Magnetic Domain Structures in Ultrathin Co/Au(111): On the Influence of Film Morphology

M. Speckmann, H. P. Oepen, and H. Ibach
Institut für Grenzflächenforschung und Vakuumphysik, Forschungszentrum Jülich, D-52425 Jülich, Germany
(Received 21 February 1995)

The magnetic domain structures in thin hcp Co films grown on Au(111) have been investigated by means of scanning electron microscopy with polarization analysis. The domain pattern in films with out-of-plane magnetization were analyzed in the as-grown state and after annealing. The heat treatment causes the morphology and the magnetic microstructure to change drastically. The films become smoother and the domain pattern exhibits a characteristic thickness dependence. A collapse of the domain size near the reorientation transition is found, consistent with micromagnetic theory.

PACS numbers: 75.70.Kw, 75.30.Kz, 75.70.Ak, 75.70.Cn

Current research on the magnetic structure of ultrathin films is largely focused on the interplay between surface and interface anisotropies and shape anisotropies. Whereas surface and interface anisotropies can cause an alignment of the magnetiziation perpendicular to the plane of the film, the shape anisotropy due to dipolar magnetic interactions tends to orient the magnetization in the plane of the film. With increasing thickness, dipolar interactions gain strength and hence may cause a transition from vertical to in-plane magnetization. Furthermore, the micromagnetic structure of the films is determined by the relative strength of surface and interface anisotropies, on the one hand, and the shape anisotropies, on the other hand [1–4]. Both the reorientation transition as well as the micromagnetic structure have been recently discussed for cobalt films grown on Au(111) [5,6]. Conflicting evidence concerning the micromagnetic structure and the micromagnetic behavior was, however, presented, since in Ref. [5] small domains were reported, while no domains have been found in Ref. [6]. This discrepancy was not understood until now. As substrates and preparation conditions differed in the two studies, the contradicting results could be related to a different morphology of the films.

The film morphology may modify the magnetic microstructure in various ways. On the one hand, the micromagnetic structure is influenced by anisotropies. As both the interface (including surface) and shape anisotropies are strongly dependent on the film structure and interface roughness [7–11], the morphology affects the relative strength of the various anisotropies and, hence, the micromagnetic structure. On the other hand, film imperfections can act as pinning centers for domains and walls. In that case the domain structure is directly determined by the film morphology.

This paper deals with the correlation of film morphology and magnetic microstructure in the regime of perpendicular magnetization. In order to determine the influence of structure and morphology on magnetic structures, a combined investigation, utilizing medium energy electron diffraction (MEED), Auger electron spectroscopy, and scanning electron microscopy with polarization analysis (SEMPA), has been performed. It will be shown that as-grown films are rough and become smoother by heat treatment. In the as-grown films the morphology determines the domain pattern, while the improvement of film homogeneity gives a nearly single domain configuration. It will be shown that these results can explain the differences of the previous investigations.

The experiments have been performed under UHV conditions (base pressures $\leq 10^{-10}$ Torr). The Au(111) single crystal was cleaned by cycles of Ar bombardment (600 eV) and annealing ($T \approx 700\,^{\circ}\mathrm{C}$). Cobalt was deposited at room temperature with a growth rate of 0.3–0.5 Å/min. During evaporation, the pressure did not exceed 6×10^{-10} Torr. The Co films were annealed by heating to 240 °C for 10 to 15 min.

In a first series of experiments the growth was studied by means of MEED and MEED intensity oscillations. During deposition at room temperature the MEED spots became completely diffuse and the spot intensities dropped rapidly, indicating a 3D-like growth, which has also been found in STM studies [12]. Annealing of the samples reestablished the MEED pattern. MEED intensity oscillations could be found with continuing cobalt deposition on the annealed Co film. Hence the MEED study indicated that the initially rough surface becomes smooth after annealing.

