Magnetization Wrinkle in Thin Ferromagnetic Films

T. Duden and E. Bauer

Physikalisches Institut, Technische Universitaet Clausthal, D 38678 Clausthal-Zellerfeld, Germany and Department of Physics and Astronomy, Arizona State University, Tempe, Arizona 85278-1504

(Received 9 May 1996)

Using spin-polarized low energy electron microscopy with polarization manipulation we have observed a new magnetic ordering pattern in thin Co films on W(110). The predominantly uniaxial in-plane magnetization is superimposed by a more fine-grained out-of-plane component in a striped or checkerboardlike arrangement, which is strongly modified by the step distribution. This results in a wrinkled magnetization which should occur also in many other systems. [S0031-9007(96)00917-9]

PACS numbers: 75.70.Kw, 75.25.+z, 75.30.Gw

Ultrathin ferromagnetic films have become one of the most exciting subjects in solid state physics in recent years, mainly because of their importance in magnetic storage devices and sensors. In these films there are always several ordering or "anisotropy" fields present. These include shape anisotropy, magnetoelastic anisotropy, magnetocrystalline anisotropy, and, foremost of all, surface anisotropy. "Surface" anisotropy usually contains all contributions to the anisotropy which increase as $1/t$ with decreasing film thickness t and, therefore, plays a dominating role in ultrathin films. The interplay of the various anisotropies produces a wide range of spin configurations as a function of film thickness and temperature, and transitions between them, the so-called spin-reorientation transitions (SRT, for a review see Ref. [1]).

In the past it has generally been assumed that in a SRT the magnetization rotates 90° within a narrow thickness range, either from perpendicular magnetization to in-plane or in-plane in the case of uniaxial in-plane anisotropy. Intermediate ("canted") magnetization has been attributed to spatial fluctuations in the thickness [2], that is, to surface or interface roughness. In this Letter we show that canted magnetization is not limited to rough surfaces or a narrow thickness range, but probably is a general state of magnetization in ultrathin films and leads to wrinkled overall magnetization. For this purpose we use ultrathin Co films on a W(110) surface which have a strong uniaxial in-plane anisotropy, caused by the Co/W interface [3,4], and an imaging method which gives both structural information on the monolayer (ML) scale and magnetic information, spin-polarized low energy electron microscopy (SPLEEM). SPLEEM makes use of the fact that the elastic reflection coefficient of a magnetic surface or thin layer contains a contribution which is proportional to $\mathbf{P} \cdot \mathbf{M}$ where $\mathbf{P}$ is the polarization of the incident beam and $\mathbf{M}$ the magnetization of the sample. (For a review of LEEM and SPLEEM, see Ref. [5].) In the original SPLEEM study of Co films on W(110) [3] $\mathbf{P}$ could be rotated only in plane so that only the uniaxial anisotropy could be determined. A new spin-polarized electron gun with polarization manipulation [6] allows us now to select arbitrary $\mathbf{P}$ directions so that also out-of-plane components of $\mathbf{M}$ can be determined.

The Co layers were deposited in the low to middle 10^{-10} Torr range at a typical rate of 0.1 ML/min and at a glancing angle of incidence of 15° on the well-cleaned W(110) surface and the microstructure and magnetic structure was monitored continuously. The depositions were made at several substrate temperatures but only results obtained under optimum growth conditions, i.e., ML-by-ML-like (quasi Frank–van der Merwe) growth are reported here. These optimum conditions were determined in earlier studies of the growth and stability of Co layers on Mo(110) [7] and W(110) [8,9] surfaces and confirmed in the present work. They consist in depositing the first ML at about 700 K, the subsequent MLs at about 400 K. At 700 K the pseudomorphic (ps) ML grows by step flow growth and the smoothness of the growth front serves as an indication of clean growth conditions [10]. The transition from the ps to the close-packed (cp) ML and the completion of the cp ML allow a very accurate rate calibration at the beginning of each growth experiment. The subsequent growth at 400 K occurs with sufficiently low nucleation and high diffusion rate so that large ML terraces are formed before the next ML nucleates. In this manner the growth front consists of at most three ML levels even at a thickness of 10 ML. The ML height differences are imaged by quantum size contrast [5,11]. Higher growth temperatures cannot be used because the kinetically limited Frank–van der Merwe growth converts then into the equilibrium Stranski-Krastanov growth with three-dimensional Co crystals on the first ML [7–9].

