

Domain formation near the reorientation transition in perpendicularly magnetized, ultrathin Fe/Ni bilayer films (invited)

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Ultrathin films with perpendicular magnetization convert from a single domain state into a multidomain structure as the reorientation phase transition to an in-plane magnetization is approached. Reorientation transitions in magnetic ultrathin films result from the interplay of interfacial magnetic anisotropy, the dipolar interaction, and two-dimensional thermodynamics. These transitions can be driven by changing either the film thickness or temperature. Experimental and theoretical studies of this effect are briefly discussed in the context of the thickness–temperature phase diagram of the reorientation transition. We then describe magnetic susceptibility experiments on ultrathin Fe/Ni(111) bilayers. Our experiments indicate an exponential increase in domain density of a multidomain structure with temperature and identify the region of the thickness–temperature reorientation transition phase diagram where this condensation is most pronounced. The temperature dependence of the domain density agrees quantitatively with theoretical predictions. Films that are slightly too thin to exhibit the reorientation transition with temperature are a special case. They undergo a ferromagnetic-to-paramagnetic transition from the perpendicularly magnetized state and exhibit domain-like behavior many tens of Kelvin above estimates of the Curie temperature. This surprising observation is interpreted using the two-dimensional dipolar Ising model. © 1999 American Institute of Physics. [S0021-8979(99)78408-2]

I. INTRODUCTION

Three factors can contribute to unique domain formation phenomena in perpendicularly magnetized ultrathin films. Yafet and Gyorgy¹ identified the first factor; they predicted that domain structures in these films would be very sensitive to changes in magnetostatic energy if the effective perpendicular anisotropy $K_{\perp\text{eff}}$ were very weak. $K_{\perp\text{eff}}$ includes both the interfacial anisotropy (favoring perpendicular magnetization) and the magnetostatic (dipole) interaction (favoring an in-plane magnetization). The domain structure's sensitivity to energy changes is due to the magnetostatic energy's *insensitivity* to changes in the domain structure when the lateral extent of the domains is much greater than the film thickness.

The second factor is that $K_{\perp\text{eff}}$ can change sign due to changes of the film thickness. A zero crossing of $K_{\perp\text{eff}}$ drives the reorientation phase transition from a perpendicular to in-plane magnetization direction. This transition proceeds with increasing thickness as the dipolar term overwhelms the perpendicular interfacial anisotropy.²

The third factor is temperature. In the ultrathin limit, two-dimensional (2D) thermodynamics prevail, producing an unusually strong connection between magnetic anisotropy and magnetic order. Indeed, ferromagnetic order in 2D is stabilized by magnetic anisotropy.³ Jensen and Benneman⁴ first argued that 2D thermodynamic effects would produce a temperature-driven reorientation in ultrathin magnetic films with suitably weak perpendicular magnetic anisotropy. This

effect was observed by Pappas *et al.*⁵ The temperature driven reorientation is then interesting because 2D thermodynamics play a more explicit role than in the thickness driven transition.

High temperature studies of perpendicularly magnetized, ultrathin films are discussed here in conjunction with studies of domain formation near the reorientation transition. The general features of the thickness–temperature phase diagram for the reorientation transition are introduced in Sec. II. Experimental studies of domain formation in various regions of the phase diagram are reviewed in Sec. III. The emphasis of the article is on the new magnetic behavior revealed by our magnetic susceptibility measurements of ultrathin Fe/Ni bilayers. These films exhibit perpendicular magnetism, a reorientation transition, and domain formation. Section IV summarizes the theoretical prediction for the temperature dependence of the domain-wall-motion magnetic susceptibility. Section V contains an overview of our experiments and focuses on two aspects of the results. One new result is that the domain density increases exponentially with temperature in quantitative agreement with the theory of Kashuba and Pokrovsky. Another new issue arises in the case of strictly perpendicular films that have a weak $K_{\perp\text{eff}}$ but do not exhibit a reorientation transition with temperature. Under these conditions, the Curie temperature T_C may not have its usual meaning. Specifically, the magnetic susceptibility remains domain-like to temperatures well above the best estimates of T_C , for which no signature is apparent in the data.

