Temperature-induced magnetic anisotropies in Co/Cu(1 1 17)

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The temperature dependence of magnetic anisotropy in cobalt films grown on Cu(1 1 17) has been studied by means of the magneto-optic Kerr effect. An in-plane uniaxial anisotropy is found in the as-grown films. At elevated temperatures the films exhibit changes in anisotropy. At temperatures around 100 °C a transition to nearly biaxial behavior is found which transforms again into uniaxial behavior at higher temperatures. Both transitions generate pronounced secondary maxima in the temperature-dependent susceptibility.

One of today's most interesting topics in magnetism is ferromagnetism in thin films and multilayers. Due to recent developments in the field of epitaxy it becomes feasible to create materials in different forms of condensation, e.g., regarding their crystal structure and/or lattice constant.[1] Such tailoring of material parameters allows one to investigate the dependence of magnetism on distinct, well-characterized properties of ferromagnets. This tendency in experimental magnetism is accompanied by strong activities in theory.[2] Some of the theoretically modeled systems may be realized as thin films now. Thus first-principals calculations of magnetic properties may be compared with experimental findings giving a strong impetus to the understanding of magnetism in general.

The dependence of magnetization on particular directions is known as magnetic anisotropy, which includes crystalline (i.e., symmetry), strain as well as shape effects. Hence, structure and morphology determine the anisotropy. Because the selection of a substrate and/or growth conditions offers the possibility of manipulating the magnetic behavior, a huge number of papers are dealing with anisotropy behavior of thin films.[3–6] The major point with the studies on magnetic anisotropies is to figure out which structural properties are predominantly reflected in the magnetic behavior.[7–10]

A strong impetus on activities in thin-film magnetism is related to the concept of surface anisotropies proposed by Néel.[11] Néel pointed out the importance of the reduced symmetry at the surface of a ferromagnet. A new facet of Néel's approach of anisotropy has been proposed by Albrecht *et al.*[12] recently. The reduced symmetry at surface steps caused additional twofold contributions to surface anisotropy. The authors could extrapolate such step anisotropy from their results.[13] An elegant way to demonstrate the influence of steps on anisotropy is to study ferromagnetic films grown on vicinal surfaces.[14,15] Because of the step alignment and high step density on such surfaces, the influence of steps can be observed immediately as a strong twofold anisotropy. Recently it was demonstrated that first-principals calculations of electronic structure in fcc lattices are in good agreement with a Néel ansatz considering only nearest-neighbor interactions. Symmetry-based contributions to the anisotropy energy have been given.[16] Based on this, a complete Néel ansatz including all energy contributions for a stepped ultrathin ferromagnet has been worked out.[17] The rigorous description shows twofold volume contributions to the anisotropy of magnetocrystalline and magnetostrictive origin, which are due to the reduced symmetry in the films. Such magnetostrictive anisotropies have been proposed as the origin for the uniaxial anisotropy found in Co/Cu(1 1 13) films with thicknesses above three monolayers (ML).[18] Besides these contributions determined by the film volume, surface and particularly step anisotropies have to be expected. To minimize the influence of the volume contributions and to be more sensitive to surface and step anisotropies, we have investigated very thin films (about 2 ML). The dominance of surface and interface manifests in the strong dependence of the in-plane anisotropy found on temperature. The results presented here show in-plane anisotropy changes, which are driven by temperature-induced structural changes at the surface and interface. This behavior is completely different from previously published anisotropy changes, which were caused by transitions to bulk behavior due to increase of film thickness.[19–21] We would also like to stress that the effect is not due to the interplay of anisotropy and shape effects, which is responsible for magnetization flipping from vertical to in plane.[22]

