Nanoscale magnetic domain structures in epitaxial cobalt films

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We have analyzed the magnetic domain structure in high quality molecular beam epitaxy (MBE) grown (0001)-hcp cobalt films of thicknesses varying from 10 to 500 nm for which the magnetic domain width is of the order of the film thickness. As the thickness of the film is increased, the magnetization turns from fully in plane (10 nm) to fully out of plane (50 nm) over a relatively large thickness range. Above 50 nm, the local domain imaging by magnetic force microscopy (MFM) together with overall magnetization measurements confirm the characteristic multidomain structure with perpendicular orientation predicted by Kittel. The periodicity of domains is then described by a $\sqrt{d}$ law (where d is the film thickness) that leads to the determination of domain wall energies. The occurrence of magnetic bubbles and periodic stripe patterns is visualized by variable field MFM. [S0163-1829(96)05429-X]

I. INTRODUCTION

A key issue in magnetic thin films is to find out under which conditions a film shows preferential orientation of the magnetization perpendicular to the film plane. This question was at the beginning of a great wealth of studies motivated mainly by the technological relevance of these films for data storage devices and sensors.[1–3] Recently, techniques like SEMPA (secondary electron microscopy with polarization analysis) (Refs. 1 and 2) and MFM (magnetic force microscopy) (Ref. 3) have been used to investigate films with perpendicular magnetization in the thickness range from 2 ML to several nanometers. These systems are usually dominated by a strong surface-interface anisotropy that holds the magnetization perpendicular to the plane of the film. As far as Co on Au (111) is concerned, it was shown from SEMPA that the domain structures in Co films a few monolayers thick are made of large irregular domains (typically 1 μm) with perpendicular ''up'' and ''down'' magnetization.[1,2] For Co films thicker than about 7 ML, the surface contribution is no longer sufficient to overcome the shape anisotropy term and the magnetization switches in plane.

As the film thickness increases, it was predicted by Kittel[4] on energetic arguments (competition between wall and shape energy) that for films of materials with a strong perpendicular magnetocrystalline anisotropy K, the magnetization may switch again out of plane. The predicted value of the film thickness d for which the magnetization turns from in plane to out of plane was calculated to be[4,5]

$$d_1 \approx 6.8\sigma_w \left(\frac{M_s}{K}\right)^2, \tag{1}$$

where M_s is the saturation magnetization and σ_w is the wall energy. When this condition is applied to hcp cobalt films, the crossover is predicted to occur at about 30 nm.[6] The only way for the system to lower its energy is then to split up into small domains with up and down magnetization. Therefore, these films are expected to show an interesting magnetic history with a magnetic bubble to stripe transition.[7–9] Macroscopic and mesoscopic arrangements of bubbles and stripes have been observed on a wide class of systems ranging from magnetic garnets to organic films. In all these systems, the patterns are stabilized by competing interactions leading to periodic variations of the order parameter.[10]

Magnetic domain structures have been observed by the Kerr effect in Co slabs 3–75 μm thick, cut into a single crystal with hexagonal c axis normal to the plane of the slice.[11] However only recently have films of sufficiently high crystallinity (to show the expected effects) been synthesized by MBE.[12,13] In a recent study Donnet *et al.*[12] analyzed the magnetic domain structure of MBE-grown Co films in a narrow range of thicknesses (20–60 nm), close to the predicted transition. Interestingly, from their Lorentz microscopy study in the phase contrast mode, they found that a 40-nm film exhibits periodic stripe domains whose weak contrasts are produced by an alternatively up and down deviation of the mainly in plane magnetization.

This paper is devoted to the detailed MFM investigation of magnetic domains of hcp Co films grown by MBE. Up to 11 samples have been analyzed in the thickness range from 10 to 500 nm. We have identified the presence of bubbles and periodic stripes with perpendicular magnetization for thicknesses of 50 nm and above as a function of magnetic history. To our knowledge, this is the first MFM investigation of Co films in this thickness range. The smallness of the observed domains shown in our work (of the order of the film thickness) clearly demonstrate the power of MFM. Their width D was found to obey the predicted $D \propto \sqrt{d}$ dependence.[4]