Next, we studied the influence of annealing on film morphology by means of Auger electron spectroscopy. The ratio of the peak-to-peak intensities of the low energy Auger transitions I = Co(53 eV)/Au(66/69 eV) was measured before and after annealing. Annealing films in the thickness range of 2–4 monolayers (ML) leads to a decrease of I, which indicates that in the topmost atomic layers the amount of Au has been increased. To clarify whether an intermixing or a clustering happens at the surface or if diffusion of Au leads to a covering of Co by gold, Auger depth profiles have been taken by sputtering the samples with argon ions (600 eV) at low current densities (100 nA/cm^2). Figure 1 shows the Auger ratio I as a function of sputtering time for an as-grown and an annealed film. For the annealed film

0031-9007/95/75(10)/2035(4)$06.00

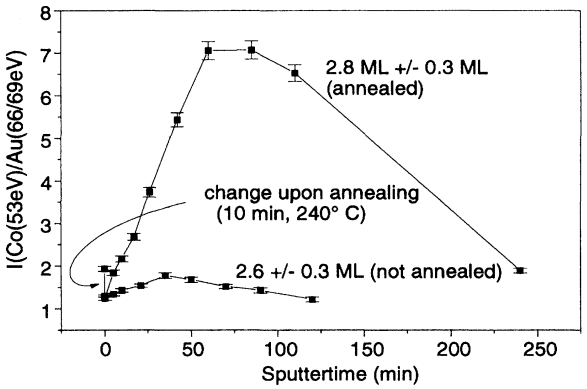

FIG. 1. Intensity ratio I of the Co(53 eV) and the Au(66/69 eV) Auger electron transition versus sputtering time. The dependence of I on sputtering is shown for an annealed and an as-grown film.

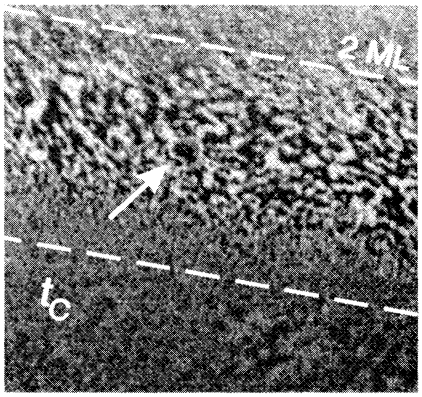

FIG. 2. SEMPA images of the magnetic domain structure in a wedge-shaped Co/Au(111) film. Scanning area is 100 μm $\times$ 100 μm. The image shows domains obtained for the perpendicular component of magnetization. The light areas indicate domains with magnetization pointing out of the image, black areas are domains with a magnetization pointing into the plane. Grey tones represent areas with vertical polarization components in between. The thickness increases from the upper right to lower left edge of the image. The dashed lines represent levels of equal film thickness. The critical thickness t_c is 3.9 ML. In the center of the image a defect serves as a marker and is indicated by an arrow.

the ratio I increases on sputtering and reaches values significantly higher than those obtained for the as-grown film. This finding rules out intermixing or clustering effects. It proves unambiguously that Au is at the surface, which is removed in the first few minutes of sputtering. Below the top Au layer, a larger fraction of the surface is covered with Co compared to the virgin films. This means that the Co islands must have been decomposed and cobalt has spread over the Au surface. Hence, the annealing produces a smoother Co film with a more homogeneous thickness.

The influence of the structural changes on magnetic microstructure was studied by means of SEMPA. The thickness dependence of the domain size and magnetization direction was investigated using wedge-shaped samples. Figure 2 shows the domain pattern obtained for the perpendicular component of magnetization of an as-grown film. The film thickness changes from 2 ML (top) to 5 ML (bottom of Fig. 2). The Co films become ferromagnetic at approximately 2 ML. The magnetization is purely perpendicular between about 2 and 3.8 ML. The domains have nearly constant size in the range of 2 to 5 μm as found in previous studies [5]. Only near t_c is the average domain size slightly decreasing. Above a critical thickness t_c [13] first indications of in-plane magnetization are found in the other spin-polarization components.

The influence of annealing on magnetic microstructure can be seen in Fig. 3 showing the same film after thermal treatment. Striking changes of the domain configuration have occurred. (1) The domain size has increased dramatically by more than 1 order of magnitude. Such an increase of domain size on annealing has been previously mentioned [14]. (2) The critical thickness t_c has shifted towards higher thickness. (3) The most remarkable effect is the strong decrease of the domain size within a very small thickness range in the vicinity of t_c. The domain size is decreasing by more than 1 order of magnitude within a thickness variation of only 0.5 ML, while the magnetization stays perpendicular as proven by vector analysis of the spin polarization.

