Domains in perpendicularly magnetized ultrathin films studied using the magnetic susceptibility

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Measurements of the complex, low-frequency ac magnetic susceptibility χ^z of Fe/2 ML Ni/W(110) films, using a small field applied normal to the film surface, were used to characterize magnetic domain formation and motion in a perpendicularly magnetized ultrathin-film system. Analysis of the real part of χ shows that a broad peak in the susceptibility roughly divides low- and high-temperature regimes, where the domains are pinned and move freely, respectively. At high temperature, the domain density increases exponentially with temperature, producing an exponential decrease of the susceptibility with decay constant $\kappa \approx 0.05\,\mathrm{K}^{-1}$, consistent with theoretical expectations. At low temperature, domain-wall motion is thermally activated, with the activation energy increasing from 2×10^3 K for 3.0 ML Fe to 9×10^3 K for 1.5 ML Fe. The systematic variation of the activation energy indicates an increasing sensitivity to monolayer steps for thinner films, and yields an average separation of pinning sites of 200 ± 30 nearest-neighbor distances. This is consistent with a 0.2°–0.3° miscut of the substrate crystal. Films with Fe thickness $\leqslant$2 ML exhibit an exponential decrease in χ^z up to, and through, the transition to paramagnetism with no marker for the Curie temperature. Films with Fe thickness $>$2 ML undergo a reorientation of the magnetization from perpendicular to in plane as the temperature is increased. However, the reorientation produces no peak in the susceptibility, giving evidence that it is a discontinuous transition for these films. [S0163-1829(99)03937-5]

I. INTRODUCTION

Many practical characteristics of bulk ferromagnetic materials are determined by the properties of the magnetic domains within the material. Domains play a correspondingly important role in determining the properties of ultrathin magnetic films, particularly those which have the magnetization vector perpendicular to the plane of the film. The perpendicular magnetic anisotropy in these films produces distinctive processes of domain formation and dynamics that are intimately connected to distinctive magnetic phenomena such as moment orientation[1] and reorientation.[2,3] Recent studies of domains in perpendicularly magnetized films have concentrated on observations of the static domain distribution,[4–7] investigations and modeling of hysteresis loops,[8–10] the measurement of relaxation from the saturated state,[11–13] and the dynamics of magnetization reversal.[14,15] The present paper reports measurements of the magnetic susceptibility measured in small fields (8–16 Oe) and demonstrates that the susceptibility is dominated by domain-wall formation and motion. In contrast to earlier qualitative reports,[16,17] the quantitative analysis of the measurements permits the investigation of a range of phenomena exhibited by perpendicularly magnetized films. For example, it is possible to study the changes in the domain density that accompany the reorientation transition as a function of temperature, and to characterize the effective pinning potential through which the domain walls move.

The link between perpendicular magnetization and domain formation in ultrathin films derives from the enormous aspect ratio of ultrathin films and from the form of the magnetic dipole-dipole interaction.[18] If the magnetization lies in the plane of a film, the dipole-dipole interaction is effectively ferromagnetic, and favors a single domain state. When a perpendicular magnetic anisotropy holds the moments perpendicular to the film, the dipole-dipole interaction is effectively antiferromagnetic and favors the formation of alternating ''up'' and ''down'' domains. Furthermore, the size of the perpendicular magnetic anisotropy effectively determines the characteristic length scale of these domains.[19]

The resulting equilibrium density of domain walls is inextricably linked to magnetic phenomena in these systems. If the perpendicular anisotropy changes sign as a function of either the film thickness or temperature, the magnetization vector reorients from perpendicular to in plane.[20,21] Since the anisotropy is small near this spin-reorientation transition, it is preceded by a rapid change in the domain density[6] and a consequent reduction in the remanent magnetization.[2,22] Theory further predicts a series of distinct domain phases,[23,24] which have not yet been observed experimentally. The transition from ferromagnetism to paramagnetism in perpendicularly magnetized films may also be altered by the presence of a domain structure. Since the domain walls themselves can accept thermal energy, an alternate route to the paramagnetic state is through the introduction, fluctuation, and unbinding of domain walls. Numerical simulations show indications that this may occur.[25,26] Finally, as hysteresis properties are among the most important in technological applications, it is important to characterize the effective potential through which the domain walls move and to attempt to relate it to the microstructure.[9,14]

The remainder of the paper addresses these issues through analysis of the real part of the magnetic susceptibility of Fe films grown on 2 ML Ni/W(110). Section II presents a model of the magnetic susceptibility in perpendicularly magnetized thin films to aid in the subsequent analysis. Section III discusses experimental techniques and the application of the model to the measured susceptibility. Section IV contains a quantitative analysis of domain formation and motion as a function of film thickness.

II. MAGNETIC SUSCEPTIBILITY OF ULTRATHIN FILMS

The magnetic susceptibility has contributions from two mechanisms—critical fluctuations near phase transitions and domain-wall motion. For the first of these, consider the relation between the susceptibility $\chi_{\rm ext}$ as measured using an external field $\boldsymbol{H}_{\rm ext}$, and the suceptibility $\chi_{\rm int}$ due to the variation of the field $\boldsymbol{H}_{\rm int}$ internal to the film. Following MacDonald,[27] the fields are related by

$$\mathbf{H}_{\rm int}=\mathbf{H}_{\rm ext}+\mathbf{H}_{\rm eff}, \tag{1}$$

where the effective field $\boldsymbol{H}_{\rm eff}$ arises from variations in the total energy due to geometric and crystalline anisotropies:

$$\mathbf{H}_{\rm eff}=-\left(\frac{1}{\mu_0}\right)\frac{\partial E_{\rm anis}(\mathbf{M})}{\partial \mathbf{M}}. \tag{2}$$