II. EXPERIMENT

Cobalt films with thicknesses varying from 10 to 500 nm were prepared by molecular-beam epitaxy at 400 °C. An initial 20-nm-thick Ru layer was deposited onto an $(1,1,-2,0)$ sapphire substrate, the Co film of thickness d was then deposited and capped by another 5-nm-thick Ru layer. The crystallographic structure of the films was studied *in situ* by reflection high-energy electron diffraction (RHEED) and *ex situ* by x-ray diffraction. The RHEED patterns obtained during the growth of the film reveal well-defined rods which

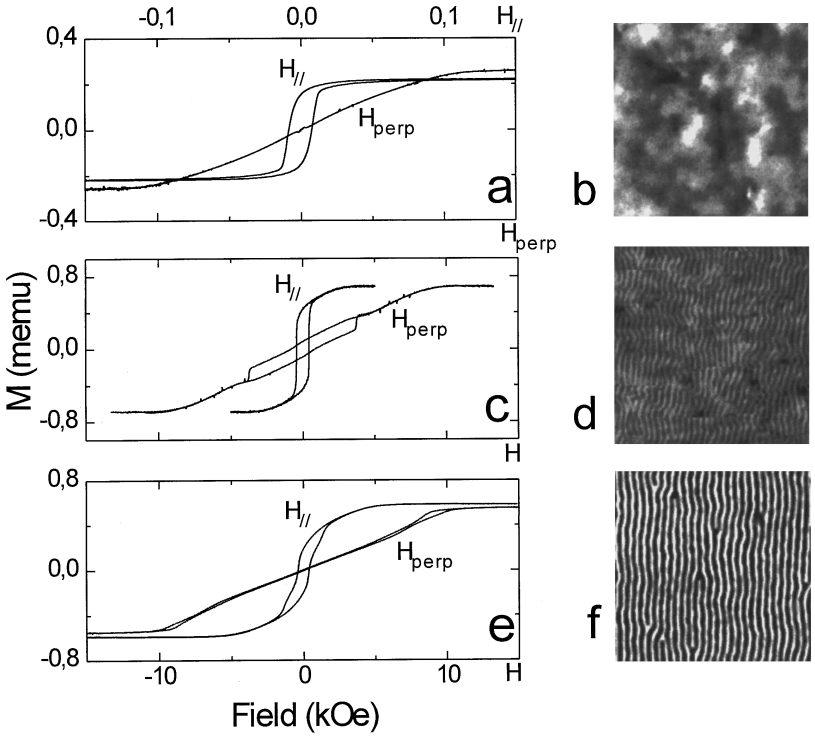

FIG. 1. *M-H* curves in a configuration with the magnetic field perpendicular and parallel to the plane of the film: (a) 10 nm, (c) 25 nm, (e) 50 nm. Corresponding MFM images 3 μm$\times$3 μm of films previously demagnetized in plane: (b) 10 nm, (d) 25 nm, (f) 50 nm.

suggests good surface crystalline quality and a smooth surface. Using three-dimensional (3D) RHEED diffraction analysis (relative position of the spots on the rods), we observe that the free surface crystal structure is mostly hcp (0001). $\Theta-2\Theta$ x-ray-diffraction scans of the (222)fcc/(0004)hcp Bragg peak revealed one peak located at the expected angle for the hcp stack. The dominant (0001)hcp structure gives rise to a large perpendicular magnetocrystalline anisotropy of $K=4.6\times10^6$ erg/cm^3 which locks the magnetization perpendicular to the layer. The average roughness over the whole surface was estimated to be 1.5 nm by atomic force microscopy.

Magnetic force microscopy was shown to be a very efficient technique to study a wide range of micromagnetic problems.[14,15] Up to now, most of the knowledge about this novel technique has been gained from a systematic study of magnetic recording media where well-defined magnetic information can be inscribed in magnetic films.[14] In particular, it has been shown that previously developed analytical forms for domain walls[16] could be applied to the description of MFM data.[17] On the other hand, tip effects have been addressed and from experiments on well-controlled samples it was even possible to determine the orientation of the tip magnetization.[18]