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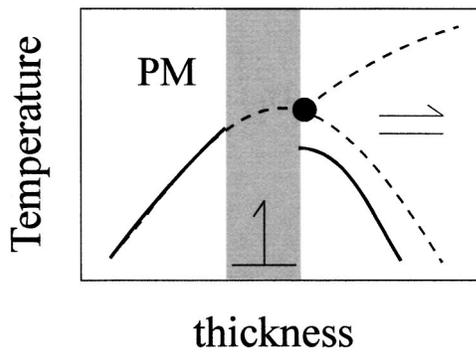


FIG. 1. A sketch of the temperature–thickness phase diagram of an ultrathin film with perpendicular magnetic anisotropy shows regions of perpendicular (\perp) and in-plane (\rightleftharpoons) ferromagnetism and paramagnetism (PM). Horizontal and vertical trajectories through the \perp and \rightleftharpoons boundary correspond to thickness and temperature driven reorientation phase transitions, respectively. A line of Curie temperatures separates \perp and PM phases. The two boundaries meet at a multicritical point (\cdot). Solid lines indicate parts of the topology qualitatively verified in experiments by Qiu *et al.* (Ref. 7).

II. TEMPERATURE–THICKNESS PHASE DIAGRAM OF THE REORIENTATION TRANSITION

Figure 1 is a phase diagram after Politi *et al.*,⁶ that describes both thickness- and temperature-driven transitions. It is based on a Heisenberg model modified by a uniaxial anisotropy that favors a perpendicular orientation of the moments. The dipolar field and its spatial dependence was partly incorporated into the model, although domain formation was excluded. The model predicts a perpendicular ferromagnetic phase at low temperature and thickness, an in-plane ferromagnetic phase at higher coverage and intermediate temperature, and a high temperature paramagnetic phase. In principle, the boundary between out-of-plane and paramagnetic phases is a line of Curie temperatures. Also, a single reorientation-transition boundary separates out-of-plane and in-plane phases. The two boundaries meet at a point which we refer to here as the multicritical point. Horizontal and vertical cuts through the reorientation transition boundary correspond to the thickness and temperature driven transitions, respectively. Portions of this diagram were verified by Qiu *et al.* for Fe/Ag(100),⁷ as indicated by the solid boundaries. The actual phase diagram of the FeAg(100) system is slightly skewed compared to the theoretical diagram. Specifically, the experimental reorientation boundary appears shifted to lower temperatures relative to the T_C boundary of the perpendicular phase. This difference is not significant in the following discussion for which Fig. 1 provides a context.

III. EXPERIMENTAL OBSERVATIONS OF DOMAIN FORMATION IN ULTRATHIN FILMS

Domain formation in the perpendicularly magnetized phase plays a prominent role near the reorientation transition,^{8–10} and at high temperature.^{11–13} Theories of domain formation in this context are now highly developed.^{14–20} Indeed, the ground state of a perpendicularly magnetized, ultrathin film is not the single domain state, but is instead a multidomain state such as a striped pattern.

Single domain states are either metastable,²¹ supported by defects which hinder domain-wall motion, or are possible because the lateral sample dimensions are less than the equilibrium diameter of the domains.¹⁵ The vanishing perpendicular magnetic anisotropy in the approach to the reorientation transition reduces the energy cost of inserting domain walls and an exponential condensation of the domain structure occurs.¹⁵ Magnetic-imaging experiments by Allenspach and Bischof verified this type of domain condensation qualitatively at a number of discrete temperatures,⁹ and Speckmann *et al.* observed a similar effect in the thickness-driven transition.¹⁰ Those experiments correspond to vertical and horizontal slices through the reorientation boundary of Fig. 1.

Single-domain behavior has been observed in some perpendicularly magnetized ultrathin films. For example, a few perpendicularly magnetized ultrathin film/substrate systems have been studied to determine their Curie temperatures and identify their thermodynamic universality classes.^{22,23} The temperature dependence of the magnetization in these experiments was consistent with the 2D Ising model. Referring again to Fig. 1, these experiments correspond to magnetization measurements along vertical slices through the line of Curie temperatures on the low thickness side of the diagram.