The strong dependence of domain size on thickness becomes more obvious with the plot in Fig. 4, showing the number of domain boundaries per length as a function

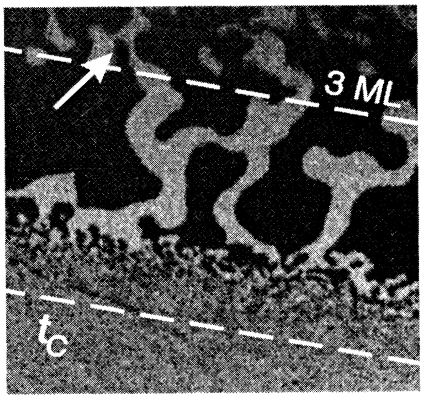

FIG. 3. Domain image of the same film as shown in Fig. 2, after annealing. Scanning area is 100 μm $\times$ 100 μm. The grey scales have the same meaning as in Fig. 2. The critical thickness t_c given in the images is 4.9 ML. The image shows a part of the film which is slightly shifted against that shown in Fig. 2. The link to Fig. 2 is possible via the defect indicated in both images.

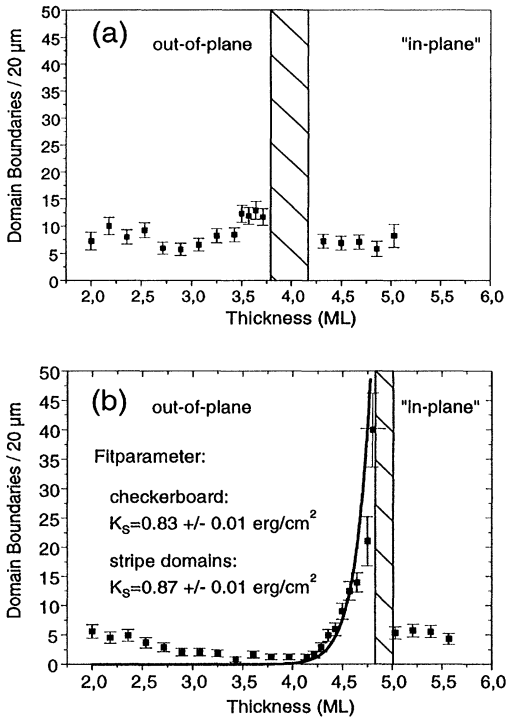

FIG. 4. Number of domain boundaries per length versus film thickness for an as-grown (a) and an annealed (b) film. Error bars give the 1σ uncertainty. The outlined region represents the thickness interval where the transition from perpendicular to in-plane magnetization takes part. In the thickness regime indicated by "in-plane" still some canting of magnetization is found. The solid line (b) is a fit of Eq. (1) to the data.

of film thickness for an as-grown (4a) and an annealed film (4b). To obtain these data the number of domain walls along lines of constant thickness have been counted. Several images of domain pattern including Figs. 2 and 3 have been used to extract the quantitative numbers. It is evident (Fig. 4) that the transition where the magnetization turns into an in-plane orientation has shifted by nearly 1 ML towards higher thicknesses upon annealing. This change indicates that the perpendicular anisotropy is also affected by the heat treatment, i.e., the perpendicular anisotropy becomes stronger.

The as-grown film [Fig. 4(a)] exhibits only a small variation of the number of domain boundaries per length, which indicates, vice versa, the nearly constant domain size. In the case of the annealed films [Fig. 4(b)] the number of domain walls decreases continuously up to a film thickness of 3 ML. In the vicinity of the onset of ferromagnetism ($t \leq 2.5$ ML) the domain size is comparable to that of the as-grown films. From this equivalence and the similarity of the domain pattern in those thickness regimes, we conclude that below 2.5 ML the domains are still determined by 3D growth and the incomplete coverage of the substrate. The annealing cannot wipe out this influence in the very low thickness regime completely. Above a thickness of 3 ML the domain size is nearly constant and large. Within a very small thickness range around 4.5 ML a strong increase of boundaries indicates the shrinking average size of the domains. High resolution images show that the collapse of domain size can be observed down to domains with lateral dimensions of 300–500 nm near the onset of transition to in-plane magnetization [15].

The strong decrease of the average domain size is predicted by theory of domain formation in thin perpendicularly magnetized films (e.g., [1–4]). It is due to the fact that $\vec{M}$ can remain perpendicular by decreasing the shape energies via domain creation. The thickness dependence of the domain size can be calculated by minimizing the total free energy with respect to the average domain size.

