Remanence due to Wall Magnetization and Counterintuitive Magnetometry Data in 200-nm Films of Ni

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200-nm-thick Ni films in an epitaxial Cu/Ni/Cu/Si(001) structure are expected to have an in-plane effective magnetic anisotropy. However, the in-plane remanence is only 42%, and magnetic force microscopy domain images suggest perpendicular magnetization. Quantitative magnetic force microscopy analysis can resolve the inconsistencies and show that (i) the films have perpendicular domains capped by closure domains with magnetization canted at 51° from the film normal, (ii) the magnetization in the Bloch domain walls between the perpendicular domains accounts for the low in-plane remanence, and (iii) the perpendicular magnetization process requires a short-range domain wall motion prior to wall-magnetization rotation and is nonhysteretic, whereas the in-plane magnetization requires long-range motion before domain-magnetization rotation and is hysteretic.

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Magnetic thin films with perpendicular anisotropy provide a model system of domain behavior in a wide variety of materials. Many (material) systems exhibit maze domain patterns that evolve to stripe domains, then large-scale domain growth upon application of an appropriate force [1,2]. Hysteresis characterizes some of these processes. These features are observed in a unique and revealing way in the magnetic thin film system studied here. The observations defy explanation based on volume averaged magnetometry measurements but come to light when magnetometry is combined with high-resolution, quantitative magnetic force microscopy.

In this Letter, we explain how the conventional interpretation of the magnetization loops can give an incorrect picture of both the magnetization processes and magnetic domain structures in the films. The system studied here is a 200-nm Ni film grown epitaxially in a Cu/Ni/Cu/Si(001) structure. This system has been studied previously [3–6] as a model for the technologically relevant phenomenon of perpendicular magnetization. We begin by noting that, for small Ni thickness t , $3 \text{ nm} < t < 14 \text{ nm}$, a strong perpendicular magnetization is observed by magneto-optic Kerr effect [3], vibrating sample magnetometry (VSM) [4], torque magnetometry and magnetic force microscopy (MFM) [5,7]. It is possible to account for these observations using a uniform magnetization model with effective uniaxial anisotropy for the free energy $f = K_{\text{eff}} \sin^2 \theta$. The coefficient K_{eff} includes contributions from the magnetoelastic and interface anisotropies [3–5,8] (>0) and the magnetostatic energy $K_{\text{ms}} (= -\frac{1}{2} \mu_0 M_s^2$, where M_s is the saturation magnetization and μ_0 is the vacuum permeability). The magnetocrystalline anisotropy K_{mc} is small and can be neglected. The magnetoelastic anisotropy K_ϵ arises from the Cu/Ni misfit, and a surface anisotropy K_s is

evident for each Ni/Cu interface. When $K_{\text{eff}} > 0$, the perpendicular magnetization is stable in a field $H = 0$.

The above model correctly describes a broad range of perpendicular magnetization observed in the aforementioned references, as well as the thickness dependence of the effective anisotropy. At 200-nm Ni thickness, it predicts an in-plane magnetization [9].

Our Cu/Ni/Cu/Si(001) films were made in a molecular-beam epitaxy machine with a base pressure in the 10^{-9} Pa range. The first Cu layer on the Si(001) substrate is 50 nm thick. Ni grows epitaxially on this layer. The 200-nm Ni film is capped with 5 nm of Cu. Lattice mismatch between Ni and Cu results in strain, which has been measured by synchrotron x-ray scattering [8].

The MFM results were obtained at room temperature and in a vacuum in the 10^{-5} Pa range [10]. A Si cantilever with a force constant $c = 0.2 \text{ N/m}$ and a resonance frequency $f_0 = 12.3 \text{ kHz}$ was used. One side of the tip was coated with iron and gold. Subsequently, the tips were magnetized in a field of $\approx 0.4 \text{ T}$ oriented along the tip axis. The MFM measurements were made at a constant tip-sample distance, in the dynamic variable frequency mode, which determines the shift δf of the cantilever resonance frequency f_0 [11]. Magnetization structures (M_z) can be inferred from the stray fields (H_z) [12] with knowledge of the instrument calibration or the tip transfer function $\sigma_{\text{tip}}^*(\mathbf{k})$, which was determined as described elsewhere [11].