The domain structure has been observed in zero and finite fields up to $|H|=500$ Oe by MFM. We used a CoCr coated Si tip magnetized along the tip axis in a permanent magnet. A Nanoscope III equipped with a magnetic tip, scanned 40 nm above the surface, was used in the interlace mode developed by Digital Instruments. This mode allows one to disentangle the long-range magnetic and the short-range topographic information during the same image acquisition. Since the detected signal (frequency shift of the vibrating cantilever) is proportional to the second derivative of the local field, this technique provides a good signal over noise ratio, unfortunately it does not allow the local field information to be recovered easily. The interpretation is simplified in the case of samples with perpendicular magnetization, since only contrasts from magnetic walls between ''up'' and ''down'' domains are visible by MFM. Independent magnetization measurements were performed for all samples at room temperature in an alternating gradient field magnetometer (AGFM) with the field applied perpendicular and parallel to the film plane.

III. RESULTS

A. From 10 to 50 nm: The transition

We first investigate the reorientation of the magnetization direction from in plane to out of plane as the thickness of the film increases from 10 to 50 nm. Figure 1 shows a first set of magnetization curves, with the field applied either perpendicular or parallel to the plane of the film, measured by AGFM together with the corresponding zero-field MFM images.

The perpendicular magnetization curve obtained for the 10-nm-thick film [Fig. 1(a)] is characteristic for a film with in-plane magnetization.[19] The corresponding MFM image shows large domains typically 1 μm wide [Fig. 1(b)]. On the other hand, the perpendicular *M-H* curve for the 50-nm-thick film [Fig. 1(e)] is characteristic of perpendicular alternatively up and down stripe domains[9] for which the magnetization in the middle of the stripe is fully perpendicular to the film. Interestingly, the in-plane hysteresis loop in Fig. 1(e) shows considerable remanence, which we attribute to the magnetization at the center of the walls, turning from one in-plane direction to the opposite in-plane direction when sweeping the field from one direction to the other.[20] The characteristic domain structure shown in the MFM image of Fig. 1(f) has been evidenced for all thicknesses above 50 nm and will be discussed together with the full MFM interpretation in the next section.

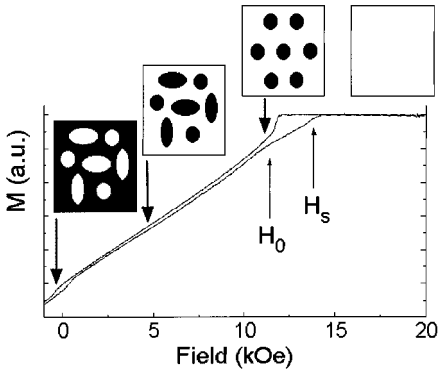

FIG. 2. Positive field branch, of the perpendicular M-H characteristics of a 150-nm-thick Co film. The nucleation field of bubbles H_0 as well as the saturation field H_s are indicated by arrows. The insets show schematically the magnetic domain contrasts.

The 25-nm-thick film depicts an intermediate regime. A hysteresis loop of extension ± 4 kOe opens up into the core of the perpendicular M-H curve as shown in Fig. 1(c), while the corresponding MFM image [Fig. 1(d)] already exhibits a weak stripe domain structure characteristic for a nonzero perpendicular component. While MFM provides the periodicity of the domain structure, it hardly informs on the profile of the magnetization across the domains. Our results however are quite similar to those obtained for a 40-nm-thick Co film grown under similar conditions,[12] where the hysteresis has been shown (on the basis of Lorentz microscopy), to correspond to a mainly in-plane magnetization with a small perpendicular component, oriented alternatively up and down.[12,21] Within a simple model[21] the thickness at which the weak stripe domain structure first appears is given by

$$d_2 \approx 27\left(\frac{8}{\pi^2}\right)^2\left(\frac{AM_s^4}{K^3}\right)^{1/2}, \qquad (2)$$

where A is the exchange constant. It is equal to 20 nm for a hcp cobalt film in good agreement with our experimental results. As can be seen from our figures, the MFM contrast for the 25-nm-thick film is weaker than for the 50-nm-thick film, denoting that the main component of magnetization is still in plane.

