Magnetic Domains in Thin Epitaxial Co/Au(111) Films

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The formation of magnetic domains in thin epitaxial Co/Au(111) films is investigated. Three-monolayer films are shown to decay into out-of-plane domains of micron size. We find a smooth transition from out-of-plane to in-plane magnetization at a crossover thickness of 4.5 layers and determine that the domain size depends linearly on film thickness below crossover.

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The experimental progress in growing epitaxial magnetic thin films down to monolayer thickness has revealed a wealth of unusual phenomena. Most remarkably, ferromagnetism has been shown to occur in very thin layers[1] or even truly two-dimensional systems.[2] The theoretical prediction that thin magnetic films exhibit large magnetic anisotropies perpendicular to the surface[3] has been substantiated in a large variety of systems.[1] The prospects of out-of-plane magnetization are especially attractive for data-storage devices. In particular, thin epitaxial cobalt films[1,4-9] are beginning to play a prominent role in view of potential future applications. This Letter studies the formation and growth of magnetic domains in epitaxially grown Co/Au(111) layers. Domains are shown to be formed in three-monolayer films, and the switching behavior from perpendicular to parallel magnetization is characterized at a domain-size level.

The principle of the experiment is known as spin scanning electron microscopy (SEM) or SEM with polarization analysis and has been described previously.[10,11] It essentially consists of scanning a highly focused primary electron beam along a ferromagnetic sample, thereby producing secondary electrons whose spin polarization is analyzed. In our setup the primary electrons are generated by a field-emission gun of variable energy (0.5 to 25 keV). The secondary electrons are collected by an energy-prefiltering transfer optic and are then energy analyzed in a cylindrical mirror analyzer. Subsequently, the low-energy secondary electrons (0 to 10 eV) are fed into a Mott spin detector,[12] which is capable of simultaneously measuring the out-of-plane and one in-plane polarization direction.

The thin epitaxial cobalt films were grown *in situ* onto a Au(111) single crystal by slow evaporation from a tantalum crucible heated by electron bombardment at a growth rate of 0.2 Å/min. The pressure during evaporation was less than 5×10^{-10} mbar. Film thicknesses have been determined independently by Auger spectroscopy[13] and stylus profilometry for thicker films, giving agreement to within 10%. The growth characteristics had been investigated in detail earlier:[4,6] The interface is sharp on the atomic level; no interdiffusion is known to occur. The large lattice mismatch between Au(111) and Co(0001) of 14% causes stress at the interface which is relieved by misfit dislocations. We monitored the quality of our films by Auger spectroscopy and low-energy electron diffraction (LEED). The sharp spots of the Au(111) substrate broaden for the first two monolayers (1 ML = 2 Å), and then sharpen again to finally produce the well-known sixfold symmetry characteristic of hexagonal Co(0001). From LEED, we find a tensile strain in the surface plane of $\simeq$14% for 1 ML and $\simeq$8% for 3 ML compared to the Co bulk value, which is achieved at 6 ML; these values are in accordance with those found earlier.[14] Both evaporation and measurements have been performed at 300 K.

Figure 1 depicts the magnetic domain image for a 3-ML Co/Au(111) film, showing irregularly shaped domains with magnetization vectors along the surface normal. The simultaneously measured in-plane magnetization vanishes. On a micron scale, this shows the importance of a high uniaxial anisotropy for the magnetization direction in thin films.[1,4-9] The most remarkable fact and new finding of Fig. 1 is not this presence of a vertical magnetization, but its decay into a domain structure. In fact, classical theories[15] predict a single domain for thin films. Experimentally, single-domain behavior has been indirectly inferred from hysteresis loops for various systems,[2,16,17] and also directly observed for Co/Cu(100)

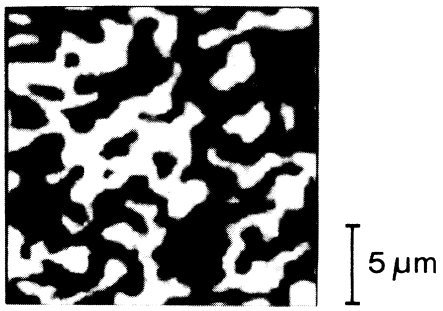

FIG. 1. Magnetic domain structure of a 3-ML Co/Au(111) film, showing out-of-plane domains with magnetization up (white) and down (black). The corresponding in-plane magnetization vanishes. Primary energy, 5 keV; beam current, 0.1 nA. The average measured spin polarization is +15% and −15%, respectively.

films magnetized in plane.[18] In contrast to these systems, we always find micron-size domains in our out-of-plane magnetized films.