An expression for the anisotropy free-energy density of the ferromagnetic film is given by

$$E_{\rm anis}=\tfrac{1}{2}\mu_0 DM^2\cos^2\theta+K_2(M)\sin^2\theta+K_4(M)\sin^4\theta, \tag{3}$$

where θ is the angle the magnetization $\mathbf{M}$ makes with the surface normal, D is the demagnetization factor for the sample geometry, and $K_2(M), K_4(M)$ are the second- and fourth-order crystalline anisotropy constants (which depend implicitly on temperature). Taking the partial derivative of Eq. (1) with respect to M_z, and considering the special case of $\mathbf{H}_{\rm ext}$ applied along the surface normal of the film yields the ''perpendicular'' susceptibility[28]

$$\chi^z_{\rm ext}=\frac{\partial \mathbf{M}_z}{\partial \mathbf{H}_{\rm ext}}$$

$$=\frac{\chi^z_{\rm int}}{1+\dfrac{1}{\mu_0}\left(\dfrac{\partial^2 E_{\rm anis}}{\partial(M\cos\theta)^2}\right)\chi^z_{\rm int}}. \tag{4}$$

The Curie transition and reorientation transition represent two distinct cases. For the first, the magnitude of the magnetization changes, but the orientation (and therefore cos θ) remains constant. Assuming $|K_4|\ll|K_2|$, Eq. (4) yields

$$\chi^z_{\rm ext}=\frac{\chi^z_{\rm int}}{1+\left[D+\left(\dfrac{\tan^2\theta}{\mu_0}\right)\left(\dfrac{\partial^2 K_2(M)}{\partial M^2}\right)\right]\chi^z_{\rm int}}. \tag{5}$$

While $\chi_{\rm int}$ diverges at a continuous phase transition, the behavior of $\chi_{\rm ext}$ depends on the geometry of the system. If the film is magnetized in plane, $\theta=\pi/2$, and $\chi^z_{\rm ext}\to 0$. Thus the measurements with a perpendicular $\mathbf{H}_{\rm ext}$ are insensitive to the phase transition. If the film is perpendicularly magnetized, $\theta=0$ and it is possible to measure a signal externally. However, the susceptibility saturates at the phase transition as

$$\chi^z_{\rm ext}=\frac{\chi^z_{\rm int}}{1+D\chi^z_{\rm int}}\to\frac{1}{D}. \tag{6}$$

In the thin-film geometry, $D\lesssim 1$, and the susceptibility is a very broad peak of order unity.[29] Thus, the present susceptibility measurements of the perpendicularly magnetized film are also insensitive to the transition to paramagnetism.[30]

At a reorientation transition, cos θ changes but the magnitude of $\mathbf{M}$ is fixed. A stability analysis of the free energy in Eq. (3) with respect to cos θ yields the equilibrium orientation,[20,21] which depends on the *effective* anisotropy $K_{\rm eff}=K_2-\frac{1}{2}\mu_0 DM^2$, on K_4, and on temperature through these parameters. If $K_4>0$, a continuous reorientation transition from a perpendicular to an in-plane magnetization begins when $K_{\rm eff}$ becomes negative,

$$\cos^2\theta=1+\frac{1}{2}\frac{K_{\rm eff}}{K_4},\quad 0\geqslant K_{\rm eff}\geqslant -2K_4, \tag{7a}$$

$$\chi^z_{\rm ext}=\frac{\chi^z_{\rm int}}{1+4\dfrac{(K_{\rm eff}+2K_4)}{\mu_0 M^2}\chi^z_{\rm int}}. \tag{7b}$$

The susceptibility diverges at the temperature T_R, when $K_{\rm eff}=-2K_4$ and $\mathbf{M}$ just falls in plane. As $K_{\rm eff}$ continues to change with temperature,

$$\cos^2\theta=0,\quad -2K_4>K_{\rm eff}, \tag{8a}$$

$$\chi^z_{\rm ext}=\frac{\chi^z_{\rm int}}{1-2\dfrac{(K_{\rm eff}+2K_4)}{\mu_0 M^2}\chi^z_{\rm int}}. \tag{8b}$$

Measurements with perpendicular $\mathbf{H}_{\rm ext}$ should therefore show a peak at a continuous reorientation transition.[31] Far from the transition, the susceptibility in Eq. (8b) continues to produce a signal essentially due to the torque applied on the in-plane magnetization by the applied field.

When $K_4<0$, the reorientation transition is discontinuous, and the equilibrium susceptibility is given by Eq. (8b) in the entire range $K_{\rm eff}<0$. $\chi^z_{\rm ext}$ now has no divergence and the measurements should not show a peak at the reorientation. There may again be a smaller response from the torque applied to $\mathbf{M}$ once it lies in plane.

Domain-wall motion also contributes to the susceptibility of perpendicularly magnetized films. The domain density n is determined by balancing of the magnetostatic energy gained by forming a region of reversed magnetization against the energy E_w required to insert a domain wall. A number of authors have calculated the resulting domain density.[7,19,32,33] Kashuba and Pokrovsky[23] use a microscopic, continuum spin-field model, and further calculate the response to an applied field, such that the susceptibility can be simply expressed. They find that

$$n=\left(\frac{\pi^2}{l}\right)\exp\left(-\left(\frac{\pi E_w}{2\Omega t}\right)-1\right), \tag{9}$$

where $\Omega=\frac{1}{2}\mu_0(g\mu_B S)^2/(a^4b^2)$ is the energy of the dipole interaction per unit volume, given by the Bohr magneton μ_B, the gyromagnetic ratio g, the in-plane separation a of the spins on a square lattice, and the film layer separation b. The thickness of the film is $t=Nb$. $E_w=4[\Gamma K_{\rm eff}]^{1/2}$ is the domain-wall energy per unit area and $l=\pi[\Gamma/K_{\rm eff}]^{1/2}$ is the domain-wall thickness. These are both given in terms of the effective perpendicular anisotropy per unit volume, $K_{\rm eff}$, and

the magnetic exchange energy per unit volume of domain wall. This last quantity scales with the parameter $\Gamma = 4JS^2/b$. J is the Heisenberg exchange constant for spins of magnitude S. As $K_{\rm eff}$, and thus E_w, approaches zero with increasing temperature near the reorientation transition, the domain density increases exponentially, and the magnetization averaged over an experimental sampling area disappears.[2,3,17]

