Symmetry-Induced Uniaxial Anisotropy in Ultrathin Epitaxial Cobalt Films Grown on Cu(1 1 13)

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(Received 3 September 1991)

Uniaxial anisotropy has been found in ultrathin cobalt films grown on a Cu(1 1 13) surface. Our studies using scanning electron microscopy with polarization analysis clearly show that the easy axis of magnetization is parallel to the direction of the step edges of the Cu(1 1 13) substrate. In spite of the different anisotropy behavior, the domain structures in Co/Cu(001) and Co/Cu(1 1 13) are similar, which indicates that the domain pattern in ultrathin films is little affected by the anisotropy.

PACS numbers: 75.30.Gw, 75.60.Ch, 75.70.Ak

Recent scientific and technical advances in surface science and thin-film preparation methods have opened up a new class of research activities, i.e., the investigation of new artificially grown materials. One fascinating aspect of such studies is the ability to compare properties of the same material in different forms. Different crystal structures can be stabilized by the appropriate choice of substrates, phases that otherwise do not occur in nature such as bcc Co [1], fcc Co [2], fcc Fe [3], and bcc Cu [4]. A further item of interest with these artificial materials concentrates on the exploitation of the transition from three- to two-dimensional crystals. For that purpose, the investigation of ultrathin films, i.e., films of a few monolayer thickness, has gained more and more importance in recent times. Apart from the general interest in studying dimensionality effects, there is a profound interest in understanding ultrathin films from the technological point of view, as novel devices continue to shrink in size.

Investigations of magnetism in ultrathin films impressively demonstrate the variety of effects which can be found in ferromagnets of diminishing thickness. In many cases the effects manifest in the magnetic anisotropy of the films [5]. For the interpretation and understanding of the magnetic properties it turns out that it is of great importance to distinguish the purely magnetic properties from those induced via magnetoelastic interactions by the film/substrate interface [6]. One approach to solve that problem is to study the ultrathin-film magnetism in a film system with a perfect and ideal substrate/film interface. The system Co/Cu(001) is well known from the literature to fulfill the above condition, and is well characterized concerning growth as well as magnetic properties [2,7–9]. The system exhibits layer-by-layer growth. The anisotropy behavior is determined by the film symmetry; no interface (i.e., magnetoelastic) effects altering these symmetry properties have been found for the cobalt films. Thus Co/Cu(001) is the ideal reference system for the investigations of substrate-induced magnetic film properties. These considerations let us use a slightly different template with a well-characterized and defined modification of the Cu(001) surface, i.e., the Cu(1 1 13). The main difference between Cu(001) and Cu(1 1 13) is the reduced symmetry of the Cu(1 1 13) surface due to the existence of well-oriented steps. The influence of the symmetry on the magnetic properties of the epitaxial cobalt films is the issue of this paper. One item of the paper deals with the correlation of the magnetic anisotropy with the film symmetry. The second topic is related to the micromagnetic structure in the Co/Cu(1 1 13) films. The influence of the magnetic anisotropy and/or the substrate topography on the domain structure in ultrathin films is addressed. This item is important for the understanding of the micromagnetics in ultrathin films, particularly in view of the complex domain structure found in Co/Cu(001) films [10].

(1 1 13) surfaces are vicinal to (001) surfaces. The orientation deviates from the low-index plane by a tilt angle of 6.2°, towards the [110] direction. The Cu(1 1 13) surface has been investigated by means of a scanning tunneling microscope quite recently [11]. The (001) terraces have been found to be separated by monatomic steps with the steps running essentially along {110} (see Fig. 1). The terrace width distribution, centered around a width of 6.5 atom distances, is rather broad. From the measured beam splitting of the LEED diffraction pattern in our study we deduce a mean terrace length of 7 ± 1

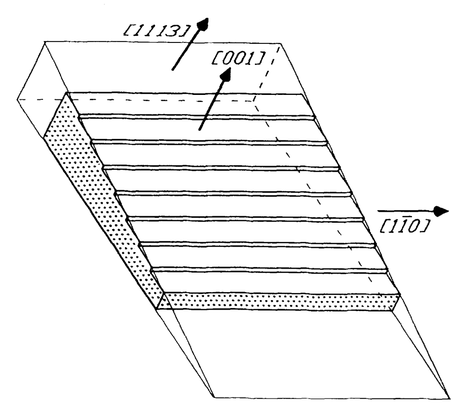

FIG. 1. Sketch of the Cu(1 1 13) surface. The step edges are parallel to the {110} direction which lies within the (1 1 13) surface.

atoms. The splitting indicates that the step edge direction is parallel to {110}, consistent with [11].