Kaplan and Gehring [3] give an analytical approximation for the thickness dependence of the domain size for both stripe and checkerboard domains:

$$D = xt \exp\left(\frac{\sigma_w}{4M_s^2 t}\right). \quad (1)$$

M_s is the saturation magnetization and t is the film thickness. The prefactor x has a value of $x = 0.955$ for stripe domains and $x = 2.62$ for the checkerboard pattern [3].

We have fitted Eq. (1) to our data (see Fig. 4) using the well-known expression for the domain wall energy in magnets with uniaxial anisotropy $\sigma_w = 4\sqrt{AK}$. For the anisotropy constant we use the effective value $K_{\text{eff}} = K_b + K_s/t - 2\pi M_s^2$ with K_b and K_s as the anisotropy constants of bulk hcp cobalt and the Co interfaces, respectively. A is the exchange constant.

As a fit parameter K_s was used while literature values have been taken for all other material parameters [16]. The best fit yields $K_s = 0.83 \pm 0.01$ erg/cm^2 for the checkerboard and $K_s = 0.87 \pm 0.01$ erg/cm^2 for the stripe domain configuration. The values are larger than the anisotropy of a *single* Co/Au(111) interface [17]. This result indicates that more than one Co/Au interface contributes to K_s. Hence, the magnetic behavior of the film is consistent with the results of the Auger analysis, which showed that the Co film is covered with Au after annealing. The Co/Au interface anisotropy is larger than the Co surface anisotropy, as shown by Ould-Mahfoud *et al.* [18]. Thus, in total, K_s increases on annealing, which causes the shift of t_c already mentioned above.

The theory describes the observed collapse of the domain size (see Fig. 4) very well. The model, however, predicts a single domain state in the thickness range below approximately 3.8 ML, which could not be observed in the wedges shown. By investigating wedges with different shapes it turned out that the maximum domain size is strongly affected by macroscopic dimensions, like, e.g.,

the thickness gradient. It is possible to generate a large domain in wedges with very small thickness gradients. Only in the lowest thickness regime ($\leq$3 ML) do small domains persist. Thus we conclude that apart from the low thickness regime the observed thickness dependence of the domains is very close to the equilibrium size distribution determined solely by magnetic properties, as the comparison with theory implies.

On magnetizing the films in an external field, the magnetic microstructure can be transformed into a metastable single domain state with perpendicular magnetization. The transition to in-plane magnetization is pulled to even higher thicknesses and a continuous rotation of the magnetization from vertical to in-plane orientation is observed. This single domain configuration with perpendicular magnetization could not be changed by demagnetization procedures. Heating the sample reestablishes qualitatively the same domain structure as before applying the field.

The findings presented in this paper can explain the differences in domain size found previously for Co/Au(111) [5,6]. One has to conclude that in the thin film limit and for as-grown films the small domain size is induced by morphology. Such domain patterns have been investigated in [5]. It is not surprising that in this case a comparison of experimental results with magnetic models must fail, if influences of film morphology are ignored. An improvement of film quality causes an increase of domain size up to a single domain state and enlarges the thickness regime of perpendicular magnetization. In a high quality film, domains appear only in a very narrow thickness interval close to but below t_c, which is hard to find without wedge-shaped films. Hence, a single domain state reported by Pommier *et al.* [6] can be explained by a smooth film, or by a metastable state artificially produced by applying an external field.

The authors would like to thank U. Linke for preparing the Au(111) single crystal and A. Suciu for performing the MEED intensity measurements. Discussions with A. Berger (University of California) are gratefully acknowledged.