The magnetic susceptibility results from the growth of one domain type at the expense of the other when a field is applied.[23] As the equilibrium domain density increases, a larger external field is required to drive the film to a single domain state, and the susceptibility decreases. In the limit of a small applied external field, the susceptibility is given by

$$\chi^z_{\rm ext} = \frac{4}{\pi n t} = A \exp\left[\left(\frac{\pi E_w}{2\Omega t}\right) + 1\right], \tag{10}$$

where $A = 4l/(\pi^3 t)$. The susceptibility therefore decreases exponentially as the temperature is increased.

The temperature dependence of the argument in the exponential in Eq. (10) depends on the system under study. Γ and Ω and K_2 and K_4 all depend on temperature because of thermal fluctuations of the spins. Furthermore, the anisotropies have distinct contributions from the surface and interior layers of the film (and possibly a magnetoelastic contribution induced by strain in the film), which are generally different functions of temperature.[34] Even if a reliable functional form was available for $E_w(T)/\Omega(T)$, it would be difficult to apply to the analysis of the experimental susceptibility because of the presence of contributions from processes other than domain-wall motion. That is, when $\chi^z = \chi^z_{\rm domain} + \chi^z_{\rm crit}$, even a small contribution from $\chi^z_{\rm crit}$ due to Eqs. (6) or (8b) will effectively mask the true temperature dependence of $\chi^z_{\rm domain}$ in a plot of $\ln \chi^z$. In the present experiments, the equilibrium susceptibility will be shown to be well described by the *ansatz*

$$\chi^z(T) = B + A^* \exp\{-\kappa T\}, \tag{11}$$

where B is a constant of order unity due to a very broad contribution from $\chi^z_{\rm crit}$. The connection between this expression and Eq. (10) can be made using a simplified model presented by Kashuba and Pokrovsky.[23] These authors and others[22,35] have outlined the classical temperature renormalization equations for $\Omega(T)$, $\Gamma(T)$, and $K(T)$ for the case where the anisotropy is dominated by the surface term, $K_2(T) = K_s(T)/t$ and $K_4(T)$ is neglected. This topic is treated in the Appendix, and expressions for κ and A^* are presented in Eqs. (A11) and (A12) in the context of this model.

If the walls are impeded by film defects such as inhomogeneities in the film thickness, terrace edges, or other types of pinning sites, the susceptibility due to domain-wall motion is affected. Bruno *et al.*[9] have summarized a relaxation model of impeded domain-wall motion, where the relaxation time τ is described by the thermally activated escape of the domain walls from the minima of a one-dimensional effective potential:

$$\tau = \tau_0 \exp\left(\frac{E_a}{k_B T}\right). \tag{12}$$

The activation energy E_a is in turn related to the mean separation of pinning sites ζ and the changes in the domain-wall energy due to pinning sites. Bruno *et al.*[9] present a model for the changes in the activation energy due to thickness variations, and show that the domain walls respond as if they are moving in a one-dimensional effective potential with modulations of amplitude $t\zeta(\Delta E_W)^2/E_W$, where ΔE_W is the variation in E_W due to the pinning site. The depth of these minima is identified with the activation energy. Estimating ΔE_W due to thickness variations Δt by differentials yields

$$E_a = \frac{64\zeta}{E_w^3}\left(\frac{\Gamma K_s}{t}\right)^2 \frac{(\Delta t)^2}{t} \approx \zeta(\Gamma K_s b)^{1/2}(\Delta N)^2 N^{-3/2}. \tag{13}$$

The approximation in Eq. (13) is valid when the anisotropy is dominated by the surface term K_s/t and uses $t = Nb$ to express the total thickness in terms of the layer thickness.

Finally, an expression for the measured susceptibility using an applied oscillatory field with angular frequency ω is obtained by having the domains relax to the equilibrium value $\mathbf{M}(t=\infty) = \chi^z_{\rm ext}\mathbf{H}(t)$ in the presence of an applied field. It is simple to show within the relaxation approximation that[9,36]

$$\chi^z = \frac{(1 - i\omega\tau)\chi^z_{\rm ext}}{1 + \omega^2\tau^2}. \tag{14}$$

The measured susceptibility due to domain-wall motion therefore exhibits a broad peak, which falls off at low temperature due to the exponential increase in the relaxation time, and falls off at high temperature due to the exponential increase in the domain density. The peak in the susceptibility is given approximately by the condition $\omega\tau = 1$.