From symmetry considerations one might interpret the Cu(1 1 13) surface as the superposition of a Cu(001) lattice with a one-dimensional superlattice with lattice constant of ≃6.5 atoms (on the average). Thus a reduced twofold symmetry characterizes the Cu(1 1 13).

In view of the fact that Cu(1 1 13) consists dominantly of (001) terraces it is not surprising to obtain qualitatively the same growing behavior, i.e., epitaxial layer mode growth, for cobalt on (1 1 13) and (001). The films were grown at room temperature with a deposition rate of 2 monolayers (ML) per minute. By means of medium-energy electron diffraction (MEED) intensity oscillations we could follow the layer-by-layer growth of the Co films. For Co/Cu(1 1 13), strongly damped oscillations have been found, while the MEED intensity maintained a high level during the growth up to 10 layers. The intensity never dropped below 75% of the initial level which indicates an even better layer mode growth on the stepped surfaces than on Cu(001), most probably due to a predominant step edge growth. The incident beam was directed along the [1$\bar{1}$0] direction. The LEED diffraction pattern after cobalt deposition exhibited the same structure as the clean Cu(1 1 13) (within the limits of our instrument) although the splitting, which has been found on Cu(1 1 13), was smeared out due to some surface roughening. Hence the whole film is characterized by a reduced symmetry (twofold), and a new artificial solid has been produced and stabilized. Novel properties of such films can be expected and are indeed found with the magnetic anisotropy of the Co films.

By means of the scanning electron microscope with spin polarization analysis of the secondary electrons (SEMPA) [12] we have studied the domain structure in 5-ML Co films on Cu(1 1 13). As a result of the vector analysis of the polarization measurements, the orientation of the spontaneous magnetization of the domains can be determined and thus the easy axes of magnetization are directly established with the domain structure investigation. The main findings of our studies are summarized in the following and are compared with the findings obtained with Co films grown on Cu(100) [13].

(i) With Co/Cu(001) as well as Co/Cu(1 1 13) we find the magnetization to lie within the film plane. No perpendicular component within the limits of our experiment could be found (uncertainty $\leq 10°$).

(ii) Completely different properties of the Co films on Cu(1 1 13) and Cu(001) have been found with regards to their easy axes of magnetization. Whereas with the films grown on Cu(001) a fourfold anisotropy has been found with {110} as easy axes [10], a uniaxial anisotropy determines the Co/Cu(1 1 13) films. The easy axes of magnetization are parallel to the step edges of the Cu(1 1 13) template. We have tried to force a remanent magnetization orientation of domains perpendicular to the step edges. Several magnetization as well as demagnetization procedures have been tried to create domains with M_s perpendicular to the step edges. Although the external magnetic field was sufficiently high to saturate the films perpendicular to the steps, the domain pattern after removal of the field exhibit only domains magnetized parallel to the edge orientation.

(iii) In spite of the completely different anisotropies of the cobalt films on Cu(001) and Cu(1 1 13) their domain structures exhibit amazing similarities. Figure 2(a) shows the domain pattern in a 5-ML Co/Cu(1 1 13) film, and Fig. 2(b) shows the domain structure in a 9-ML Co/Cu(001) film. The similar, irregular shape of the domains directly demonstrates that the symmetry of the domain structure in ultrathin films is not determined by the anisotropy of the films, if the easy axes of magnetization lie in the film plane. Thus the micromagnetic behavior of ultrathin films is completely different from that at the surface of bulk magnets and of thick-film magnets. In ultrathin films the wall orientation is no longer deter-

(a)

(b)