B. From 50 to 500 nm

In Fig. 2 we show a detail of the magnetization curve for the 150-nm-thick sample. The field is applied in the easy direction perpendicular to the surface of the film. The curve of Fig. 2 appears like a signature of domains with perpendicular magnetization and can be understood quite well in the model developed by Cape and Lehmann[7] for films with an easy axis perpendicular to the film plane (see also an earlier work by Kooy and Enz[9]). By describing the hysteresis loop counterclockwise from the high-field saturation, where all magnetic moments are oriented in the field direction, towards lower positive fields, a singularity of the M versus H curve appears at some point H_0. This singularity is ascribed to a sudden nucleation of magnetic bubbles with opposite magnetization. It is due to the existence of a critical bubble radius R_0 (H_0) under which a bubble network does not form a stable configuration. Thiele[8] has shown that the nucleation field is subject to geometrical constrains, namely that H_0 is a growing function of the film thickness. This fact nicely shows why in a real systems, where the film thickness may vary from one point to another, a dispersion in nucleation fields may occur, resulting in a smoothening of the transition. Note in Fig. 2 the narrow range of fields (300 Oe) over which the nucleation of bubbles spreads throughout the whole film.

By further decreasing H, the bubbles grow larger while the lattice parameter of the bubble network shrinks.[7] The problem now becomes one of wall motion that also explains the quasilinear decrease of M with H.[9] It was shown by Thiele[8] that above a given radius R_1 the bubbles tend to adopt an elliptic shape which is the precursor state of stripes. Interestingly, the repulsive interaction between the bubbles[22] may prevent their growth as H decreases so that in some cases bubbles may be the stable configuration even at $H=0$ as is confirmed by our MFM measurements on thick Co films [Fig. 3(c)]. The dependence of saturation fields and nucleation fields on the film thickness has been analyzed in detail elsewhere.[13] From the saturation field in the parallel configuration we were able to find an estimate of the perpendicular demagnetizing field which is close to zero for a 500-nm-thick Co film, confirming independently the perpendicular easy-axis interpretation. Furthermore, the onset of fully perpendicular magnetization was found to be 50 nm determined both from the appearance of a kink in the M-H curve and from the detection of stripes by MFM in good agreement with the prediction.[7]

C. Interpretation of MFM data

To achieve an unequivocal interpretation of our MFM data, we calculated the second derivative of the local field in the z direction for various domain configurations,[23] assuming that the tip can be approximated by a dipole (which is reasonable in first approximation), and taking the magnetization across the wall to be of the form arctan (x/w), where w is the wall width. Figure 4 shows the calculated MFM response to a stripe domain structure of normalized width D/d where d is the film thickness. The calculation was done for a tip magnetization perpendicular to the surface and $(z+w)/d = 0.5$, where z is the tip to surface distance. Similar profiles have been obtained for bubbles.[23] Contrasts are always due to domain walls, however, real experimental situations cover the range $D/d = 0.3$–1.5 for which the walls are so close to each other that they produce one single bright or dark area centered on the domain itself. Therefore the contrasts in Fig. 3 may be unequivocally identified as corresponding to domains (not to domain walls). That contrasts are identical to domains is seen quite well on the images on the basis of topological arguments: in Figs. 3(a) and 3(b), bright stripes that come to an end in an otherwise dark environment are virtually forbidden in a domain-wall type of interpretation. Unfortunately, wall widths cannot be determined easily by fitting the data since w and z take on the same status in the equations. Also small magnetic domain closures at the surface[24] would hardly be detectable by MFM.

In order to analyze the samples by MFM, we have performed both magnetization and demagnetization experiments in the direction either *perpendicular or parallel to the plane of the film*. During the *parallel demagnetization* the field is