III. MAGNETIC SUSCEPTIBILITY OF Fe FILMS GROWN ON 2 ML Ni/W(110)

Fe films grown on a 2-ML Ni buffer on a W(110) substrate offer an ideal system for the study of domain formation and the reorientation transition in perpendicularly magnetized films.[37–39] The Ni buffer forms slightly strained fcc (111) that has two different layer stacking domains present, and acts as a template for the subsequent growth of slightly strained fcc Fe. The nickel buffer films are magnetized in plane, but adding even 1/4 ML of Fe is sufficient to create a perpendicular magnetization.[17] This implies a small perpendicular anisotropy for the composite system, so that the reorientation transition from perpendicular to in-plane magnetization (as a function of temperature) appears for films with an Fe coverage >2 ML. The strained fcc Fe does not begin to relax towards a bcc structure until after the third monolayer is complete. Thus the properties of perpendicularly magnetized domains may be studied without the complication of reorientation in the thickness range of 1–2 ML Fe, and the effect of reorientation can be studied in the thickness range of 2–3 ML Fe. The films were grown by evaporation in a vacuum of 1×10^{-9} Torr, with the pressure falling to 4×10^{-10} Torr when the evaporation was complete. Total evaporation time was approximately 20 min. Thicknesses were calibrated to within ± 0.1 ML in separate calibration runs, using Auger electron spectroscopy. This was possible

because the first monolayer of either Fe or Ni on W(110) is much more stable than subsequent layers, and the Auger yield of successively deposited films, annealed to 600 K after each deposition step, shows a very distinct breakpoint in the uptake curves at 1 ML. For the films used as samples, the first monolayer of the Ni buffer was grown at a substrate temperature of 550 K to achieve almost perfect wetting.[38] The second Ni layer, and the Fe layers, were grown at a substrate temperature of 350 K.

In situ measurements of the ac magnetic susceptibility were made using the magnetooptic Kerr effect.[40,41] In the apparatus used in the present experiments,[42,43] linearly polarized light from a He-Ne laser passes through a polarizing crystal, then a UHV window, and falls on the film at 45° to the surface normal. A coreless coil creates a modulated **H** field at the sample position, resulting in a modulated Kerr ellipticity in the reflected light. The reflected light passes through the exit window and polarizer, and the magnetic signal is detected using a photodiode and lock-in ampliflier. The measurements presented here were made using the polar Kerr effect, with **H** parallel to the surface normal. Longitudinal Kerr effect measurements were also made using an in-plane field and reference is made to these published experiments. The magnetic susceptibility of the films was measured with $\mathbf{H}_{\text{ext}}$ of 8–16 Oe modulated at 210 Hz. The films were grown in a region where the magnetic field was compensated to less than 0.01 Oe, and were not explicitly magnetized prior to the measurements. Each susceptibility trace required approximately 0.5 h, including cooling time, and repeated measurements were very reproducible, so long as the temperature was not taken above 400 K.

One series of measurements of the real part of χ^z (in phase with the modulated field) was made on the same film after successive depositions, and is shown in Fig. 1. Figure 1(a) uses a linear scale for χ^z, whereas Fig. 1(b) uses a natural logarithmic scale. There is a consistent factor of approximately 2 uncertainty in the common absolute scale of χ^z, since the Voigt parameter of the films has been estimated from the Kerr rotation shown by previously published results for other Fe films.[44] The peak in the curve moves to lower temperature as the thickness increases from 1 ML to 3.25 ML. All the measurements show a response, indicating perpendicular magnetization, with a broad peak 30–60 K in width. Measurements made with smaller field amplitudes, down to about 0.5 Oe, show that this is an intrinsic width. Similar measurements on other samples[17,36] show that hysteresis, as indicated by the imaginary part of the susceptibility, persists to substantially higher temperature than the peak in the real part. This is very different from the susceptibility of in-plane magnetic systems that undergo a Curie transition.[41,17] These have a much larger (10^3–10^4 SI units), much narrower peak in the susceptibility near the critical temperature T_C, and hysteresis does not persist beyond T_C. It is clear, therefore, that the data in Fig. 1 does not represent observations of the Curie transition in perpendicularly magnetized films. This is in agreement with Eq. (6), which indicates that such a transition should reveal, at most, a very broad peak of order unity.

Domain-wall formation and motion is the source of the overwhelming contribution to the susceptibility in this system. The quantitative agreement between the data and the

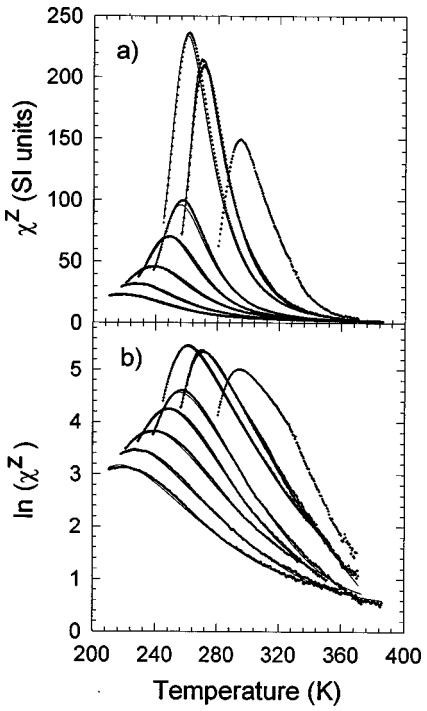

FIG. 1. The real part of the ac magnetic susceptibility of Fe/2-ML Ni/W(110) films measured using the polar Kerr effect and an applied field perpendicular to the film surface. The same data are shown on linear (a) and logarithmic (b) scales. The measurements were made on a single film after stepwise total Fe deposition of (progressing from right to left in the figure): 1, 1.5, 1.75, 2, 2.25, 2.5, 2.75, 3, and 3.25 ML. The lines fit to the data are discussed in the text.

domain model of susceptibility is illustrated in Fig. 2 using the measurements for a 1.75 ML Fe film. In Fig. 2(a), the logarithm of the real part of the susceptibility is plotted against temperature. At high temperature, where $\omega\tau \ll 1$, the domains are freely moving and the plot in principle gives the function $E_w(T)/\Omega(T)$. As has been previously discussed, in practice there is a very broad contribution due to the Curie transition and the data are fitted instead to the *ansatz* in Eq. (11). The curve in Fig. 2(a) is given by the parameters $B = 1.71 \pm 0.02$, $A^* = (7.52 \pm 0.08) \times 10^8$, and $\kappa = 0.0546 \pm 0.0004\,\text{K}^{-1}$. B implies a value of $D \approx 0.6$. An ideal film would have $D = 1$, but both finite thickness, roughness, and the inhomogeneity of the bilayer will reduce the demagnetization factor.[29] It is shown in the Appendix that the simplified model of Kashuba and Pokrovsky leads to an estimate $\kappa = 0.09\,\text{K}^{-1}$ if the film thickness refers only to the 2 ML of Fe, or $\kappa = 0.06\,\text{K}^{-1}$ when using the entire 4 ML of magnetic material (Fe plus Ni) as the thickness. The fitted values are therefore very reasonable.