The experiments including film preparation were performed under UHV conditions (base pressure $\leq 2\times10^{-10}$ Torr). The Cu(1 1 17) substrate has been prepared by cycles of Ar-ion etching (glancing incidence, 500 eV) and annealing (T > 670 °C). Low-energy electron diffraction (LEED) patterns of the annealed substrate have shown a splitting of the lattice spots, which are in good agreement with an average terrace width of 8.5 atomic distances. The films were grown at a rate of 1 ML/min with the substrate held at room temperature. During growth the pressure stayed below 5×10^{-10} Torr. As in case of Co/Cu(1 1 13),[14] no MEED-intensity oscillation could be found, while a high reflectivity was conserved during growth. We might conclude from the high reflectivity and the missing of intensity oscillations that the growth mode is preferably determined by step edge flow. No contamination could be found in the films within the detection limits of Auger electron spectroscopy. The magnetic characterization were performed *in situ* by means of the longitudinal magneto-optic Kerr effect. The setup is similar to the Kerr experiment of Bader and co-workers.[23]

Besides conventional Kerr-hysteresis measurement, the high sensitivity of our experiment allows us to measure ferromagnetic as well as paramagnetic susceptibilities. These measurements are made *in situ* and can be easily performed at films of a few monolayer thickness (≤ 2 ML).[24]

In the following a brief summary of the properties of Co/Cu(1 1 17) is given. A more detailed discussion of the magnetic properties of Co films grown on Cu(1 1 17) will be given elsewhere.[25] The vicinal Cu surface consists of terraces, which are separated by monatomic steps.[26] In the average the terraces on Cu(1 1 17) have a width of 8.5 atomic distances. The magnetic properties are mainly influenced by the parallel arrangement of the steps similar to Co/Cu(1 1 13).[27] In particular, that means that for all thicknesses studied (2 ML$< D <$ 14 ML) an in-plane magnetization is found with a uniaxial anisotropy behavior. At room temperature the easy axis of magnetization is as on Cu(1 1 13) parallel to the step edges.

An interesting issue in the ultrathin ferromagnets is the dependence of magnetic properties on temperature.[28] We have investigated the susceptibility[29] as a function of temperature (see Fig. 1). The susceptibility was measured parallel to the step edges. For the sake of simplicity the findings for cooling down from the paramagnetic temperature range is discussed first (Fig. 1, circles). The plot exhibits the well-known temperature behavior of susceptibility, i.e., a peak in the susceptibility at the Curie temperature. In the ferromagnetic regime the susceptibility signal is very low, which indicates that the film is in a single domain configuration, and the squareness of the hysteresis loop is very high. The "cooling" curve is reproducible in repeated heating and cooling cycles.

A completely different temperature dependence is found by heating up the as-grown films (see Fig. 1, squares). The susceptibility displays two strong, sharp peaks below the transition temperature. From the theory of susceptibility it is well known that in the ferromagnetic regime the susceptibility is inversely proportional to the magnetic anisotropy constant.[30] Hence, a peak in the susceptibility (below T_C) occurs each time the anisotropy becomes small.[31] In order to find out what causes vanishing anisotropies, we have taken hysteresis loops at different temperatures in a freshly prepared film (which happens to be slightly thicker). The loops obtained at different temperatures are shown in Fig. 2 . On the left-hand side the hysteresis loops obtained parallel to the step edges are shown. The right-hand side gives the magnetization curves perpendicular to the step edges within the film plane. Figures 2(a) and 2(b) are the hysteresis loops obtained far below the temperature of the first maximum, T_1. The loops indicate a strong uniaxial behavior with easy axis parallel to the step edges (notice the different scales of the abscissa). Above T_1 the magnetization curves have drastically changed [Fig. 2(c) and (d)]. The film exhibits nearly a fourfold symmetry with a very small uniaxial contribution, which, however, favors the direction perpendicular to the step edges. Thus, it is obvious that first, a transition of the anisotropy is found and second, the uniaxial anisotropy constant must have changed its sign. Both effects can produce a strong peak in the susceptibility if higher-order anisotropy contributions are negligibly small in the transition. Crossing the temperature of the second susceptibility peak, T_2, changes the hysteresis loops again [see Fig. 2(e) and (f)]. The twofold anisotropy is reestablished with the same easy axis of magnetization as below T_1. Hence the second peak in the ac susceptibility is caused by another anisotropy change in the film. In summary, two different changes of the in-plane magnetic anisotropy are found at temperatures slightly below and above 110 °C.