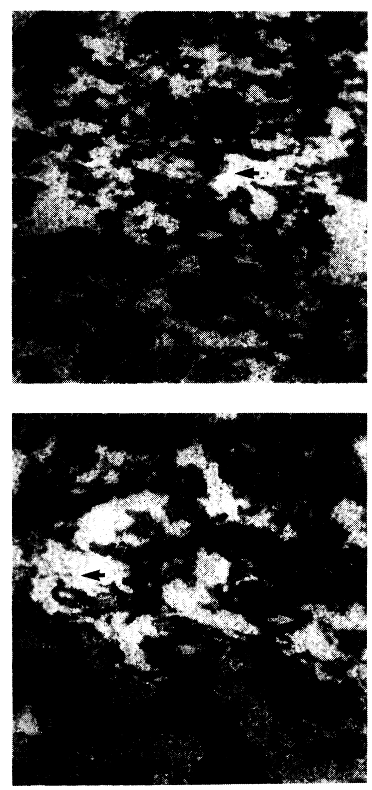

FIG. 2. Domain structure in cobalt films. Arrows indicate the magnetization orientation within the domains. (a) A 5-monolayer-thick film grown on Cu(1 1 13). The size of the image is 1×1 mm^2. (b) A 9-monolayer film grown on Cu(001). The size of the image is 500×500 μm^2. Note the existence of three gray levels (white, gray, black). The gray domains consist of two different types of domains, magnetized upward and downward, which has been proven by the second polarization component. Those domains, however, are not indicated in the image by arrows.

mined by the orientation of the magnetization in the adjacent domains. As a result of the vanishing thickness and thus the negligible extension of the wall in the third dimension (i.e., depth) the aligning force on the wall becomes very weak. The long-range magnetostatic interactions of magnetic fields created within the walls due to their Néel-like structure gain importance in the ultrathin films and influence the domain pattern [13].

(iv) No correlation of the domain shape or wall orientation with the preferred step orientation of the Cu(1 1 13) template is observed [see Fig. 2(a)]. This result suggests that pinning of domain walls at steps may not be the dominant mechanism responsible for the symmetry properties of the domain structure in ultrathin films.

The uniaxial anisotropy of the films on Cu(1 1 13) opens one question which will be addressed in the following section: What is the origin of the uniaxial anisotropy and can the uniaxial anisotropy be explained in the framework of purely bulk cubic properties, generally described by the anisotropy constants K_1 and K_2 [14]? To clarify that point the influence of the K_2 term of the orientational free energy has to be discussed in detail for the (1 1 13) surface. It is well known that {110} directions become the easy axes of magnetization for $K_2 > -\frac{9}{4}K_1$ ($K_1<0$) [15], whereas otherwise the {111} directions are the easy axes of magnetization. As the (1 1 13) surface contains one {110} direction, i.e., the direction parallel to the steps, the balance between the K_1 and K_2 terms has to be taken into consideration. A straightforward evaluation of the orientational free energy based on bulk symmetries of a cubic material yields the following items for the (1 1 13) surface ($K_1<0$ [16,17]). First, the influence of the K_2 free-energy contribution is very small compared to the K_1 energy term. The ratio of the prefactors of the K_1 and K_2 energy terms is 100:1. That result is not surprising as the deviation of the (1 1 13) from the (001) surface is only small and the K_2 term equals zero in the (001) orientation. For the same reason—the similarity of the surfaces—the second conclusion from the free-energy equation is obvious. The angle dependence of the orientational free energy for (1 1 13) is similar to that of the (001) surface. Two marked energy minima parallel and perpendicular to the steps are found, which correspond to the equivalent energy minima along {110} for the (001) surface. Third, the absolute energy minimum depends on the K_2 value as mentioned above. For $K_2 \simeq -3K_1$, the minima parallel and perpendicular to the steps are equal. For $K_2 > -3K_1$, the absolute energy minimum coincides with the direction parallel to the step edges, whereas for smaller K_2 values the direction perpendicular to the steps edges is favored. The relative differences for reasonable K_2 values, however, are very small compared to the absolute energy minimum, e.g., varying K_2 from $2K_1$ to $-8K_1$ gives a 10% increase of the energy in the direction perpendicular to the steps. Thus for the (1 1 13) surface the cubic ansatz of the orientational free energy yields pronounced minima perpendicular and parallel to the step directions. More than that, as negative K_2 values have been found with cobalt films grown on Cu(011) [17], we have to expect from the cubic ansatz the absolute free-energy minimum in the direction perpendicular to the step edges. Hence, domains magnetized perpendicular to the step edges should obviously be expected. It should even be possible to saturate the film in that particular direction.

To test that prediction we performed the following experiment. First of all the film was saturated parallel to the step edges. The domain structure investigation by means of SEMPA showed that the film was in a single-domain state. Even at the sample edges we could not find any domains. We then tried to saturate the single-domain film with the in-plane direction perpendicular to the steps. Again we studied the domain structure and found domains in the film. The domain magnetization, however, was again aligned with the [1$\bar{1}$0] or [$\bar{1}$10] direction (parallel to the steps). This finding demonstrates first that the magnetic field was strong enough to rotate the magnetization from the orientation parallel to the steps into the perpendicular direction. Second, it clearly proves that the direction perpendicular to the steps is neither an axis of easy magnetization nor a direction of a local minimum of the orientational free energy, as proposed from the calculations based on the cubic ansatz. Thus we may conclude that the observed uniaxial anisotropy cannot be described by the semiclassical approach via cubic bulk anisotropies.