At lower temperatures the domains do not move freely, but rather respond with a thermally activated time constant τ. For quantitative analysis of the low-temperature tail of the susceptibility, Eqs. (11), (12), and (14) are combined to show that the activation energy may be displayed by plotting $\ln[(A^* e^{-kT}/\chi) - 1] = \ln(\omega^2 \tau^2)$ vs $1/T$. Figure 2(b) shows such a plot for the film of thickness 1.75 ML Fe. The linear region at low temperature yields an activation energy of $E_a/k_B = (7.43 \pm 0.02) \times 10^3\,\text{K}$. For comparison, Berger and Hopster[12] measured the relaxation of the magnetization of a

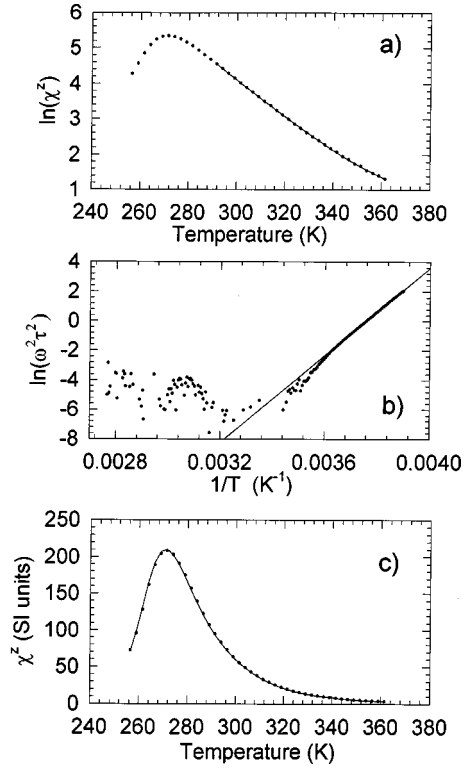

FIG. 2. The real part of the susceptibility of the film with 1.75 ML Fe is fit to the domain model. (a) The high-temperature portion ($\omega\tau \ll 1$) is fit to Eq. (12). (b) An Arrhenius plot of $\ln(\omega^2\tau^2)$ against $1/T$ demonstrates that the domain-wall motion is thermally activated at low temperature. The scatter at high temperature represents the deviations from the fit in part (a). (c) A comparison of the fitted line and data over the entire range, using the parameters determined in parts (a) and (b). In parts (a) and (c), every fifth data point is plotted to allow the fitted line to be seen.

saturated 4-ML Fe/Ag(001) films with perpendicularly magnetized domains, yielding $E_a/k_B = 3.9 \times 10^3$ K (with considerable variation from sample to sample). The resulting fit for the complete susceptibility curve is shown on Fig. 2(c). The fitted curve agrees very well with the data, particularly when it is considered that only the high- and low-temperature tails were used in the fitting process.

Returning to Fig. 1, a similar procedure has produced the lines fitted to all the data traces. The fitted parameters are presented and interpreted in Sec. IV. It is clear that the measured susceptibility is well described by domain-wall formation and motion. (The susceptibility measured for 1 ML of Fe has a different shape and does not fit the model of domain motion. This has been observed in many films of thickness 1 ML of Fe or less, and could be the result of more complicated domain pinning processes in films that are not yet contiguous.) It is now possible to understand the striking reduction in the magnitude of the susceptibility in Fig. 1 that occurs once the film is thick enough ($\geqslant 2\frac{1}{4}$ ML) for the reorientation transitions to occur. This is the result of the increase in domain density as a function of *thickness* at a given *temperature* in the approach to the reorientation transition, as has been observed previously by Speckmann *et al.*[6] using domain images of wedged films.

As a final point, consider the absolute size of the SI susceptibility. Equation (10) gives the characteristic size of the domains $d = 1/n$ at high temperature, where $\omega\tau \ll 1$ and the domains are freely moving. The data in Fig. 1 then yield freely moving domains of 600 Å, which shrink to 60 Å as the temperature is increased. These very small values cannot be correct, since the lower limit of the domain size[23] should be of order $\Gamma/\Omega t \approx 150a = 450$ Å. While an approximate calibration of the magneto-optical effect was used, the order of magnitude should be correct. The origin of this discrepancy is not yet clear. A second discrepancy in the size of the susceptibility was originally pointed out in Ref. 36, and is noted here without proof. A complete characterization of the Re[χ] should also determine Im[χ] through Eq. (13). However, the measurements yield Im[χ] that is much smaller than predicted from Re[χ].