Figure 1 shows a typical in-plane (a), out-of-plane (b), magnetization tilt angle (c), and structural (d) image. Contrary to previous belief, the layer shows not only uniaxial in-plane magnetization (a) but also a measurable out-of-plane $\mathbf{M}$ component (b) with completely different domain structure. The two images show the asymmetry $A = P(I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow)$ distribution whose grey levels are directly proportional to the $\mathbf{M}$ component parallel to $\mathbf{P}$. In a grey level scale from -128 to $+128$ the white and black regions in (a) correspond to an in-plane component

 0031-9007/96/77(11)/2308(4)$10.00

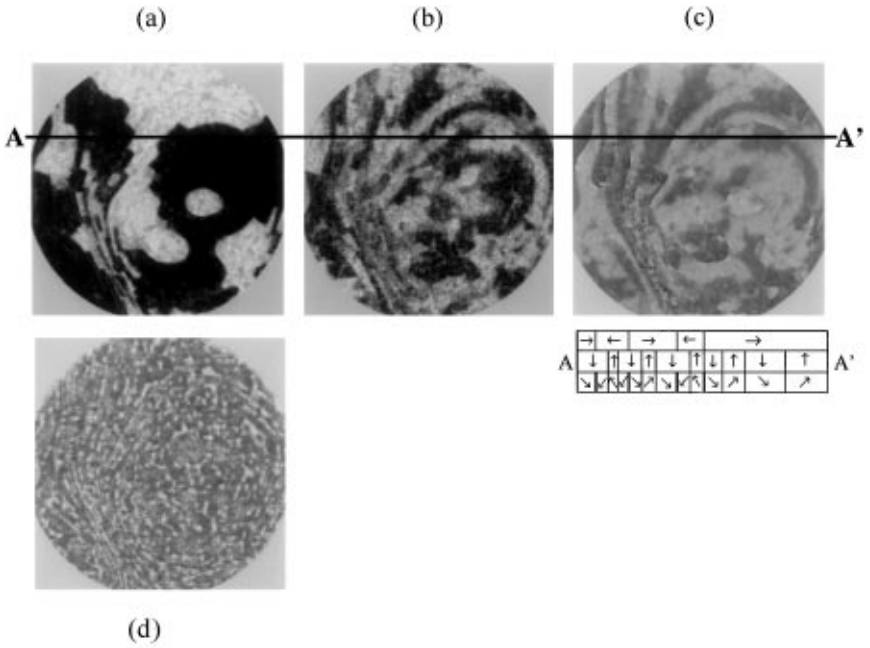

FIG. 1. SPLEEM images of a Co layer on W(110). Coverage 5.0 ML, electron energy 2.1 eV, field of view 8 μm. (a) In-plane **M** image, (b) out-of-plane **M** image, (c) **M** tilt angle image, and (d) structural image. Below (c) a schematic presentation of the **M** distribution along the section AA′ is shown.

$\mathbf{M}_{\parallel}$ parallel to $[1\bar{1}0]$ and $[\bar{1}10]$ in the W(110) surface, respectively.

The less extreme grey levels in (b) clearly indicate a much smaller out-of-plane component $M_{\perp}$. The magnitude $M(x, y)$ of the magnetization can be obtained from the square sum of the signal components $I_{\perp} \propto M_{\perp}$ and $I_{\parallel} \propto M_{\parallel}$ as a function of position x, y. It shows slight variations within the domains which can be correlated with the thickness variations seen in the topographic image [Fig. 1(d)]. This is a consequence of the spin dependence of the quantum size contrast which makes extraction of the *magnitude* of **M** difficult without calibration of the thickness and energy dependence of the contrast [5].