In other experiments studying perpendicularly magnetized films at high temperatures, deviations from ideal critical behavior were observed close to T_C . Kolhepp *et al.* observed a substantially different behavior for the temperature dependences of the remanent and saturation moments $M_r(T)$ and $M_s(T)$, of Co/Cu(111) resulting in a difference of >10 K in the respective Curie temperature determinations.²² The authors attributed this effect to sample imperfections. More recently, Pouloupoulos *et al.* argued that a similar difference between $M_r(T)$ and $M_s(T)$ for 8–10 ML Ni/Cu(100) films was due to domain formation close to, but below, T_C .¹¹ We show in Sec. V that domain formation is most prolific in Fe/Ni bilayers at thicknesses marginally less than those where a temperature-driven reorientation occurs. These films correspond to the shaded intermediate region in Fig. 1, where Qiu *et al.*⁷ did not report results.

IV. THE DOMAIN-WALL-MOTION SUSCEPTIBILITY OF AN OUT-OF-PLANE, ULTRATHIN FILM

Kashuba and Pokrovsky considered the response of a domain structure to an applied field.¹⁵ At a given thickness and temperature, the equilibrium configuration of their perpendicularly magnetized films was a stripe domain structure of alternating “up” and “down” magnetization. The application of a small magnetic field in the up direction causes the up domains to grow at the expense of the down domains, thus producing a magnetic response. Energy minimization then determines the perpendicular susceptibility of the domain structure, which is found to vary inversely with the stripe linear density. The dipolar and exchange interactions and interfacial anisotropy determine the equilibrium stripe density. 2D thermal fluctuations renormalize these terms to temperature-dependent quantities. The effective perpendicular anisotropy crosses 0 at the reorientation transition. A

vanishing effective anisotropy produces an exponential increase in the linear density of the domain structure. This, in turn, produces an exponential decrease in the perpendicular susceptibility. Using the T-dependent expressions of Abanov *et al.*,⁵ and expanding to first order in the temperature, a simple exponential decay of the perpendicular susceptibility is obtained,^{24,25}

$$\chi_{\perp}(T) \propto \exp(-\alpha T). \quad (1)$$

A more detailed description of this derivation is found elsewhere.²⁴ The same expression was recently obtained by De'Bell *et al.* for the 2D dipolar Ising ferromagnet using simple scaling arguments, although the constant α is unphysically large in that calculation.²⁶

This approximation is valid for $T \ll T_R$, the temperature where reorientation occurs, or for $T \ll T_C$ in the case of perpendicularly magnetized films that does not exhibit a reorientation transition. Using values appropriate for a 2 ML Fe film investigated in Sec. V, where $T_R = 300$ K, we expect the decay exponent is to be approximately 0.07 K^{-1} ; χ_{\perp} is predicted to drop by an order of magnitude every 35 K. This rate of decay is much slower than that of an in-plane susceptibility in the temperature range just above the Curie temperature.^{12,27-29}

V. MAGNETIC SUSCEPTIBILITY EXPERIMENTS ON ULTRATHIN Fe/2 ML Ni/W(110) FILMS

The thickness-temperature phase diagram of ultrathin Fe films grown on a 2 ML Ni buffer was investigated using magnetic susceptibility measurements. Ultrathin films of Fe were grown in ultrahigh vacuum by electron-beam evaporation on a 2 ML Ni buffer on the (110) face of a W single crystal. The preparation methods and film structure have been studied and published previously.³⁰ The face of the 2 ML Ni buffer is nearly identical to the bulk Ni(111) and provides a template for fcc Fe growth. The Fe structure was slightly strained fcc for thicknesses less than 3 ML, and a gradual transition to bcc proceeded at greater thicknesses. The films were ferromagnetic at low temperatures, with a moderate perpendicular magnetic anisotropy. 1 ML Ni buffers were not suitable as fcc templates. Presumably, this is because the first monolayer on W(110) is strained further from the Ni(111) structure than the second monolayer. Fe grew on the 1 ML buffer in a strained bcc fashion and exhibited poor long-range order. Fe on 1 ML Ni films were magnetic, however, with an in-plane magnetization along the $W(\bar{1}10)$ direction.