Next it is important to discuss the irreversible behavior during the first heat treatment. With a fresh film the temperature was raised to different values characterized by the anisotropy changes discussed above. Figure

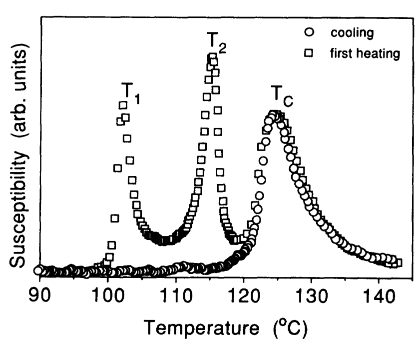

FIG. 1. Susceptibility of Co/Cu(1 1 17) ($D \approx 2.2$ ML) measured along the steps as a function of temperature. Squares are the susceptibility of an as-grown film during first heating. Circles indicate the susceptibility during successive cooling.

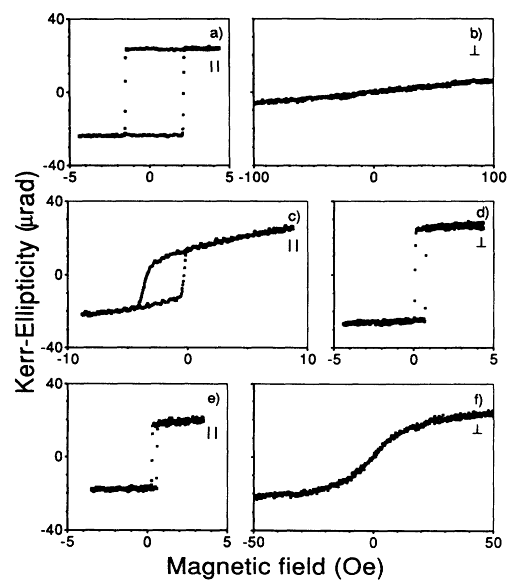

FIG. 2. Hysteresis curves of Co/Cu(1 1 17) ($D \approx 2.5$ ML) obtained with the longitudinal magneto-optic Kerr-effect at different temperatures (a/b : $T < T_1$; c/d : $T_1 < T < T_2$; e/f : $T_2 < T < T_C$). The loops on the left-hand side are measured along the steps, while the loops on the right-hand side are measured perpendicular to the steps.

3 shows the temperature behavior obtained by increasing the temperature slightly above the first susceptibility maximum and cooling down again. As one can see (Fig. 3) a new maximum appears at a lower temperature T_1'. The magnetization curves identify this peak as the transition temperature between the uniaxial and nearly biaxial anisotropy behavior. The transition is the same as found in the first heating cycle at T_1 shifted to lower temperatures now. That transition turns out to be reversible. As long as the temperature is not raised too high ($< 80\,^\circ$C) the transition was observed on heating and cooling repeatedly.[32]

Raising the temperature to T_2, however, causes a complete irreversible change of film properties. After reaching T_2, a temperature range with fourfold anisotropy behavior cannot be found any longer. The cooling down curve exhibits the same shape as already seen in Fig. 1. Thus the irreversible change of the magnetic properties appears below the Curie temperature. A stable film configuration is obtained, which is not changed on further heating. No interdiffusion of Co and Cu could be found with Auger electron spectroscopy below 200 °C.[25]

A hint of what determines the film properties in the 2.2-ML film can be implied from the findings obtained with Co/Cu(1 1 13), i.e., the results from Brillouin light scattering (BLS) and Kerr experiments.[18,27] From the BLS results one can extract an interface anisotropy, which favors magnetization perpendicular to the step edges. As the films in the BLS measurements were covered with Cu, this anisotropy has to be attributed to properties of the two Co/Cu interfaces. The BLS data show that the interface anisotropy is dominant below a critical thickness of (2.9 ± 0.6) ML, which means that the easy axis is perpendicular to the step edges below that thickness. With Kerr experiments it was demonstrated, however, that the films with an uncovered surface behave differently.[27] Two monolayers Co/Cu(1 1 13), for example, show an easy axis that is parallel to the step edges in contradiction to the findings for the covered film. Thus, one might infer that the uncovered surface (free surface) gives a stronger anisotropy contribution than the interface, which yields the resultant easy axis parallel to the step edges.