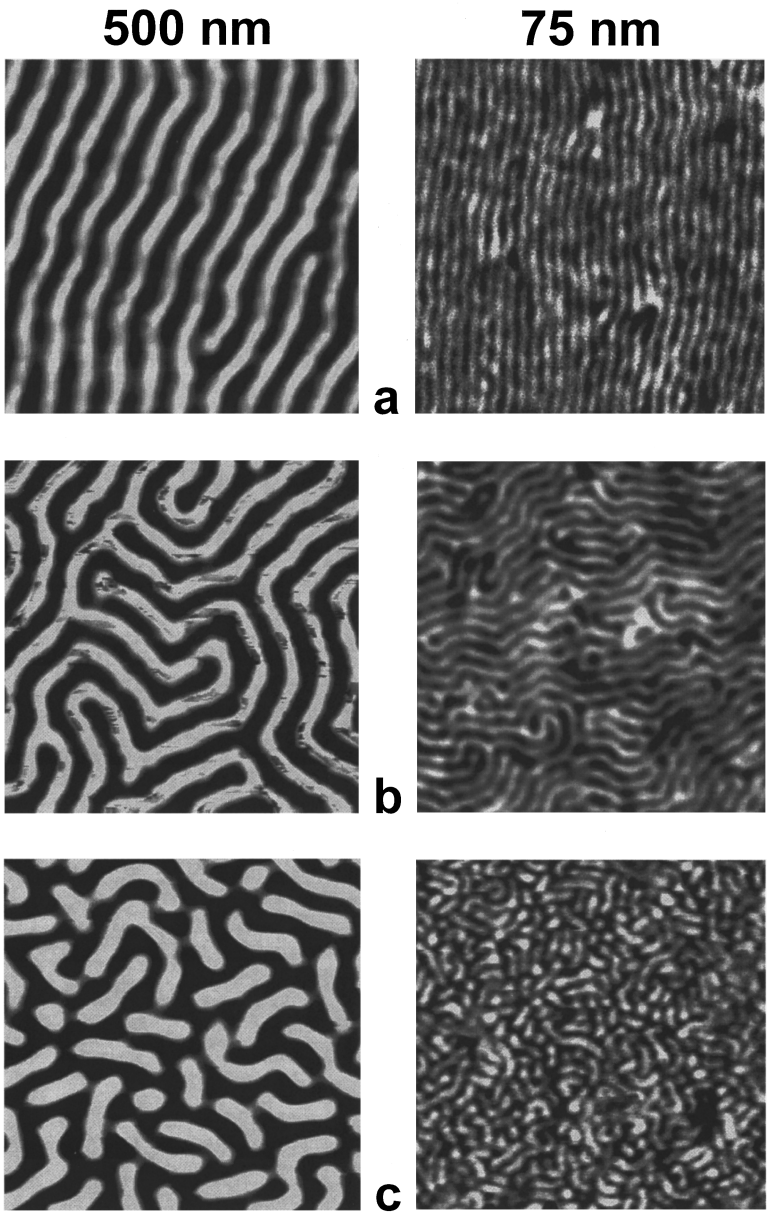

FIG. 3. Zero-field MFM images 5 μm$\times$5 μm. (a) Demagnetized parallel, (b) demagnetized perpendicular, and (c) magnetized perpendicular states for 500- and 75-nm-thick Co films.

swept from positive to negative values while reducing simultaneously the area of the hysteresis loop. The magnetic moments first follow the field direction and the system evolves in a quasistatic way. At some small value of the magnetic field, the magnetic moments get trapped out of plane (due to the strong crystalline anisotropy). The alignment of the parallel component of the magnetization with the field in order to minimize the Zeeman energy leads to a periodic pattern of parallel stripes [Fig. 3(a)] predicted by Muller.[24] Qualitatively similar results are obtained for the *parallel magnetization*, in which the sample is brought to saturation and the field turned off. MFM images reveal evenly spaced parallel stripes oriented in the direction of the applied field (not shown). The additional degree of freedom introduced during *perpendicular demagnetization* leads to labyrinthine stripes [Fig. 3(b)]. Finally, a quite different situation is obtained for *perpendicular magnetization* [Fig. 3(c)], where the remnant state is reminiscent of the elliptic bubbles predicted by Thiele.[8] Although these films have a complicated magnetic behavior it is generally admitted that the stripe state is ener-

