Epitaxial growth of thin magnetic cobalt films on Au(111) studied by scanning tunneling microscopy

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The initial nucleation and the subsequent growth of Co on Au(111) have been studied with scanning tunneling microscopy. At low coverage the cobalt nucleates forming polygonal islands whose location and spacing are determined by the underlying Au(111) zigzag reconstruction geometry. The islands are two atomic layers high and grow laterally with increasing coverage. A model for the nucleation and growth is proposed and a comparison with the growth of other metals on gold is given. We also qualitatively relate our structural results to published magnetic measurements.

Epitaxially grown magnetic thin films have revealed a variety of interesting physical phenomena. Most remarkable are the realization of a ferromagnetic monolayer[1] and the perpendicular magnetic anisotropy observed in some thin films.[2] Initially, the focus has been on the new magnetic properties of these films, while the details of their structure have not been fully investigated. In this paper we report the results of a scanning-tunneling-microscopy (STM) study of the growth of Co on the Au(111) surface. This system is of particular interest for, in addition to its role in the study of perpendicular magnetization in thin magnetic films, it is also a promising candidate for magneto-optical storage technology.[3,4]

Since the observation of a perpendicular magnetization anisotropy in Au/Co/Au(111) sandwiches by Chappert *et al.*,[2] an easy-magnetization axis perpendicular to the film was found by several authors. The films display square hysteresis loops[5,6] giant magnetoresistance,[7] and magneto-optical effects.[4] All of these studies find that the magnetization axis turns perpendicular to the plane of the film for the layers thinner than ~ 7 monolayers (ML). While these investigations probed the magnetic effects over macroscopic lateral dimensions, recently the observation of magnetic domain structure has been reported.[8,9]

Little structural information about ultrathin Co/Au(111) films is available. The relative Co/Au lattice mismatch was measured with electron diffraction.[6,7] For films thicker than 10 (ML, x-ray-diffraction measurements indicate that the cobalt layer has a hexagonal (hcp) structure.[10] Given the large lattice mismatch between Co and Au (14%), one would anticipate that the growth does not occur layer by layer. However, little is known about the nature of the epitaxial growth of Co films. Below, we report on the initial nucleation steps and the growth of Co on Au(111), and we relate the structural results to recent findings from magnetic measurements.

The experiments were performed with a STM at pressures of 1×10^{-10} mbar. The Au/(111) crystal was cut and polished to within 0.4° of the desired orientation, which corresponds to an average terrace width of ~ 400 Å. The crystal was cleaned by Ne-ion bombardment (1.5 keV) and annealed at ~ 600 °C for 5 min. Cobalt evaporation and STM measurements were performed at 300 K. The cobalt was evaporated, *in situ,* from an electron-beam evaporator, at a rate of 0.1–0.5 ML/min. During the evaporation, the pressure was $<1\times10^{-9}$ mbar. The cobalt flux was measured with a quartz-crystal microbalance and agreed to within 15% with the coverage obtained from the STM images. The images were obtained in the constant-current mode with tunneling currents of 0.2–1.0 nA using positive-bias voltages ranging from 0.5 to 2.5 V.

It is instructive to examine the Au(111) surface first since its structure influences the nucleation and subsequent growth of the Co layer. Gold is the only fcc metal where the close-packed (111) surface has been observed to reconstruct. Previous STM measurements have atomically resolved the $(23\times\sqrt{3})$ reconstructed unit cell.[11,12] The surface is uniaxially contracted (4.2%) along $\langle 1\bar{1}0\rangle$ directions. This long-range reconstruction separates regions of fcc (*ABC*) and hcp (*ABA*) stacking on the surface. The regions with the surface atoms in different kinds of hollow sites are separated by partial surface dislocations in which the surface atoms are near bridge sites (i.e., higher than in hollow sites). In the STM image (Fig. 1), the dislocation regions appear as 0.15-Å high ridges separating the fcc from hcp regions. The double rows in Fig. 1 (63-Å periodicity) appear because the fcc stacking regions are broader (lower free energy) as compared to the hcp stacking regions. In large-area scans, we observe a regular alternation of ~ 150-Å-long uniaxial reconstruction domains (along the $\langle 1\bar{2}1\rangle$ direction), which form a zigzag pattern. This pattern leads to a more uniform contraction on the surface, while a single uniaxial domain would lead to a strain relieve only in one direction. The left ridges of the double-ridge structure in Fig. 1 are quite regular, while the right-hand ones bulge out and narrow alternately at the kinks. A closer look at the surface crystallography of the Au(111) reconstruction shows that the left and right ridges join in a different way at the kinks of the zigzag pattern (domain boundaries).[13] The left segment of the double ridges in Fig. 1 have the same Burgers vectors and join without additional dislocations. Two joining right segments have different Burgers vectors,[13] which leads to an additional dislocation at the domain boundaries. At the kink locations, we often ob-