The magnetization loop $\mathbf{M}(\mathbf{H})$ for the 200-nm film is displayed in Fig. 1. Parameters of this film are indicated in Table I. As can be seen from the figure, $M(H_\perp)$ has the form of a hard axis loop with zero remanence, saturating at $H_\perp^{\text{sat}} = 380 \pm 20 \text{ kA/m}$. It indicates a net uniaxial anisotropy $K_{\text{eff}} = -\frac{1}{2} \mu_0 M_s H_\perp^{\text{sat}} = -115 \text{ kJ/m}^3$, favoring in-

FIG. 1. Room temperature VSM measurements of the magnetization versus applied field in a Cu/Ni/Cu/Si(001) sample with a Ni thickness of 200 nm. Fields $H_{\perp}$ and $H_{\parallel}$ are applied, respectively, perpendicular to the film or in-plane (either [100] or [110] with identical results).

plane magnetization. Comparison of this value with the effective anisotropy energy ($K_{\text{eff}} \approx K_{\epsilon} - \frac{1}{2} \mu_0 M_s^2 = -100 \text{ kJ/m}^3$ —cf. Table I) supports a homogeneous magnetization description based on K_{eff} . The in-plane magnetization loop $M(H_{\parallel})$ saturates more easily than the perpendicular loop, as is expected for an easy magnetization direction. Furthermore, $M(H_{\parallel})$ displays remanence and coercivity (of about 42% and $16 \pm 2 \text{ kA/m}$, respectively), both of which suggest domain wall pinning. Accordingly, the conventional uniform magnetization interpretation of this magnetization loop is that the film has in-plane anisotropy. However, upon careful observation, several inconsistencies in this interpretation appear, which ultimately demand its change.

Notice that if the plane of the film is “easy,” then an applied field that saturates the magnetization in-plane produces a configuration which essentially minimizes the anisotropy energy. Except for rather small demagnetization fields, removing the applied in-plane field should not destabilize this in-plane magnetization. That is, a large remanence would be expected. Yet a remanence of only 42% is determined experimentally (Fig. 1), indicating that a substantial fraction of the magnetic moments is swinging away from the previously saturated direction. In fact, the magnetization swings out of the plane, as is readily seen in the MFM measurements in Fig. 2. These images of as-received film [maze pattern, Fig. 2(a)] and of films pre-

FIG. 2 (color online). MFM images of Cu/Ni/Cu/Si(001) structures with a Ni thickness of 200 nm. (a) As-received film in the absence of an applied field. (b) Image of the film relaxed to zero field after saturation in-plane (along $\hat{x}$).

viously saturated in-plane and allowed to relax to zero field [stripe pattern, Fig. 2(b)] suggest perpendicular magnetization and an anisotropy energy which is not minimal in the plane.

Quantitative MFM [7] can be used to find the magnetization structure which fits the measured frequency shift pattern $\delta f(M(x, y))$ in the images. In this technique, candidate structures are assumed, and their $\delta f(M(x, y))$ pattern is simulated (i.e. calculated with the calibrated transfer function) and then compared with the measured image to determine which provides the best match. For reference, Figs. 3(a)–3(c) show this technique applied to 7-nm Ni films, which are known to have perpendicular anisotropy [7]. Good agreement assuming perpendicular domains through the thickness is apparent. Using the same method of analysis for the 200-nm film, the results of Figs. 4(a)–4(c) are obtained. In this case, although the shape of the pattern is well reproduced by the simulation, the simulated contrast amplitude is too big (overall by a factor 1.58); i.e. $\mathbf{M}$ is not perfectly perpendicular. Different assumptions degrade or improve the match. Simulations of magnetization ripples, where the magnetization oscillates out of the plane with amplitude $\alpha(y) \approx \alpha_{\text{ripple}} \sin(k_r y)$, can match the range of measured contrasts for a ripple angle $\alpha_{\text{ripple}} \approx 39^\circ$.