Magnetic susceptibilities were measured in a small, ac field for three orthogonal field orientations: two in-plane orientations and one perpendicular to the film plane. The convention here will be to label the susceptibilities according to the direction of the applied field. Hence, the susceptibility corresponding to the perpendicular field orientation is χ_{\perp} . In contrast to the remanent magnetization, that is small or even zero the magnetic susceptibility of the domain structure is expected to be finite or even large. Complex magnetic susceptibilities $\chi = \chi' + i\chi''$ were measured as a function of temperature with the ac magneto-optic Kerr effect

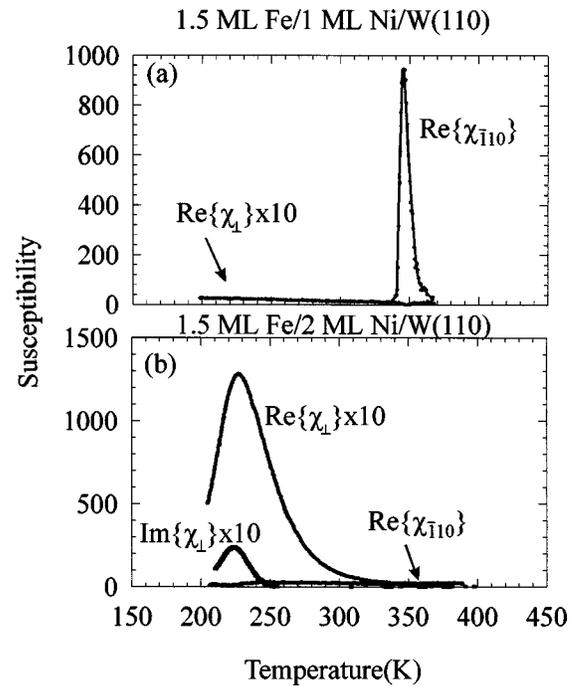


FIG. 2. Magnetic susceptibilities of (a) an *in-plane* magnetized 1.5 ML Fe/1 ML Ni film and (b) an *out-of-plane* magnetized 1.5 ML Fe/2 ML Ni films on W(110) illustrate the qualitatively different magnetic responses of in- and out-of-plane magnetized ferromagnetic films. Both figures show the susceptibility with the applied field perpendicular to the film χ_{\perp} and along the in-plane $W(\bar{1}10)$ direction $\chi_{\bar{1}10}$. The susceptibility along the $W(100)$ direction is omitted for simplicity, but was 0 in both cases. (a) measures the critical susceptibility at the ferromagnetic-to-paramagnetic transition while (b) measures domain wall motion.

technique²⁹ using a frequency of 210 Hz. A single phase lock-in amplifier was used to measure the real (in-phase) and imaginary (quadrature) components separately. The field amplitude was 1300 A/m for the χ_{\perp} measurements, and ranged from 150 to 500 A/m for the in-plane measurements. The in-plane susceptibilities are labeled $\chi_{\bar{1}10}$ and χ_{100} for fields applied along the in-plane $W(\bar{1}10)$ and $W(100)$ directions, respectively.²⁹

Figure 2 contrasts the susceptibility results for an in-plane magnetized 1.5 ML Fe on a 1 ML Ni film with a perpendicularly magnetized 1.5 ML Fe on a 2 ML Ni film. The in-plane $\text{Re}\{\chi_{\bar{1}10}\}$ of Fig. 2(a) has a half width at half maximum (HWHM) of only a few kelvins and peaks to nearly 1000 in SI units. Furthermore, $\text{Re}\{\chi_{\bar{1}10}(T)\}$ is well described by a universal divergence $(T - T_c)^{-\gamma}$. $T_c = 343.9$ K and $\gamma = 1.78 \pm 0.09$ were determined for the in-plane film. The susceptibility of the in-plane film is therefore the critical susceptibility associated with the order-disorder transition between ferromagnetic and paramagnetic states at T_c . The critical exponent is in good agreement with the 2D Ising value of 7/4.

Figure 2(b) shows a markedly different behavior for χ_{\perp} of the out-of-plane film. It peaks at about 130 SI units and has a HWHM of about 30 K. Structural studies proved that the order of the in-plane films was considerably worse than that of the out-of-plane films.³⁰ Indeed, we find that the peak value of the susceptibility for 1.5 ML Fe/1 ML Ni/W(110) is a factor of 10 less than that of in-plane magnetized Fe/