Noteworthy by its absence is any sizable contribution to the susceptibility by the critical fluctuations. For films of Fe thicknesses 2 ML or less, experiments with $\mathbf{H}_{\text{ext}}$ applied in plane indicate that no reorientation transition occurs. The corresponding susceptibility data in Fig. 1 continue to display an exponential decrease at high temperature, indicative of perpendicular domain formation, but this process evidently ends in the paramagnetic state. The data show no marker for T_C, other than the presence of the small constant *B*. Because the data are insensitive to this transition, it is difficult to test recent calculations,[25,26] which suggest that the transition to paramagnetism proceeds continuously by fluctuations of the domain walls. For films of Fe thickness greater than 2 ML, previous measurements[17] with $\mathbf{H}_{\text{ext}}$ applied in plane show clearly that a reorientation transition to in-plane magnetization occurs at a temperature T_R in the temperature range 340–270 K for films of 2.25–3-ML Fe thickness. Close examination of Fig. 1 in this range shows no indication of a departure from the domain model, and accordingly, no indication of the diverging susceptibility of Eqs. (7b) and (8b) that would accompany a continuous reorientation transition. It is not likely that this contribution is ''washed out'' by the use of too large a modulation field or by the presence of too large an uncompensated background field. It is certainly true that these effects can distort and greatly reduce the magnitude of the measured susceptibility, but if they mask a divergence peak due to the reorientation transition, one would expect that they would have similarly affected the critical susceptibility at the Curie temperature for films magnetized in plane, measured under identical conditions. Published data show that this is not the case.[17] Once again, the only evidence for the presence of the critical susceptibility in the thicker films is the small constant, *B*. This small contribution is consistent with the appearance of an in-plane magnetization that responds to the torque applied by the perpendicular field $\mathbf{H}_{\text{ext}}$ according to Eq. (8b). While it is not possible to draw a definite conclusion from the *absence* of a peak, these observations suggest a discontinuous (first-order) reorientation transition that passes through a metastable state where regions of in-plane and perpendicular magnetization coexist. There are previous reports of discontinuous reorientation transitions.[20,45]

IV. THICKNESS DEPENDENCE OF DOMAIN PROCESSES

Having established that the susceptibility measurements result from domain-wall formation and motion, the param-

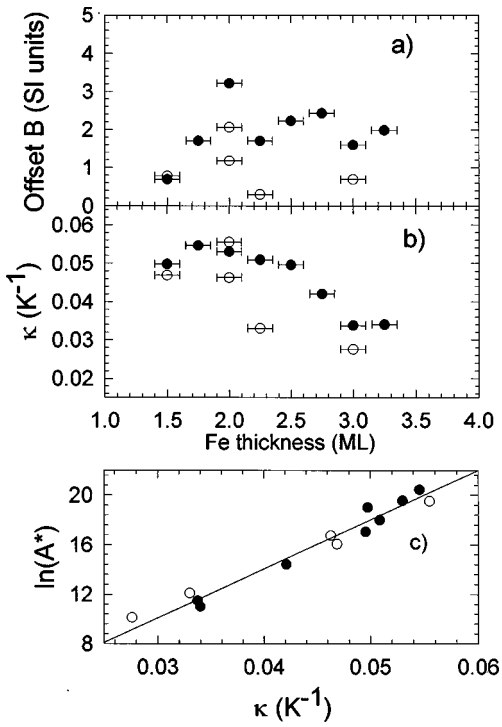

FIG. 3. The fitting constants that describe the exponential change in domain density in Eq. (11) are shown for the data in Fig. 1 (solid symbols), and for independently prepared films (open symbols). These are (a) the temperature-independent contribution B; (b) the simple exponential decay constant κ; and (c) the magnitude A^*. The magnitude is presented as a correlation plot against κ.

eters extracted from the fits to the data in Fig. 1 and to other similar measurements, can be used to gain insight into these processes in perpendicularly magnetized films. Since domain formation and pinning is very sensitive to the microstructure of the film, the data shown in figures in this section were divided into two groups. Values derived from the measurements made on a single film after successive depositions (i.e., those in Fig. 1) are expected to have a common microstructure and be highly correlated. These will be indicated by filled symbols. Values derived from independently grown films are expected to show more variation and are represented by open symbols.

Figure 3 presents the constants derived by fitting Eq. (11) to the susceptibility at high temperature where $\omega\tau \ll 1$. The values for B are shown in Fig. 3(a). Since reorientation is first observed for 2.25-ML Fe films, the interpretation of B depends on the thickness. For a thickness of 2.0 ML and less, it represents a paramagnetic susceptibility of a perpendicularly magnetized system, which is saturated due to the demagnetizing field. The fitted values of order unity in SI units are reasonable for this mechanism. For a thickness of greater than 2 ML, the constant term represents the response of regions of in-plane magnetization to a perpendicular field. Comparison to measurements for Fe films grown on a 1-ML Ni buffer, where the system is magnetized in plane regardless of temperature, show a perpendicular susceptibility of 3–4 SI units.[17] Thus the values in Fig. 3(a) are consistent with the two proposed mechanisms, but provide no independent confirmation that two distinct mechanisms are required.

The value of κ, which characterizes the domain formation process, is presented in Fig. 3(b). Since κ is given by a temperature expansion of $E_w(T)/\Omega(T)$, it is not possible to interpret it in a straightforward manner. In the model presented in the Appendix, it is assumed that the anisotropy is given entirely by the surface anisotropy, K_s, which remains constant as the thickness changes. (Of course, K_s/t changes.) While the model predicts the correct range of values of κ, according to Eq. (A11), κ is expected to increase with thickness as the reorientation temperature decreases. The data, however, show a decrease in κ with thickness. This discrepancy is likely due to too simple a treatment of the anisotropy in the model. To simulate the experimental result requires that K_s increase slightly with thickness. There are two mechanisms by which this could occur in the extreme low-thickness regime probed by the experiments. First, the Fe is strained and is known to begin a transformation toward a bcc structure once the thickness reaches 4 ML. Strains can induce an anisotropy[34] that scales as $1/t$ and could augment K_s. Second, in a 2-ML Fe/2-ML Ni/W(110) film, there are no magnetic atoms that have bulk Fe or Ni coordination. In this thickness regime, it is plausible for the surface anisotropy averaged over the sample to increase as the surfaces and interfaces are formed. The data presently available do not permit these ideas to be tested.