FIG. 3. Measurement and simulation of the magnetization of Cu/Ni/Cu/Si(001) structures. (a) MFM image for 7-nm-thick Ni, measured at $31 \pm 1 \text{ nm}$ tip sample distance. (b) Image simulated from perpendicular domains obtained from (a) with the calibration function $\sigma_{\text{tip}}^*(\mathbf{k})$. (c) Comparison of the sections indicated in (a) (solid line) and (b) (dashed line).

TABLE I. Material parameters (room temperature).

Sat. magnetization	$\mu_0 M_s$	0.603 T
Magnetostatic energy	$-\mu_0 (M_s^2/2)$	-145 kJ/m^3
Magnetocryst. anisotropy	K_{mc}	-4.5 kJ/m^3
Mag. elastic anisotropy	K_{ϵ}	$45 \pm 1 \text{ kJ/m}^3$
Magn. coupl. coeff.	B_1	6.2 MPa
Strains	$e_{xx} = e_{yy}$	$+3.18 \times 10^{-3}$
	e_{zz}	-3.8×10^{-3}
Bloch wall width	$\delta_{\text{dw}} = \pi \sqrt{A/K_{\epsilon}}$	$47 \pm 1 \text{ nm}$

FIG. 4 (color). (a) MFM image for a 200-nm-thick Ni film. (b) Simulation assuming perpendicular domains obtained from (a). (c) Comparison of the sections indicated in (a) and (b). (d) Histogram of image intensities for (a) and (b), a simulation assuming a ripple pattern with $\alpha_{\text{ripple}} = 39^\circ$, and simulation (b) with the contrast adjusted by $1/1.58$, as per domain closures.

(from in-plane). However, due to the $\sin(k_y y)$ dependence, ripples are inherently incapable of reproducing sharp contrast changes observed in the MFM data. On the other hand, perpendicular domains capped with closure domains [13–15] having a canting angle of $\approx 51^\circ$ off the plane normal produce the same pattern as through-thickness perpendicular domains (due to the linearity of the transfer function) and do give the appropriate amplitude.

The above discussion is summarized in the δf histogram in Fig. 4(d). It shows that the domain pattern generated by perpendicular domains, adjusted by a factor $1/1.58$ (corresponding to closures canted at 51°) closely matches the measured histogram, whereas the ripple simulation has an intermediate frequency shift level that is too large, in accordance with their “fuzzy” magnetization profile. In conclusion, the film is shown to have perpendicular domains capped with canted closure domains. This quantitative result complements earlier model results indicating that closures would be consistent with observed domain widths [15].

The perpendicular anisotropy in 200-nm films thus having been established, our previous interpretation of Fig. 1 must be revisited. Indeed, it is recognized that $M(H_\perp)$ must be a combination of short-range domain wall motion processes and closure magnetization rotation, whereas $M(H_\parallel)$ must result from long-range rearrangement of domain walls and domain-magnetization rotation. Further clarification is required because coercivity and high remanence are expected for domain wall motion but not for magnetization rotation. The greater difficulty (smaller susceptibil-

FIG. 5. Schematic of the magnetization in the film's Bloch walls. A net magnetization in the x direction could exist.

ity) of the perpendicular magnetization of the film via domain wall motion also merits discussion.

In addressing the first point, consider the film saturated in-plane along $\hat{x}$ as it relaxes to zero field [Fig. 2(b)]. The magnetization of the closures lies in the yz plane and cannot give rise to remanence in $\hat{x}$ (a slight canting toward $\hat{x}$ over the underlying Bloch wall could be possible, but minor). Furthermore, the symmetry of the closures requires that their magnetization have a vanishing average in the direction perpendicular to the domains. Closures therefore cannot be the primary cause of remanence, as other authors have suggested [15].