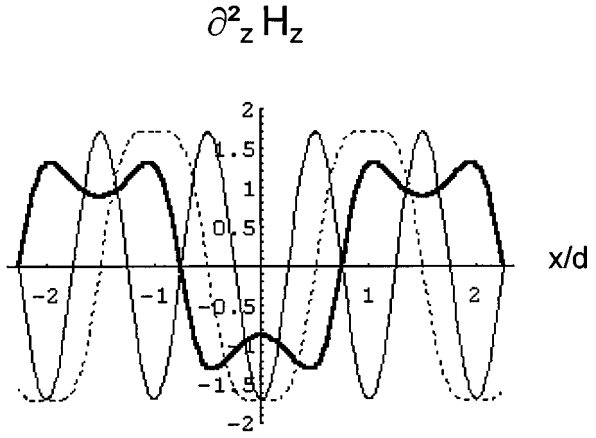

FIG. 4. MFM response to a perpendicularly magnetized stripe for normalized widths (—) $D/d=0.5$, (---) $D/d=1.0$, (—) $D/d=1.5$. The calculation has been done for $(z+w)/d=0.5$. Real experimental situations cover the range $D/d=0.3$–1.5.

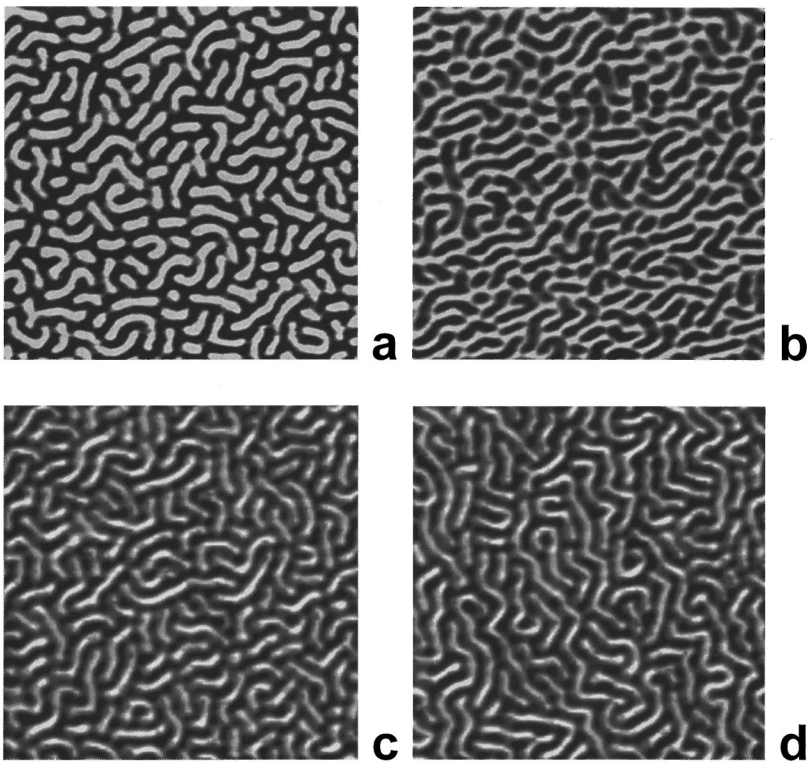

FIG. 5. MFM images 10 μm×10 μm for 500-nm Co films. At finite field values on the major loop (a) $H=+400$ Oe, (b) $H=-400$ Oe. At $H=0$ after interrupted perpendicular demagnetization at (c) 13 kOe and (d) 12 kOe, respectively.

getically more favorable at $H=0$.[7] This is clearly what we observe after *parallel* [Fig. 3(a)], as well as after *perpendicular* [Fig. 3(b)] *demagnetization*. While in the first case, the symmetry breaking along an in-plane direction forces the stripes to align in one direction, in the latter case, only the local order is preserved and free-energy minimization leads to a globally disordered pattern. It is the unbinding of disclination defects, visible in the Y shape of domain bifurcation in the stripe pattern of Fig. 3(b) ($d=500$ nm), that preludes to disorder. The threefold symmetry comes about because 120° chevrons form the precursor state for the labyrinthine structure.[10]

