin films" under contract No. CHRX-CT93-0316 and the Deutsche Forschungsgemeinschaft, Project No. SFB 341 is gratefully acknowledged.

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