On the other hand, the magnetization of the Bloch walls inside the film imaged in Fig. 2(b) lies in the xz plane, for the most part. This is shown schematically in Fig. 5. As is clear from this figure, each wall has a net contribution to the remanence along $\hat{x}$, which can be positive or negative but is not prescribed by symmetry to take either value. However, when the magnetization relaxes from saturation in $+\hat{x}$, practically all the magnetic moments in the walls will have a positive component along $\hat{x}$. The resulting remanence can be large given the wall density of the present film. Integration of $\mathbf{M} \cdot \hat{x}$ over a domain width $D \approx 125$ nm yields $M_x = M_s(\delta_{\text{dw}}/D) = 38\%$ (cf. Table I), essentially matching the measured remanence of 42% and in agreement with magnetoelastic stress results [6]. Thus, the remanence is not a net magnetization of the domains: It is the net magnetization of the *domain walls*.

FIG. 6. Macroscopic return point memory. Total magnetization level as percent black (from MFM images [17] normalized to point 8) after successive field excursions (labels in T).

TABLE II. Energy for saturation.

Estimate	Expected	Measured
$\Delta U_{\perp} \approx \frac{1}{2} \mu_0 M_s^2 - \sigma_{\text{dw}}/D$	123 kJ/m ³	$E_{\perp} = 121 \pm 3 \text{ kJ/m}^3$
$\Delta U_{\parallel}^+ \approx K_{\epsilon} - \sigma_{\text{dw}}/D$	23 kJ/m ³	$E_{\parallel}^+ = 21 \pm 3 \text{ kJ/m}^3$
$\Delta U_{\parallel}^- \approx K_{\epsilon} - \sigma_{\text{dw}}/D$	23 kJ/m ³	$E_{\parallel}^- = 32 \pm 3 \text{ kJ/m}^3$

Notice that in a maze pattern such as Fig. 2(a) the net in-plane magnetization averages out macroscopically. A transition to this state from Fig. 2(b) has a very low driving force (small demagnetization fields) while requiring a large reconfiguration of the walls. This accounts for the nonzero coercivity observed in the in-plane magnetization loop.

In contrast to the in-plane loop, the out-of-plane magnetization loop displays no significant remanence or coercivity. Domain wall motion and macroscopic return point memory [16] are confirmed via MFM in Fig. 6. It is apparent that field excursions up to $\mu_0 H^{\perp} = 0.4 \text{ T}$ can move the domain walls so that they can recover the initial magnetization when $H^{\perp}$ is removed. Domain wall pinning does not result in appreciable coercivity here because a large magnetization change can occur by a relatively small displacement of (many) walls between or around pinning centers. These conclusions do not hold for films with larger domains (thickness around 10 nm and below).

The work required to saturate in- or out-of-plane can be quantified from Fig. 1 as $E_{\parallel}$ or $E_{\perp}$, respectively. As well, the energy differences $\Delta U_{\perp, \parallel}$ between zero-field and saturated states can be estimated. In-plane saturation results in peak anisotropy energy K_{ϵ} and negligible magnetostatic energy. Out-of-plane saturation results in the reciprocal case of negligible anisotropy energy and maximum magnetostatic energy. In a remanence state, the energy is the wall energy. Table II compares $\Delta U_{\perp, \parallel}$ with $E_{\perp, \parallel}$ (domain spacing $D = 125 \text{ nm}$). Here the superscripts + and – refer to positive and negative remanence in the initial state. A reasonable agreement is apparent, despite having neglected the energy of the closures in the initial state and possibly also additional components in $\Delta U_{\parallel}^+$ from the removal of wall magnetization opposing the applied field.