A point of interest is whether this advent of domains is the true "ground state" of the film or rather some metastable state. The concept of a lowest-energy state is somewhat obscured owing to the inherent hysteretic properties of ferromagnets. It is related to the demagnetization procedure of the sample with respect to temperature and/or applied magnetic field. We note that all films have been evaporated in a field-free configuration ($H < 1$ A/m), giving demagnetized films. Domain imaging itself has been performed in a residual magnetic field caused by the focusing optics of the electron microscope of < 250 A/m, much smaller than any relevant field for the system under study.[19]

Substrate or film imperfections are known to serve as pinning centers for domain walls. Interestingly, in our films we found that the domain size is much larger than the average step width of the substrate of $\simeq 50$ nm.[20] Defects in the film can cause local alterations of the preferred magnetization axis. We note, however, that at extended irregularities visible in the topography we do not observe any variation; in particular, we do not find any in-plane magnetization component. We state therefore that most surface imperfections do not influence domain formation.

The lowest-energy configuration of a ferromagnetic sample can be expected to be reached by annealing above the Curie temperature T_C and subsequent cooling in zero field. Upon annealing, we find that the location of the domains changes irregularly. In no case were we able to achieve a single-domain state. Hence we conclude that at room temperature the lowest-energy state of a 3-ML Co/Au(111) film is an irregular arrangement of vertical domains, with a typical domain size of microns. We note that this result can explain why hysteresis loops in very thin Au/Co/Au layers display a strongly reduced remanent magnetization compared to saturation.[19]

A recent theoretical model predicts domains even in ML films, based on an elaborate calculation including dipolar magnetic energy.[21] As a starting point we compare this zero-temperature calculation to our room-temperature data, bearing in mind that finite temperature even favors the creation of domains. The relevant quantities are $\delta = w/D$ (w denotes domain-wall width and D domain size) and f, which is proportional to the ratio of uniaxial anisotropy to demagnetization energy. For domains to be present, $1 \lesssim f \lesssim 1.4$. For our 3-ML film, we find $\delta < 0.02$, $D \simeq 2$ μm, and hence[21] $f \simeq 1.06$. Therefore finite domains must form. To our knowledge, Fig. 1 represents the first experimental support for the prediction that domains should form in very thin films with large perpendicular anisotropy, owing to the small dipolar magnetic energy. Much more work is required to test the implications of this novel view of regarding domain formation as being driven by dipolar energy.

The next point we address deals with the switching behavior from out-of-plane to in-plane magnetization at a certain thickness. Any film magnetized vertically must change its magnetization direction at a critical thickness d_c, owing to the predominance of magnetostatic energy over anisotropy. This switching has been investigated earlier with hysteresis-loop experiments.[4,22] The result is that near d_c, the out-of-plane remanent magnetization decreases, whereas the in-plane magnetization increases. Depending on the film parameters, d_c varies between 0.5 and 1.9 nm.[4,5,8] The exact mechanism of such a transition, however, is not clear. The crucial question is whether the out-of-plane remanence decrease is due to a breakup into domains or rather to a rotation of the magnetization from out of plane towards in plane. We emphasize that from hysteresis loops one cannot discriminate between a continuous rotation of the magnetization and only two allowed discrete values. Only the monitoring of the magnetization direction in a domain allowed us to conclude unequivocally that the magnetization crossover takes place by a continuous rotation from 0° to 90°, within a thickness increase of 2 ML.