serve an increased corrugation of 0.6–0.9 Å.

When Co is deposited at room temperature, a regular array of Co islands nucleates at the domain boundaries of the zigzag reconstruction, i.e., at the kink positions. To be more precise, the Co islands nucleate at domain boundaries where two ridges join with an additional dislocation present. In Fig. 2(a) a surface with a Co coverage of 0.3 ML is shown. Linear arrays of Co islands are readily seen whose pattern is determined by the Au zigzag reconstruction. This results in one-dimensional chains of closely spaced islands, with the original double-ridge Au reconstruction still visible. Additionally, small islands decorate the step edges. The polygonal shape of the islands and their orientation with respect to the reconstruction ridges show that the islands grow epitaxially with their edges aligned along the close-packed rows of the substrate ($\langle 1\bar{1}0\rangle$). The islands are hexagonal with a threefold symmetry having long and short-step segments opposite to each other. The high density and regular arrangement of similar nucleation centers on this surface yield a narrow size distribution of islands as contrasted with the distribution expected from homogeneous nucleation. The two monoatomic steps in the lower part of Fig. 2(a) can be used to determine the Co island height. The line scan in Fig. 2(b) is calibrated by the 2.36-Å height of the Au(111) steps. The dotted lines indicate the height of 2-ML cobalt (0001). No substantial relaxation with respect to the bulk values was detected, and the Co islands grow as a bilayer. We attribute the double-layer growth to the large lattice mismatch between bulk Au and Co.

As the Co coverage increases, the island size increases, and at ~ 0.8 ML some islands start to coalesce [Fig. 3(a)]. The corrugation on top of the islands ($\lesssim 1$ Å) is higher than on the Au substrate and may be due to dislocations relieving the strain in the Co layer. In Fig. 3(b) the gray scale is chosen such that every atomic layer is shown with only one gray value. The image shows that the islands still have predominately double-layer height with a small fraction showing a third layer growth. With increasing coverage, the islands coalesce along the $\langle 1\bar{1}2\rangle$ direction whose periodicity is only 73 Å [$= (63\text{ Å})\times 2/\sqrt{3}$]. Upon deposition of 2–3 ML, we observe that the Co overlayer coalesces further [Fig. 4(a)]. While some of the Au(111) surface is still bare, the Co layer is nevertheless fully contiguous. The corrugation on top of the islands is ~ 1 Å, and some adatom clusters are present. Even at a coverage of 7 ML, a granular structure, resulting from the initial nucleation pattern, is observed [Fig. 4(b)]. Additionally, step edges covered by the Co layer can be identified. Upon deposition of ~ 12 ML Co, the granular structure disappears, and the surface develops 100–300-Å-size terraces which exhibit height variations of about ± 3 ML.

From our data the following model for the Co nucleation and growth on Au(111) is deduced: As the Co atoms impinge on the Au surface, they diffuse until they stick at one of the elbow sites of the reconstruction pattern. The dislocations at these sites are the nucleation centers for the growth. Islands form when additional Co atoms encounter the nucleation centers. The large lattice mismatch between Au and Co (14%) leads to a buildup of considerable lattice strain. However, the strain is already

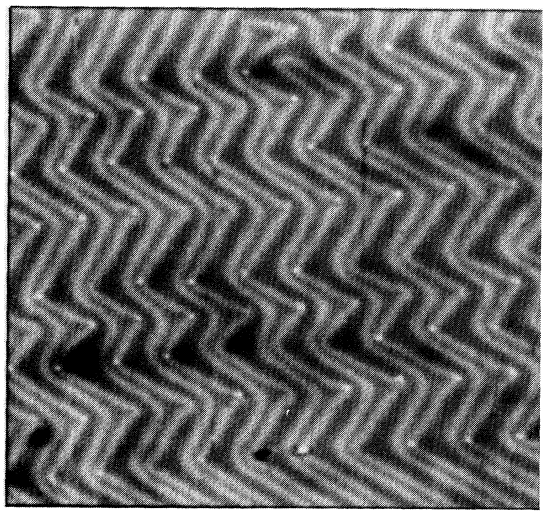

FIG. 1. Reconstructed Au(111) surface (730×730 Å^2) with rotational domains along the $\langle 1\bar{1}2\rangle$ direction forming an ordered zigzag pattern.