In conclusion, the Cu/Ni/Cu/Si(001) structure with a Ni thickness of 200 nm studied in this Letter is a thin film example which defies a conventional interpretation. The film has perpendicular anisotropy, but the out-of-plane direction is hard to magnetize because of the large magnetostatic energy. 125-nm-spaced perpendicular domains with closure domains canted at 51° exist in the demagnetized state. Out-of-plane magnetization occurs by domain wall motion and magnetization rotation, but it has essentially no remanence or hysteresis because the domain width is small, and large magnetization changes can take place by small shifts in the domain walls. The in-plane magnetization for small fields occurs by rearrangement of domain walls over ranges larger than the domain wall spacing. For large fields, it occurs by magnetization rota-

tion. It involves coercivity and hysteresis. A 42% remanence observed upon relaxing an in-plane saturation is due almost entirely to the in-plane wall magnetization of the perpendicular domains.

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- [1] D. P. Jackson and B. Gantner, Phys. Rev. E **64**, 056230 (2001).
- [2] Th. Eimüller, R. Kalchgrubber, P. Fisher, G. Schütz, P. Guttman, G. Schmahl, M. Köhler, K. Prügl, M. Scholz, F. Bammes, and G. Bayreuther, J. Appl. Phys. **87**, 6478 (2000).
- [3] R. Jungblut, M. T. Johnson, J. van de Stegge, A. Reinders, and F. A. den Broeder, J. Appl. Phys. **75**, 6424 (1994).
- [4] G. Bochi, C. A. Ballentine, H. E. Inglefield, C. V. Thompson, R. C. O'Handley, H. J. Hug, B. Stiefel, A. Moser, and H. J. Güntherodt, Phys. Rev. B **52**, 7311 (1995).
- [5] G. Bochi, H. J. Hug, D. I. Paul, B. Stiefel, A. Moser, I. Parashikov, H.-J. Güntherodt, and R. C. O'Handley, Phys. Rev. Lett. **75**, 1839 (1995).
- [6] M. Ciria, J. I. Arnaudas, L. Benito, C. de la Fuente, A. del Moral, J. K. Ha, and R. C. O'Handley, Phys. Rev. B **67**, 024429 (2003).
- [7] H. J. Hug, B. Stiefel, A. Moser, I. Parashikov, A. Klicznik, D. Lipp, H.-J. Güntherodt, G. Bochi, D. I. Paul, and R. C. O'Handley, J. Appl. Phys. **79**, 5609 (1996).
- [8] K. Ha, M. Ciria, R. C. O'Handley, P. W. Stephens, and S. Pagola, Phys. Rev. B **60**, 13780 (1999).
- [9] K. Ha and R. C. O'Handley, J. Appl. Phys. **87**, 5944 (2000).
- [10] H. J. Hug, B. Stiefel, P. J. A. van Schendel, A. Moser, S. Martin, and H.-J. Güntherodt, Rev. Sci. Instrum. **70**, 3625 (1999).
- [11] P. J. A. van Schendel, H. J. Hug, B. Stiefel, S. Martin, and H.-J. Güntherodt, J. Appl. Phys. **88**, 435 (2000).
- [12] H. J. Hug, B. Stiefel, P. J. A. van Schendel, A. Moser, R. Hofer, S. Martin, H.-J. Güntherodt, S. Porthun, L. Abelman, J. C. Lodder, G. Bochi, and R. C. O'Handley, J. Appl. Phys. **83**, 5609 (1998).
- [13] A. Hubert and R. Schäfer, *Magnetic Domains* (Springer-Verlag, Berlin, 2000).
- [14] U. Rüdiger, J. Yu, L. Thomas, S. S. P. Parkin, and A. D. Kent, Phys. Rev. B **59**, 11914 (1999).
- [15] S. Hameed, P. Talagala, R. Naik, L. E. Wenger, V. M. Naik, and R. Proksch, Phys. Rev. B **64**, 184406 (2001).
- [16] M. S. Pierce, R. G. Moore, L. B. Sorensen, S. D. Kevan, O. Hellwig, E. E. Fullerton, and J. B. Kortright, Phys. Rev. Lett. **90**, 175502 (2003).
- [17] Taken at 9.4 K.