The above considerations show that a uniaxial anisotropy term has to be introduced in the orientational free energy for the Co/Cu(1 1 13) films in contradiction to the Co/Cu(001) films. With that result we finally end up with the question of what mechanism is responsible for the uniaxial anisotropy. Two different explanations come to mind. The first mechanism is related to the stress in the films. For Co/Cu(001) a tetragonal distortion is found in the films [7]. That tetragonal distortion may be altered in the vicinity of the steps due to the different geometrical arrangement of the substrate atoms at the steps. Consequently, stresses of twofold, no longer fourfold, symmetry are introduced at the steps and may influence the geometrical arrangement of the cobalt atoms on the terraces, which results in a complete twofold-symmetrical distortion in the films. The second explanation of the uniaxial anisotropy is based on the electronic structure of the film and thus on the crystalline anisotropy. As the whole film is characterized by a reduced symmetry one might expect the electronic structure to be altered, which should change the crystalline anisotropy via the spin-orbit coupling. Strictly speaking, the latter mechanism would mean that a new artificial material has been stabilized on the Cu(1 1 13) surface. The two proposed mechanisms differ in so far as the first is directly related to the steps and has to be taken under consideration as a localized effect whenever steps do exist, whereas the second mechanism gains importance when a superlattice exists with an average terrace width on the length scale of atomic distances.

In conclusion, we may summarize as follows: Induced by the periodical step arrangement of the substrate, new magnetic properties have been created in ultrathin cobalt films. In comparison with Co/Cu(001) we have found a change of the anisotropy in Co/Cu(1 1 13), although both templates are very similar. The anisotropy reflects the reduced symmetry of the cobalt films grown on Cu(1 1 13). In spite of that dramatic change of the anisotropy, the domain patterns in both films are characterized by the same irregular shape of the domains. That result gives a hint for the understanding of the micromagnetics in ultrathin films. It demonstrates that the influence of the anisotropy on the domain structure is of minor importance in ultrathin films in strict contradiction to the experience with bulk and thick-film magnets.

The authors would like to thank Professor H. Ibach for critical reading of the manuscript and K. Hieble for his able technical assistance.

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[14] The cubic ansatz for the orientational free energy is given by $E_a = \text{const} + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_1^2\alpha_3^2) + K_2(\alpha_1^2\alpha_2^2\alpha_3^2) + E_f + E_g$, with E_f the shape and E_g the surface anisotropy energy. Both are constant for an in-plane magnetization. The α's represent the directional cosines of the magnetization with respect to the cubic axes.

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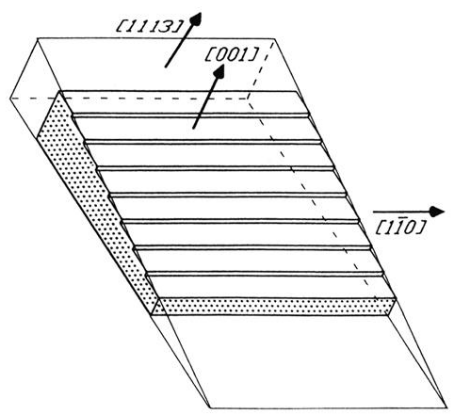

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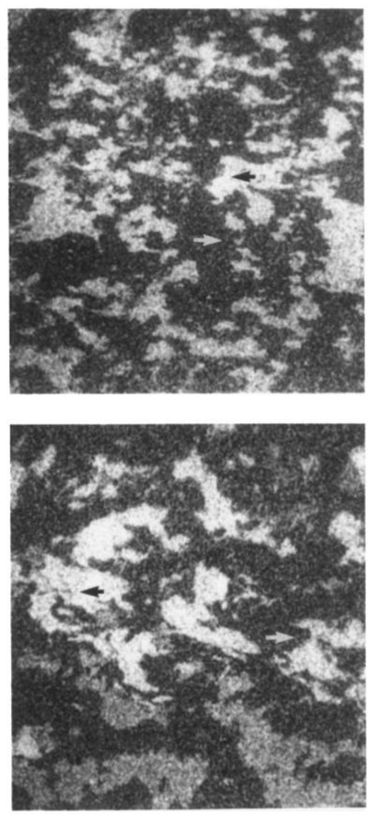
(a)

(b)



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