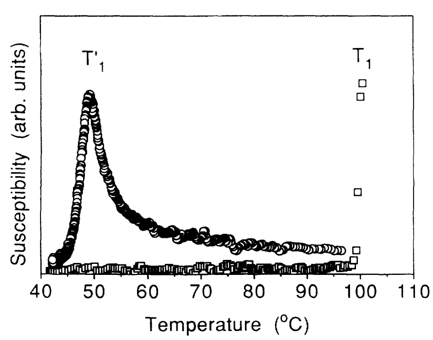

FIG. 3. Susceptibility of Co/Cu(1 1 17) ($D \approx 2.2$ ML) measured along the steps. Squares are the susceptibility of an as-grown film during first heating up to T_1. Circles indicate the susceptibility during successive cooling down from T_1.

In Co/Cu(1 1 17) an easy axis parallel to the step edges is found for uncovered films too [see Fig. 2(a) and (b)], which indicates the analogous behavior of Co/Cu(1 1 13) and Co/Cu(1 1 17). If one postulates the two competing anisotropies for Co/Cu(1 1 17) too, the temperature behavior below 3 ML can be explained. At room temperature it is obvious that the surface anisotropy overcomes the interface anisotropy, yielding the easy axis of magnetization parallel to the step edges. With rising temperature, however, the interplay of both anisotropies changes. The surface contribution becomes weaker until the contributions are nearly the same (at T_1) giving the nearly fourfold anisotropy. The slight twofold contribution (perpendicular to the step edges) supports the suggestion of competing interface and surface anisotropies. On further heating the surface contribution gains importance over interface anisotropies again.

Hence, the remaining question is that for the mechanism which could be responsible for the strong dependence of the surface and interface anisotropy on temperature variations. Actually the temperature behavior itself gives a hint to the mechanism. In the same temperature range a roughening transition is found on vicinal Cu surfaces.[33] With scanning-tunneling-microscopy studies on Cu(1 1 19), it was demonstrated that step roughness increases strongly when raising temperatures to 100 °C.[34,35] When the steps get rougher the step alignment is disturbed on the microscopic scale due to increased number (and mobility) of kinks, while the macroscopic surface orientation is still preserved. The temperature dependence of magnetic anisotropy might be directly related to similar dynamical properties of steps on the surface and interface of the films. One can imagine the following scenario during heating: The steps at the surface might get rough first because surface diffusion usually has lower activation energies than bulk diffusion, leading to the first transition. The interface contribution should change remarkably at higher temperatures, which would yield the second anisotropic change. The irreversible behavior can also be explained within this microscopic model. The interface structure produced at high temperatures cannot be reversed on cooling for thermodynamical reasons. Thus, no changes in anisotropy behavior can be found on cooling. The "frozen" roughness at the interface, on the other hand, might prevent the "perfect" realignment of steps at the surface causing lower uniaxial magnetic anisotropy.

In conclusion, in-plane anisotropy changes have been found in two monolayer films of Co on Cu(1 1 17). In a narrow temperature interval the easy axis of magnetization switches twice within the film plane. The susceptibility demonstrates that the alterations are accomplished by vanishing anisotropies. The irreversible character indicates that structural changes are most likely responsible for that magnetic behavior. As the interface and surface determine the overall anisotropy in the very thin films, studied, a model has been proposed, which links the magnetic temperature effects with the step dynamics at the surface and interface.

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