The determination of the distribution of the *direction* of **M** as expressed, for example, by the tilt angle $\alpha = \arctan(I_{\perp}/I_{\parallel})$ of **M** with respect to the surface, however, does not suffer from this complication because $I_{\perp}$ and $I_{\parallel}$ have the same thickness and energy dependence. α is shown in Fig. 1(c) in the grey level scale 128 $[1 + \alpha/(\pi/2)]$. Its significance is illustrated by the $M_{\parallel}$, $M_{\perp}$, and **M** distributions along the section AA′ in Fig. 1, taken from (a) and (b) and schematically added up to **M**. Clearly, a wrinkled magnetization results with several poles indicated by heavier lines. The black-white double lines in Fig. 1(c) are a result of the zero crossing of the in-plane signal I which lets the argument of the arctan function diverge. In the lower left region the domain walls are so closely spaced that it is difficult to trace the **M** distribution. This is connected with the high step density ($\approx 15/\mu$m) at the lower left side of a large flat region with about 3 μm diameter on the right side of the image, which can be seen in the original of Fig. 1(d). The energy in Fig. 1(d) was chosen such that both step contrast and quantum size contrast were obtained. (For an explanation of these contrast mechanisms, see Ref. [5].) Because of the quasi Frank–van der Merwe growth the substrate steps have propagated through the layer. They are frequently the loci of domain walls [see (a, b)]. In addition to the steps, predominantly two grey levels are seen on the terraces which indicate a predominant two-atomic level system. The dark/bright area ratio gives a coverage of 4.7 ML, the rate calibration 5.0 ML. Closer inspection of the originals shows a third grey level but these regions are too small for quantitative evaluation. Thus some third level contribution is already present too. The dark (ML) islands have lateral dimensions of a few hundred nm.

Numerous measurements on individually and cumulatively deposited layers show that the wrinkled magnetization is always present in the thickness range studied (3–8 ML). Below 3 ML the magnetic contrast is too weak as to allow **M** distribution analysis, above 8 ML α is too small as to allow its determination in view of the width of the α distribution which is illustrated in Fig. 2 for the film region shown in Fig. 1. Two well-pronounced peaks at about $\pm 22°$ with a FWHM of about 13° are seen, with a significant contribution from $\alpha \approx 0°$. The most probable nonzero $|\alpha|$ value depends nonmonotonically on film thickness t as shown in Fig. 3. The error bars presently do not allow a distinction between an oscillatory and a stepwise decrease with t. The apparent double layer periodicity makes a spin-dependent quantum size effect

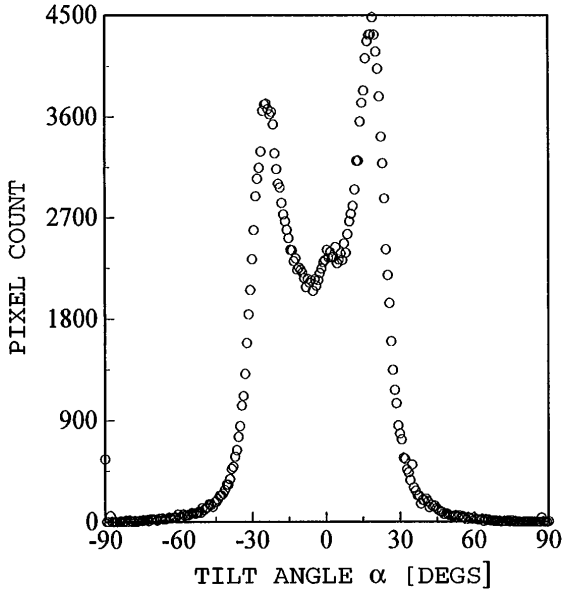

FIG. 2. Histogram of the **M** tilt angle distribution in the film shown in Fig. 1.

unlikely because the Fermi wavelength of the minority spin electrons $\lambda_F \approx 2.09$ nm [12] is incommensurate with the double layer thickness of 0.407 nm. It rather suggests a connection with the hcp stacking sequence $(ABAB\ldots)$ deduced from x-ray photoelectron diffraction measurements [8]. More work with improved image acquisition and more data points are needed to obtain a more accurate $\alpha(t)$ relation before a discussion of this point is warranted.