[1] Z. Málek and V. Kamberský, Czech. J. Phys. **21**, 416 (1958).
[2] Y. Yafet and E. M. Gyorgy, Phys. Rev. B **38**, 9145 (1988).
[3] B. Kaplan and G. A. Gehring, J. Magn. Magn. Mater. **128**, 111 (1993).
[4] A. B. Kashuba and V. L. Pokrovsky, Phys. Rev. Lett. **70**, 3155 (1993); Phys. Rev. B **48**, 10 335 (1993).
[5] R. Allenspach, M. Stampanoni, and A. Bischof, Phys. Rev. Lett. **65**, 3344 (1990).
[6] J. Pommier, P. Meyer, G. Pénissard, J. Ferré, P. Bruno, and D. Renard, Phys. Rev. Lett. **65**, 2054 (1990).
[7] F. J. A. den Broeder, D. Kuiper, A. P. van de Mosselaer, and W. Hoving, Phys. Rev. Lett. **60**, 2769 (1988).
[8] P. Bruno, J. Phys. F **18**, 1291 (1988).
[9] G. H. O. Daalderoop, P. J. Kelly, and F. J. A. den Broeder, Phys. Rev. Lett. **68**, 682 (1992).
[10] P. Bruno, J. Appl. Phys. **64**, 3153 (1988).
[11] H. J. G. Draaisma and W. J. M. de Jonge, J. Appl. Phys. **64**, 2610 (1988).
[12] B. Voigtländer, G. Meyer, and N. M. Amer, Phys. Rev. B **44**, 10 354 (1991).
[13] Note that other authors define the critical thickness t_c as the value where the magnetization is canted by 45° to the normal. We take t_c as the thickness where the magnetization starts to deviate from the normal.
[14] R. Allenspach, J. Magn. Magn. Mater. **129**, 160 (1994).
[15] The minimum domain sizes found near the transition are in good agreement with theoretical suggestions [2,4]. We do not want to stress that issue, as further work is necessary.
[16] $K_b = K_1 = 5.1 \times 10^6$ ergs/cm^3; $M_s = 1440$ emu, $A = 1.3 \times 10^{-6}$ erg/cm as published in *Magnetic Properties I*, edited by K.-H. Hellwege and A. M. Hellwege, Landölt-Bornstein Vol. II, Pt. 9 (Springer, Berlin, 1962), 6th ed.; *3d, 4d, and 5d Elements, Alloys and Compounds*, Landolt-Börnstein New Series, Group 3, Vol. 19, Pt. a (Springer, New York, 1986).
[17] For a survey of published values see Chap. 2.3, in *Ultrathin Magnetic Structures I,* edited by J. A. C. Bland and B. Heinrich (Springer, New York, 1994).
[18] S. Ould-Mahfoud, R. Megy, N. Bardou, B. Bartenlian, P. Beauvillain, C. Chappert, J. Corno, B. Lecuyer, G. Sczigel, P. Veilet, and D. Weller, in Mater. Res. Soc. Symp. Proc. **313**, 251 (1993).

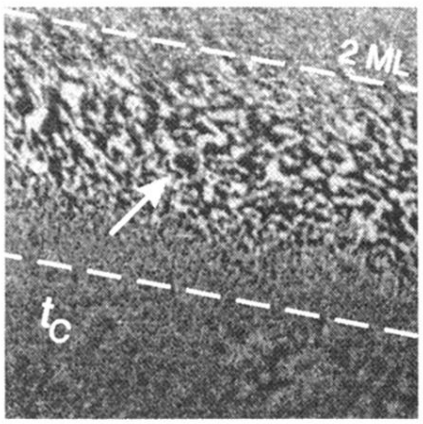

FIG. 2. SEMPA images of the magnetic domain structure in a wedge-shaped Co/Au(111) film. Scanning area is 100 μm $\times$ 100 μm. The image shows domains obtained for the perpendicular component of magnetization. The light areas indicate domains with magnetization pointing out of the image, black areas are domains with a magnetization pointing into the plane. Grey tones represent areas with vertical polarization components in between. The thickness increases from the upper right to lower left edge of the image. The dashed lines represent levels of equal film thickness. The critical thickness t_c is 3.9 ML. In the center of the image a defect serves as a marker and is indicated by an arrow.

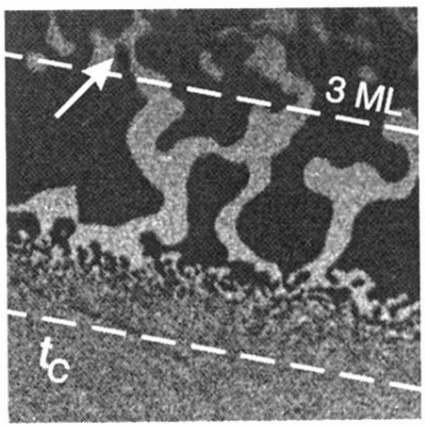

FIG. 3. Domain image of the same film as shown in Fig. 2, after annealing. Scanning area is 100 μm $\times$ 100 μm. The grey scales have the same meaning as in Fig. 2. The critical thickness t_c given in the images is 4.9 ML. The image shows a part of the film which is slightly shifted against that shown in Fig. 2. The link to Fig. 2 is possible via the defect indicated in both images.