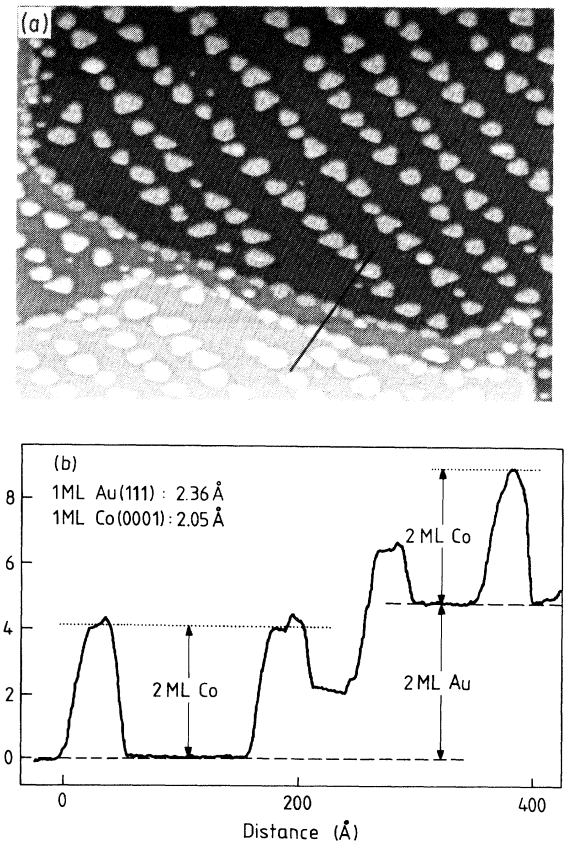

FIG. 2. (a) 0.3-ML Co coverage on Au(111) (1600×1600 Å^2); 2-ML-high polygonal Co islands nucleate at the kinks of the Au(111) zigzag reconstruction. (b) Height variation along the line indicated in (a).

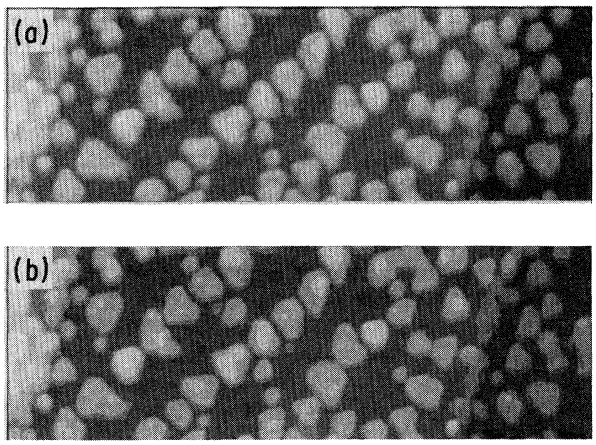

FIG. 3. (a) 0.8-ML Co coverage on Au(111) (1100×400 Å^2) showing some Co islands beginning to coalesce. (b) Same as in (a), but displayed with only one gray value per atomic layer.