The origin of the wrinkled magnetization over a wide thickness range has to be sought in the competition between different anisotropies favoring in-plane and out-of-plane magnetization, respectively. In its simplest form, the free energy density of a hexagonal layer in the absence of an applied magnetic field may be written in the form [13]

$$F = 2\pi M_s^2 \cos^2\Theta + K_1 \sin^2\Theta + K_2 \sin^4\Theta , \qquad (1)$$

where Θ is the angle between the c axis (film normal) and **M**, M_s is the saturation magnetization, and $K_i = K_{ib} + K_{is}/t$ $(i = 1, 2)$ are the second and fourth order anisotropy coefficients. In K_{ib} all anisotropy contributions are subsumed which are proportional to t ("bulk" anisotropies); in K_{is} all contributions which are independent of t (surface anisotropies). Minimization of F with respect to Θ at constant t and temperature T gives the equilibrium **M** direction $\Theta = \pi/2 - \alpha$ as a function of t for given M_s, K_{ib}, and K_{is} values. Below a critical thickness $t_{c1} = K_{1s}/(2\pi M_s^2 - K_{1b})$, $\Theta = 0°$ (perpendicular magnetization), above a second critical thickness $t_{c2} = (2K_{2s} + K_{1s})/(2\pi M_s^2 - 2K_{2b} - K_{1b})$, $\Theta = 90°$ (in-plane magnetization). Provided that $t_{c1} < t_{c2}$, Θ changes from $t = t_{c1}$ to $t = t_{c2}$ as

$$\sin^2\Theta = \frac{2\pi M_s^2 - K_{1b} - K_{1s}/t}{2K_{2b} + 2K_{2s}/t} . \qquad (2)$$

The K values are still not known well enough so that Θ cannot be reliably predicted. It should also be noted that in the t range studied a surface type contribution $-4\pi M_s^2 t_d/t$ with $t_d = 0.18$ nm has to be added to the shape anisotropy term $2\pi M_s^2$ due to the thickness dependence of M_s [4]. The data in Fig. 3 indicate that $t_{c1} < 3$ ML and $t_{c2} > 8$ ML, in apparent disagreement with previous SPLEEM studies [3] and magnetometric measurements [4]. The first disagreement is easily explained by the fact that in the earlier SPLEEM work [4,9,10] only the in-plane component of **M** could be determined. The second disagreement is also only an apparent one: In the present work the virgin **M** distribution is studied, in Ref. [6] **M** as a function of applied field. Recent work [14] has shown that this leaves Co films in a metastable magnetic domain state. The original domain structure could approximately be obtained only by annealing but not by demagnetization.

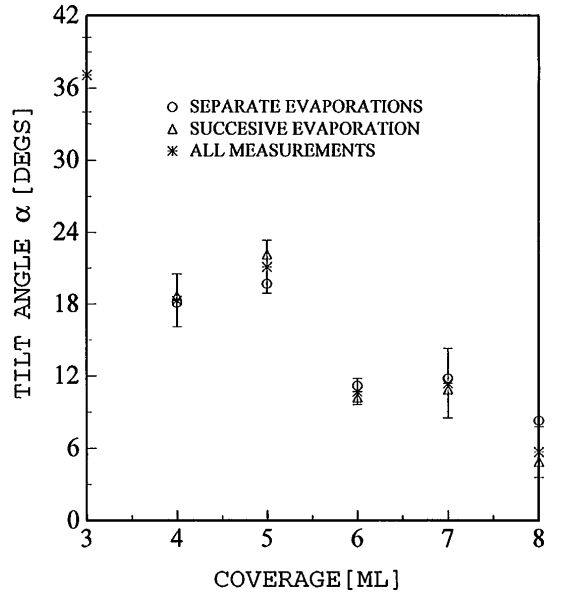

FIG. 3. Thickness dependence of the most probable nonzero tilt angle. Circles: individual deposits; triangles: cumulative deposits; stars: average values, taking into account error bars.

The canting of the magnetization can be attributed to the competition between the surface anisotropy of the system W(110)/Co/UHV and the shape anisotropy. The former has a large magnetoelastic contribution which actually is measured as a bulk contribution in the thickness range studied, in which the strain is constant [4]. It favors perpendicular magnetization as does the magnetocrystalline anisotropy of Co. With increasing thickness the shape anisotropy, which favors in-plane magnetization, increases due to the increasing magnetic moment [4] and as a consequence **M** rotates into the in-plane orientation.