The fitted magnitude of the susceptibility A^* is presented in Fig. 3(c) as a correlation plot of $\ln(A^*)$ against κ. The correlation results from the incorporation of the constant term in the expansion of $E_w(T)/\Omega(T)$ into the preexponential factor A in Eq. (10). As is shown in Eq. (A12), this gives a linear correlation $\ln(A^*)=\ln(A)+2\kappa T_R$. The slope of the correlation curve is 397 ± 22 K, which is considerably less than $2T_R$, but of the proper magnitude. This is likely due to the fact that T_R changes with thickness, and that T/T_R is not a very small expansion parameter for most of the range of the data. Nevertheless, the source of the correlation is clear.

Figure 4 presents the constants derived from fitting Eq. (14) for domain-wall pinning to the susceptibility at temperatures well below their peak value. The activation energies shown in Fig. 4(a) show a clear trend, decreasing with increasing film thickness, as is qualitatively reasonable. The data plotted with solid symbols were fit to Eq. (13) for the variation of E_a with thickness. Because of the ambiguity as to how to treat the 2 ML of Ni in determining the number of magnetic monolayers, the functional form was altered to $E_a/k_B = C(N-N_0)^{-3/2}$, with N the number of Fe layers. The best fit, which is illustrated on the figure, describes the data well and demonstrates that the pinning sites are consistent with a model based upon variations in the film thickness. The fact that a single value of ζ, the average separation of pinning sites, applies to the measurements performed on a single film grown sequentially is an important check of the self-consistency of the analysis. The fitting constants are $C=(1.2\pm.2)\times10^4$ K and $N_0=0.31\pm0.14$ ML. The fact that N_0 is close to zero suggests that only the magnetic roughness of the Fe is important. This confirms that there is little intermixing, and that the magnetic system is driven by the Fe layer—the primary role of the Ni layers being to alter the interface anisotropy of the Fe. The fitted value of C permits an estimate of ζ in this film. Using the values of Γ and K_s from the Appendix, and letting $\Delta N=1$ for thickness changes of one atomic step, gives $\zeta/a \approx 200\pm30$. Using the square

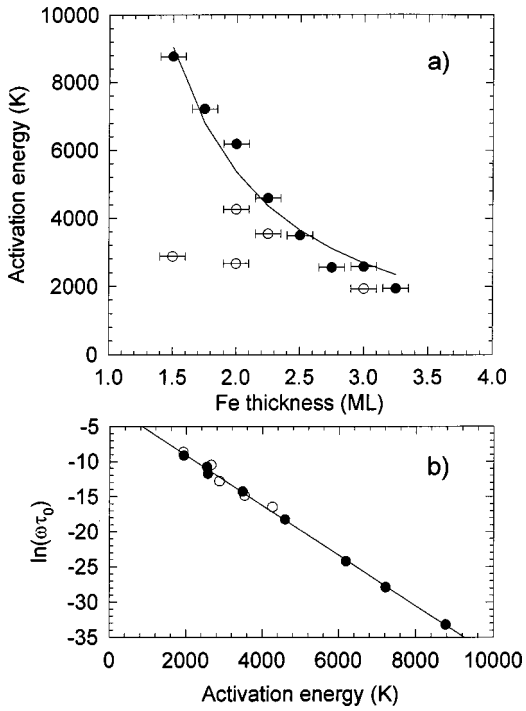

FIG. 4. The fitting constants in Eq. (12) that describe the thermal activation of domain-wall motion are shown for the data in Fig. 1 (solid symbols), and for independently prepared films (open symbols). These are (a) the activation energy, (b) the prefactor $\ln(\omega\tau_0)$. The activation energies for the data represented by solid symbols are fit to $t^{-3/2}$, as described in Eq. (13). The prefactor $\ln(\omega\tau_0)$ is presented as a correlation plot against activation energy. The correlation originates from the condition $\omega\tau\approx 1$ at the temperature where the susceptibility has its maximum.

root of the surface area per atom in the Ni(111) substrate for a, gives $\zeta = 580 \pm 90$ Å. This value is slightly larger than, but consistent with, those found for other ultrathin magnetic films.[9,14,15] Since the plot indicates that a single characteristic separation of the pinning sites applies to all Fe thicknesses, the sites are likely structural features associated with the tungsten substrate or the Ni buffer. In the first case, this is consistent with terraces of one atom step height resulting from a 0.3° miscut of the substrate crystal. In the second case, low-energy electron diffraction patterns of the Ni substrate indicate that there are structural domains of alternate fcc stacking. The pinning may arise through thickness variations at growth defects at these domain boundaries.

The fitted values of τ_0 are presented as a correlation plot of $\ln(\omega\tau_0)$ against the activation energy in Fig. 4(b). If the logarithmic fits of the low- and high-temperature tails of the susceptibility are extended, they intercept at $\omega\tau = 1$, near the maximum of χ^z. Denoting the temperature at the maximum as T_P, and substituting Eq. (12) for τ gives $\ln(\omega\tau_0) = -E_a/(k_B T_P)$. The slope of the correlation plot gives $T_P = 280 \pm 5$ K, which is in the middle of the small range of peak temperatures observed in the measurements (see Fig. 1). The intercept of -1.9 ± 0.3 is to be compared with the predicted value of zero. It is clear that this is the origin of the correlation of these fitting parameters, and that the peak in the susceptibility therefore roughly divides the regions of free and hindered domain motion.