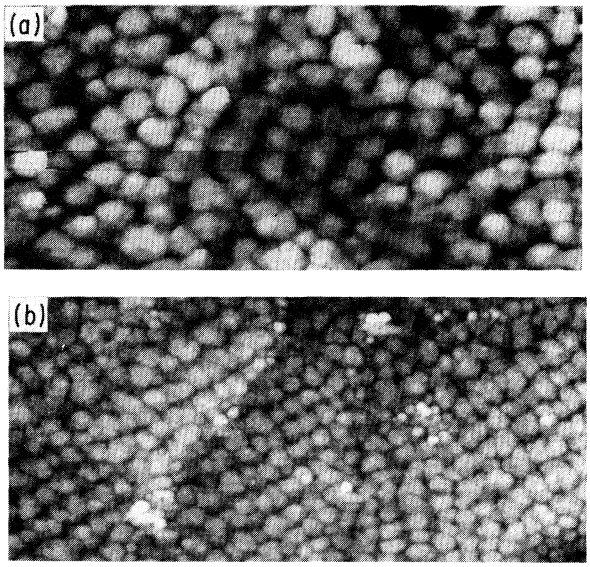

FIG. 4. (a) 3-ML Co coverage on Au(111) (1600×800 Å^2). At this coverage the Co layer is already contiguous. (b) 7-ML Co coverage (3200×1600 Å^2) showing a persisting regular array of Co islands related to the initial nucleation pattern.

relaxed to ~8% in the second layer.[17] Therefore, the total strain energy is reduced by the double-layer growth. The strain relief by misfit dislocations leads to the roughness observed on the islands. The shape of the islands, hexagonal along the close-packed $\langle 1\bar{1}0 \rangle$ directions with threefold symmetry (long- and short-step segments opposite to each other), was also found on a ion-bombarded platinum (111) surface.[14,15] Opposite step edges belong to different facets: One of the $\langle 1\bar{1}0 \rangle$ step segments belongs to a {111} facet and the opposite belonging to a {100} facet. The free energy of the {111} facets is lower than that of the {100} ones. The same may also hold for the corresponding step types leading to the largely triangular shape of the islands. It is interesting to note that triangular-shaped islands with two different orientations are present [Fig. 2(a)]. This shows that the {111} and {100} faceted steps have interchanged their orientation, which can be explained by a different stacking order for different oriented islands. When the islands coalesce, we often observe trenches between nearby islands (Fig. 3). The formation of the trenches can be attributed to the decreased area available for diffusion to these trenches.

Apart from the nucleation of Co islands on the terraces, Co islands also nucleate at the step edges at positions determined by the reconstruction ridges. The double-layer islands, which nucleate at the Au steps, are generally smaller than the islands at the kink sites of the reconstruction pattern [Fig. 2(a)].

Comparison of the growth of different metals on Au(111) shows that for Au and Ag on Au(111) the nucleation occurs predominantly at the gold step edges.[16–18] The metals with larger misfit, Ni,[13] Fe[19] and Co, nucleate predominantly at the kinks of the Au reconstruction pattern. In this dislocation region, the rather large distortion from the ideal Au(111) surface crystallography appears to favor the nucleation of metals with a large lattice mismatch.

It is interesting to correlate our structural findings with the magnetic properties of thin Co layers on Au(111).[2,4–9] The formation of perpendicular magnetic domains at coverage ≥2 ML (Ref. 9) coincides with our observation that the Co layer coalesces at this coverage over large regions (which is a consequence of the bilayer growth). One would expect that only a connected Co layer leads to ferromagnetic coupling between the islands and to the formation of domains in the film. No apparent structural change is observed in the coverage range between 4 and 7 ML, where the magnetization axis changes from perpendicular to parallel to the surface. With increasing film thickness, the magnetic shape anisotropy, which leads to an in-plane magnetization in thin films, overcomes the perpendicular anisotropy which dominates in ultrathin films.

In summary, the epitaxial growth of Co on Au(111) was probed with scanning tunneling microscopy. We propose a model to describe the initial nucleation and growth of the Co film, and we compared our results with the growth of other metals on this gold surface and related structure to published magnetic measurements.

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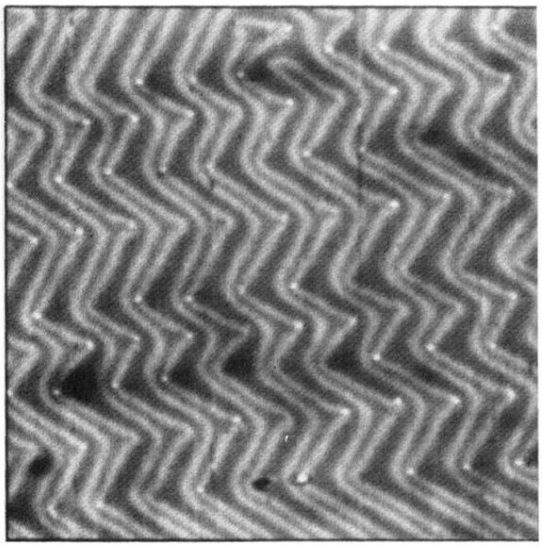