The different length scales of regions with constant in-plane component $\mathbf{M}_\parallel$ and of regions with constant perpendicular component $M_\perp$ can also be explained

easily: For in-plane **M** the single domain state is energetically most favorable, for perpendicular **M** the striped or checkerboard domain phase. In the (t, T) range in which both components exist, that is in the SRT range, the regions with constant perpendicular $M_\perp$ become very small [15]. Although the striped phase has a lower energy than the checkerboard phase [15,16], local anisotropy fluctuations can strongly influence the domain pattern. Substrate surface steps which propagate in a quasi-ML-by-ML growth into the magnetic layer obviously create such fluctuations as seen in the domain patterns of Fig. 1. Because of the different interlayer distances of the W(110) and Co(0001) layers, the Co layers adjoining a step are displaced relative to each other by 10% of their interlayer distance. This causes a significant local change in the anisotropy coefficients which is apparently more important for the domain pattern than the long-range dipolar interactions.

In summary, we have observed canted magnetization in Co(0001) layers over a wide thickness range. The different length scales for the in-plane and perpendicular magnetization components lead to a wrinkled magnetization. The canting of the magnetization is not caused by (lateral) coexistence of in-plane and perpendicularly magnetized regions as assumed in recent work [2,17] but also occurs in homogeneous systems and is generally expected in the (t, T) range of the spin reorientation transition. According to model calculations [18], this transition should always be continuous in real systems, that is, in systems with magnetic surface anisotropy.

This work was supported by the Deutsche Forschungsgemeinschaft. Stimulating discussions with H.-J. Elmers, H. Fritsche, and U. Gradmann and critical reading of the manuscript by M. Scheinfein are acknowledged.

[1] R. Allenspach, J. Magn. Magn. Mater. **129**, 160 (1994).
[2] B. Dieny and A. Vedyayev, Europhys. Lett. **25**, 723 (1994).
[3] H. Pinkvos, H. Poppa, E. Bauer, and J. Hurst, Ultramicroscopy **47**, 339 (1992).
[4] H. Fritsche, J. Kohlhepp, and U. Gradmann, Phys. Rev. B **51**, 15 933 (1995).
[5] E. Bauer, Rep. Prog. Phys. **57**, 895 (1994); in Handbook of Microscopy, edited by S. Amelinckx, D. Van Dyck, J. F. Van Landuyt, and G. Van Tendeloo (VCH Verlagsges, Weinheim, to be published).
[6] T. Duden and E. Bauer, Rev. Sci. Instrum. **66**, 2861 (1995).
[7] M. Tikhov and E. Bauer, Surf. Sci. **232**, 73 (1990).
[8] H. Knoppe and E. Bauer, Phys. Rev. B **48**, 2675 (1993).
[9] H. Pinkvos, H. Poppa, E. Bauer, and G.-M. Kim, in *Magnetism and Structure in Systems of Reduced Dimensions*, edited by R. F. C. Farrow, B. Dieny, M. Donath, A. Fert, and B. D. Hermsmeier (Plenum Press, New York, 1993), p. 25.
[10] H. Poppa, H. Pinkvos, K. Wurm, and E. Bauer, in MRS Symposium Proceedings No. 313 (Materials Research Society, Pittsburgh, 1993), p. 219.
[11] M. Mundschau, E. Bauer, and W. Swiech, J. Appl. Phys. **65**, 581 (1989).
[12] Y. Gu and J. Noffke (private communication).
[13] C. Chappert and P. Bruno, J. Appl. Phys. **64**, 5736 (1988).
[14] M. Speckmann, H. P. Oepen, and H. Ibach, Phys. Rev. Lett. **75**, 2035 (1995).
[15] A. B. Kashuba and V. L. Pokrovsky, Phys. Rev. B **48**, 10 335 (1993).
[16] A. B. MacIsaac, J. P. Whitehead, M. C. Robinson, and K. De'Bell, Phys. Rev. B **51**, 16 033 (1995).
[17] I. Lyuksyutov and E. Bauer (unpublished).
[18] A. Moschel and K. D. Usadel, Phys. Rev. B **51**, 16 111 (1995); J. Magn. Magn. Mater. **140–144**, 649 (1995), and references therein.