The experiment to find this result is quite elaborate. It involves evaporation of a thin film whose thickness is subsequently increased in well-defined intervals. At each thickness, the *identical* position on the sample has to be probed. The results of such an experiment are displayed in Fig. 2 for a selected thickness range 3 ML $\leq d \leq$ 6 ML, each with the out-of-plane and one in-plane components. Below 2 ML we do not find magnetization, in accordance with the observation that $T_C < 300$ K.[4] At 2

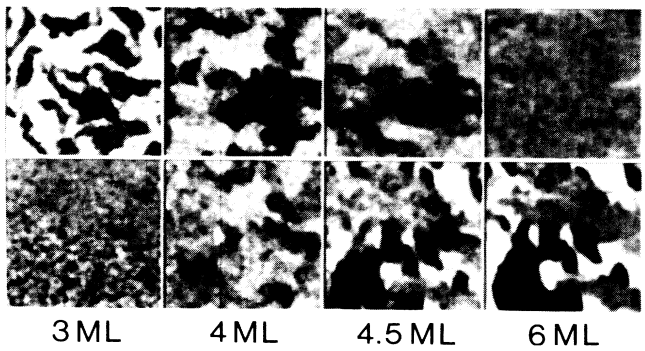

FIG. 2. Magnetic domain images for Co/Au(111) thin films, showing the evolution of domain size and switching behavior vs film thickness. The pictures have been taken at identical positions on the sample (within 2% of scan area) between evaporation of additional layers. For each thickness, the upper image gives the out-of-plane and the lower the in-plane magnetization component (along the length of the page). Gray scales from black to white indicate magnitude of magnetization component along the measured axis; scan area 20 μm × 20 μm. Note the collapse of the small domains on proceeding from 3 to 4 ML.

ML, very small domains (≤ 0.5 μm) with perpendicular magnetization begin to form (not shown). At 3 ML, an out-of-plane domain pattern similar to Fig. 1 is observed. One additional layer is sufficient to let the small domains coalesce into larger domains, and some faint contrast is already visible in plane. Evaporation of an additional 0.5 ML corresponds to the crossover thickness d_c. Another 0.5 ML forces the film to essentially switch to in plane (not shown). For $d \geq 6$ ML, no perpendicular component is left.

From Fig. 2 and the second in-plane component we are able to reconstruct the angle θ between surface normal and magnetization for all points individually. The result is summarized in Fig. 3. We clearly find a continuous switching from out-of-plane to in-plane magnetization in a finite thickness interval of 2 ML.[23] According to a recent calculation,[24] the magnetization axis might rotate or jump into the plane at a finite small temperature $\ll T_C$. However, this is applicable only far from T_C. In the following we outline how the tilted configuration can be understood by a simple energy consideration. The reason is the different angular dependence of the demagnetization energy and second hexagonal anisotropy constant K_2. The total energy

$$E = \tfrac{1}{2}\mu_0 M^2 \cos^2\theta + K_1 \sin^2\theta + K_2 \sin^4\theta,$$

with M the magnetization and K_1, K_2 the uniaxial anisotropy constants, is minimized with respect to θ. Depending on the actual numbers, the solutions are $\theta = 0°$, $\theta = 90°$, and $\sin^2\theta = (\frac{1}{2}\mu_0 M^2 - K_1)/2K_2$. Since K_1 may be split into bulk and surface contributions, $K_1 = K_{1B} + K_S/d$,[22,25] we can determine the uniaxial surface or interface anisotropy K_S from a fit to Fig. 3. Taking Co bulk values $M = 1440$ kA/m, $K_{1B} = 412$ kJ/m^3, and $K_2 = 143$ kJ/m^3, the result is $K_S = 0.62 \pm 0.05$ mJ/m^2, in good agreement with the 0.53 mJ/m^2 found earlier.[2]

We emphasize that the phenomenological quantity K_S cannot give insight into the physical origin of the uniaxial anisotropy. The present understanding is that strain might be responsible for the out-of-plane anisotropy, although the Néel surface anisotropy cannot be excluded.[8,9] Experiments with unstrained hexagonal Co films will solve this ambivalence.