V. CONCLUSIONS

The domain processes that occur in perpendicularly magnetized ultrathin films have been studied quantitatively using the magnetic susceptibility. Fe/2-ML Ni/W(110) films reveal the exponential increase in domain density that precedes the reorientation transition, as a function of either temperature or thickness. The measured simple exponential decay constant of the susceptibility (≈ 0.05 K^{-1}) due to domain formation is in agreement with theory, but the precise functional dependence of the exponent on temperature is masked by the presence of a small contribution from the critical susceptibility. The data suggest that the reorientation in this system is a discontinuous transition, as there is no indication of the strong peak in the susceptibility that would accompany a continuous transition. Films that are too thin to exhibit a reorientation transition also exhibit an exponential increase in domain density that persists into the paramagnetic state with no clear indication of the Curie temperature. At temperatures below the peak in χ^z, the susceptibility increases rapidly with temperature due to thermally activated domain-wall motion. The method readily yields the activation energy, which is seen to increase rapidly with decreasing thickness as imperfections in the film become more important. The variation of the activation energy with thickness is in agreement with the theory of Bruno *et al.*,[9] and the extracted average separation of pinning sites for small applied fields is consistent with terrace defects of the substrate. Susceptibility measurements provide a straightforward method to characterize the effective pinning potential through which the domain walls move.

APPENDIX

Kashuba and Pokrovsky[23] give the classical renormalization equations for the magnetic interactions in a spin-continuum model of a monolayer film. These are altered slightly to describe a film with N magnetic monolayers as an effective film of one monolayer. Thus the magnetic interaction strengths are expressed as energy/area instead of energy/volume. The anisotropy is considered to arise from the surface term alone,

$$\Gamma(T) = N\Gamma_0 Z(T), \tag{A1}$$

$$\Omega(T) = N\Omega_0 Z^2(T), \tag{A2}$$

$$K_s(T) = [K_0 + (3N\Omega_0/2a) I(\xi,T)] Z^3(T), \tag{A3}$$

$$K_{\mathrm{eff}}(T) = K_s(T) - N\Omega(T)/a. \tag{A4}$$

The quantities $\Gamma_0 = 4JS^2$, $\Omega_0 = \frac{1}{2}\mu_0 (g\mu_B S)^2/a^4$, and K_0 are the magnetic interaction strengths for a single monolayer at zero temperature. The temperature rescaling function is

$$Z(T) = 1 - \frac{\xi T}{2\pi N\Gamma_0}, \tag{A5}$$

with

$$\xi = \tfrac{1}{2}\ln[N\Gamma_0/K_0 a^2]. \tag{A6}$$

The term that mixes the renormalized anisotropy and dipole contributions is

$$I(\xi,T)=\int_0^{\xi} e^{-\xi'} Z^{-2}\left(\frac{\partial Z}{\partial \xi'}\right) d\xi'. \tag{A7}$$

Placing these relations into Eq. (10) gives

$$\frac{\pi E_w(T)}{2\Omega(T)}=2\pi\left(\frac{\Gamma_0}{\Omega_0 a}\right)^{1/2}\left[\left(\frac{K_0 a^2}{N\Omega_0 a}\right)+\frac{3}{2}I(\xi,T)-\frac{1}{Z}\right]^{1/2}. \tag{A8}$$

Because $T/N\Gamma_0$ is a relatively small parameter near room temperature, the temperature dependence in Z^{-1} and $I(\xi,T)$ can be linearized to yield

$$\frac{\pi E_w(T)}{2\Omega(T)}=2\pi\left[\left(\frac{\Gamma_0}{\Omega_0 a}\right)\left(\frac{K_0 a^2}{N\Omega_0 a}-1\right)\right]^{1/2}\left[1-\frac{T}{T_R}\right]^{1/2}$$
$$\equiv\alpha\left[1-\frac{T}{T_R}\right]^{1/2}, \tag{A9}$$

where the reorientation temperature

$$T_R=2\pi\Gamma_0\frac{\frac{K_0 a^2}{\Omega a}-N}{\xi+\frac{3}{2}(1-e^{-\xi})}. \tag{A10}$$

Since ξ depends only logarithmically on N, the reorientation temperature is seen to decrease roughly linearly with thickness. Films of 2 ML Fe grown on 2 ML Ni show a reorientation temperature[17] of about 350 K. Using the measured moment[39] of $2.13\mu_B$ and the observed fcc structure[37] the dipole energy at zero temperature, $\Omega_0 a=2.2\,\mathrm{K}$ and $\Gamma_0=680\,\mathrm{K}$. Equations (A10) or (A8) are used to solve for the value of $K_0 a^2$, which gives the observed reorientation temperature. One finds $K_0 a^2=5\,\mathrm{K}-10\,\mathrm{K}$ as N varies from 2–4 ML (depending on what is considered to be the thickness of the magnetic film). Equation (A3) then gives a perpendicular surface anisotropy at room temperature of 3.5–7.5 K, in reasonable agreement with the experimental value[39] of 4.0 ± 0.9 K.

Contact with the phenomenological form of the susceptibility introduced in Eq. (11) can be made in the limit that T/T_R is a small parameter. Then the simple exponential decay constant is given by

$$\kappa=\frac{\alpha}{2T_R}=\frac{\xi+\frac{3}{2}(1-e^{-\xi})}{[2N\Gamma_0(K_0 a^2-N\Omega_0 a)]^{1/2}}. \tag{A11}$$

The relation between the prefactors in Eqs. (10) and (11) is then

$$\ln A^*=\ln A+\alpha=\ln A+2\kappa T_R. \tag{A12}$$

Even though T/T_R is not a small parameter in the present experiments, Eq. (A11) should give a useful estimate. The estimate of κ is then from 0.09 K^{-1} ($N=2$) to 0.06 K^{-1} ($N=4$).

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