D. Variable field MFM

Whether stripe domains occur or not along the major hysteresis loop is the next question we tried to address using variable field MFM. We found that the stripe pattern never occurs. Instead we show that when a major loop is described counterclockwise from large positive fields to negative fields, bubbles of originally bright contrasts (oriented opposite to the applied field) begin to expand lengthwise so as to touch each other and to coalesce. This is the point where the identity of dark bubbles emerges in an otherwise bright background. Therefore the transition from bright to dark bubbles proceeds continuously in a wall motion type of procedure. MFM images of Figs. 5(a) and 5(b) have been taken on the major loop at fields of $+400$ and -400 Oe, respectively, they clearly show bubbles with opposite contrast. It is only during field cycling to minor loops that more ramified patterns are observed. How bubbles go over to stripes during the demagnetization procedure is nicely shown for 500-nm films ($H_s=16$ kOe) in Figs. 5(c) and 5(d) where the perpendicular field has been interrupted at two different values of the maximum field, namely 13 and 12 kOe leaving behind a remnant state. The bubbles are first subject to elongation and bending [Fig. 5(c)] only then does domain branching start [Fig. 5(d)]. The pattern obtained at 9 kOe (not shown) is already very close to the one of Fig. 3(b) for demagnetization down to zero field. The labyrinthine structure resulting from the *perpendicular demagnetization* is seen over the whole thickness range from 50 to 500 nm. This experiment shows that the stripe and bubble patterns must be very close in energy at $H=0$ as anticipated from the analysis.[7]

E. Thickness dependence of the domain width

In Fig. 6, we have reported the period L of the stripelike patterns observed in Figs. 3(a) and 3(b) as well as the one

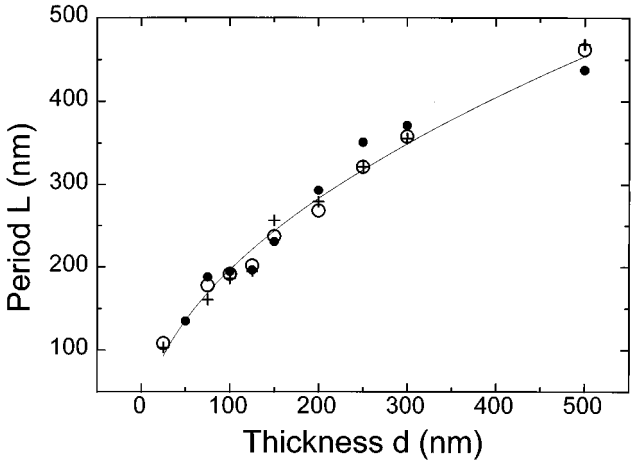

FIG. 6. Period L of the stripe pattern as a function of Co film thickness d (see text). (○) demagnetized parallel, (●) demagnetized perpendicular, (+) magnetized parallel.

from parallel magnetization (not shown) as a function of film thickness d. On the basis of minimum energy arguments, Kooy and Enz[9] found the following analytical expression for the period:

$$L^2 = 2\pi^2 d\sigma_w(1+\sqrt{\mu})/(16M_S^2\varepsilon), \tag{3}$$

where $\mu = l + 2\pi M_S^2/K$; σ_w is the wall surface energy; and ε is a numerical factor that depends on μ, on the film thickness d, and on the period L [see Eq. (10) of Ref. 9]. For our ranges of parameters, this leads to $\varepsilon = 1.04 \pm 0.01$ which is put equal to a constant in the calculation. This expression fits all three states well (solid line in Fig. 6) indicating that domain periods do not depend significantly on the magnetic state at $H=0$. The wall energy is found to be $\sigma_w = 25 \pm 3$ erg cm^{-2} and does not vary significantly with the film thickness. This value is in good agreement with recent 2D micromagnetic calculations performed for a 100-nm-thick Co film for which energies of about 20 erg/cm^2 are found.[25]

IV. CONCLUSION

Using both, conventional magnetometric measurements and local MFM domain imaging, we have identified a continuous reorientation of the magnetization in thin epitaxial cobalt films from in plane to out of plane for thicknesses comprised 10 and 50 nm. For film thicknesses above 50 nm we found that a continuous bubble-to-bubble transition through irreversible wall motion occurs along the major hysteresis loop when the field is applied perpendicular to the film. The earlier predicted lowest-energy periodic stripe pattern is obtained only after an appropriate demagnetization procedure, and the nanoscale domain width is comparable in size to the film thickness. The periodicity of stripelike structures is then found to obey a $L \propto \sqrt{d}$ law in good agreement with theoretical predictions. In a forthcoming paper we will show how a confinement of domains by geometrical boundaries down to nanometer scale modifies the magnetic properties of epitaxial Co films.

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