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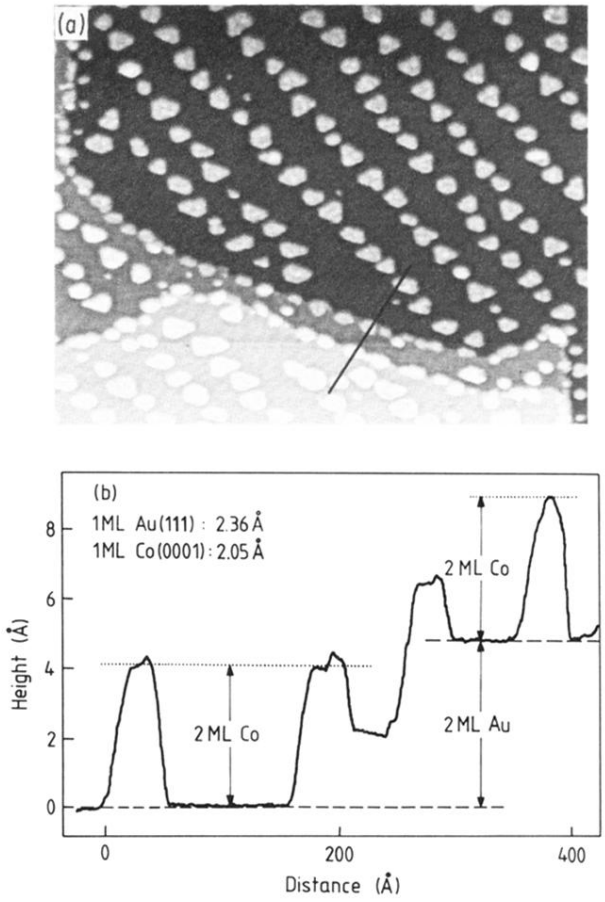

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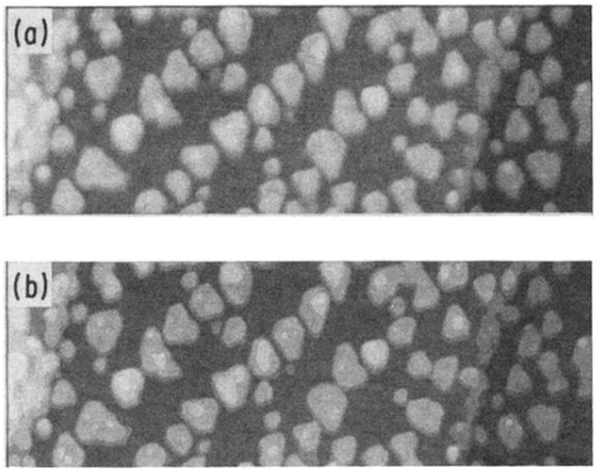

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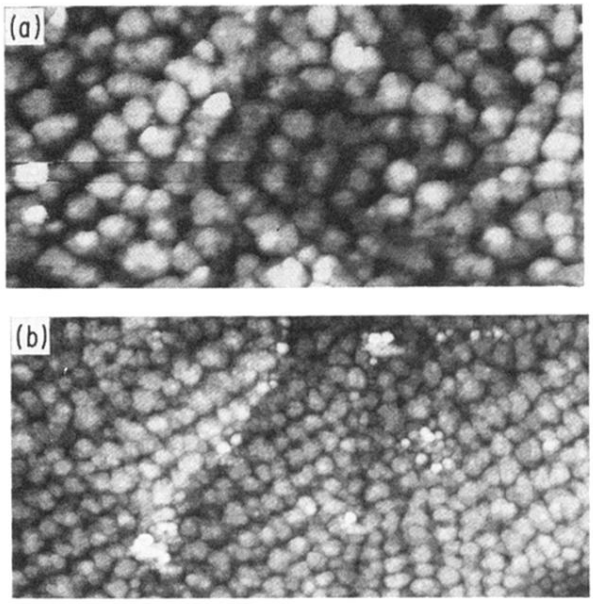

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