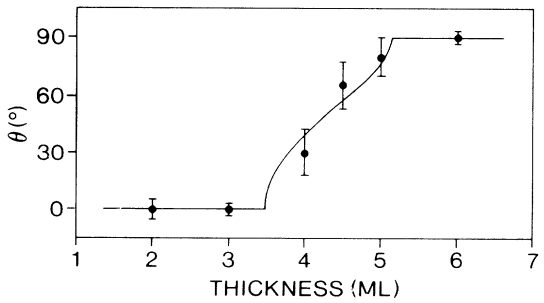

FIG. 3. Angular dependence of the magnetization direction vs thickness derived from point-by-point analysis of Fig. 2. θ is the angle between surface normal and magnetization. The line represents the best fit for $\sin^2\theta = (\frac{1}{2}\mu_0 M^2 - K_{1B} - K_S/d)/2K_2$, with the Co bulk values $M = 1440$ kA/m, $K_{1B} = 412$ kJ/m^3, $K_2 = 143$ kJ/m^3, and the fit parameter $K_S = 0.62$ mJ/m^2.

In the following we focus on the average domain size. From Fig. 2 we see that the domain size D grows with increasing film thickness d, mainly by eliminating intermediate domains and walls. A domain growth with increasing film thickness was predicted long ago,[15] with $D \sim \sqrt{d}$. However, the approximations in this model fail for $D > d$. The statistical evaluation of various films gives a linear increase from $D \simeq 0.5$ μm at 2 ML up to $D \simeq 4$ μm at d_c, and only a slow further increase for thicker films. For films with $d \geq 10$ ML, we find elongated domains, indicating an anisotropy within the film plane. In contrast to films with $d \leq 3$ ML, films at d_c can be made single domain at remanence by magnetizing in a sufficiently high magnetic field.[19] Actually, we find square hysteresis loops for films with $d = 8$ ML with the *in situ* magneto-optical Kerr effect, and a coercive field $H_C = 12$ kA/m. We verified the single-domain state over mm areas. Thus the 3-ML film ground-state domain configuration is responsible for the *occurrence* of domains in films with $d > 3$ ML, whereas the *size* of the domains depends on d.

In contrast to the domain size which also depends on the magnetic history of the sample, the domain-wall width depends on film parameters exclusively (exchange and anisotropy constants, crystallographic orientation, and film thickness). One can therefore expect to gain additional insight into these properties in thin films by measuring the domain-wall width w versus film thickness d. Furthermore, the theory for domain formation[21] predicts for perpendicularly magnetized films that w decreases as d increases. Preliminary experiments on an in-plane magnetized film of 50-ML thickness show that Néel walls[10,26] exist with $w = 40$ nm, indicating that our probing diameter is below this value. The walls in out-of-plane magnetized samples are even thinner. Experiments are under way to resolve these narrow walls also.

In conclusion, we have investigated magnetic domains in thin epitaxial Co/Au(111) films. We find out-of-plane domains of micron size in films as thin as 3 ML. The transition from out-of-plane to in-plane magnetization is shown to occur smoothly within a narrow thickness region of 2 ML. The domain size depends linearly on film thickness. At room temperature, a multidomain configuration is the ground state for a 3-ML film, whereas for films with $d \geq d_c$, the single-domain state can be reached by magnetizing in an external field.

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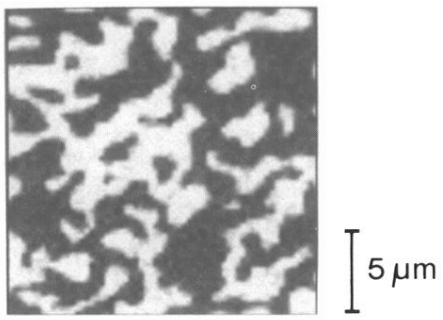

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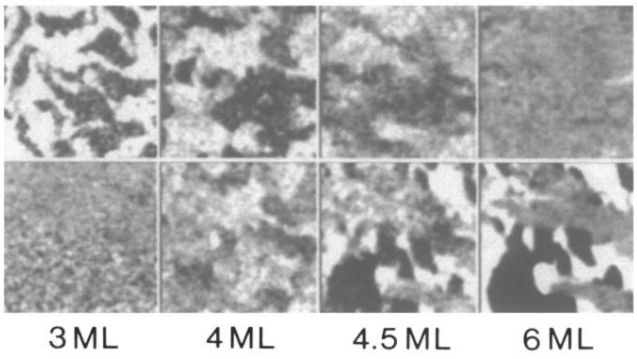

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