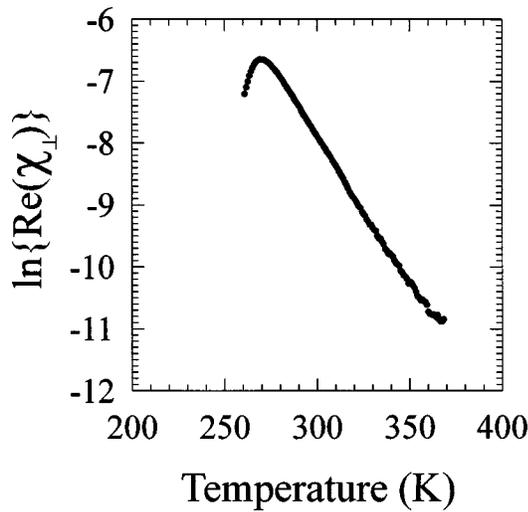


FIG. 3. A semilog plot of the real data of Fig. 2(b) indicates a broad region of exponential decay, with a decay constant of 0.04 K^{-1} . Assuming a magnetic response due to domain wall motion, exponential decay is the signature of a multidomain film with a domain density that grows exponentially with temperature (see Ref. 15). The value of the decay constant is in reasonable agreement with theoretical estimate for this film of 0.07 K^{-1} (see Ref. 15).

W(110), which has much better structural order. The narrower susceptibility peak in 2(a) compared to 2(b) is therefore not due to a difference in structural quality.

Arguments based upon the demagnetizing field predict that Figs. 2(a) and 2(b) should be fundamentally different. The external susceptibility $\chi_{\text{ext}} = dM/dH_a$, where H_a is related to the internal susceptibility χ_{int} by

$$\chi_{\text{ext}} = \frac{\chi_{\text{int}}}{1 + N\chi_{\text{int}}}, \quad (2)$$

where N is the demagnetizing factor. The internal susceptibility neglects the dipolar interaction in this calculation, and instead incorporates dipolar effects through the demagnetization field. For the in-plane case $\chi_{\text{ext}} = \chi_{\text{int}}$. Using the same χ_{int} in the perpendicular geometry with $N = 1$, we expect χ_{ext} to be always less than unity and almost independent of temperature. This shows that the mechanisms of magnetic response in Figs. 2(a) and 2(b) are different.

Instead of a broad $N = 1$ susceptibility or a universal divergence, the $\text{Re}\{\chi_{\perp}\}$ exhibits an exponential decay in a broad range of temperature, as demonstrated in the semilog plot of Fig. 3. This is the signature of the condensing domain phase as described in Sec. II. The experimental decay constant of 0.04 K^{-1} is reasonably close to the value of 0.07 K^{-1} which is estimated for this film using the theory of Abanov *et al.*¹⁴ This behavior does not persist below about 225 K, where a peak is formed in the susceptibility, and this is attributed to relaxation effects as previously described.³¹ The range of exponential decay exceeds that expected from the expansion described above, indicating domain-like behavior to 370 K.

An approximate phase diagram was constructed from a series of χ vs T measurements at thicknesses ranging from 0.25 ML Fe to 3.25 ML Fe and is shown in Fig. 4. Solid lines connect the temperatures bounding perpendicular and

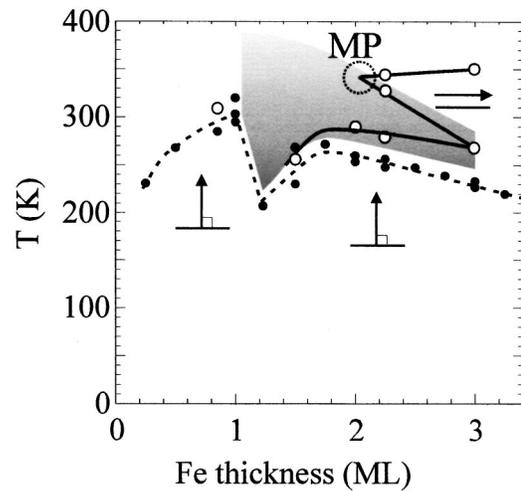


FIG. 4. An approximate phase diagram demarcates regions of perpendicular and in-plane magnetism. The solid line connects open circles (O) bounding regions of in- and out-of-plane remanence as indicated. Solid points joined with the dashed line mark maximum values in the out-of-plane susceptibility at each thickness. These points track the phase diagram indirectly (see Refs. 22 and 25). The greyed region consistently gives a broad range of exponential decay in $\text{Re}\{\chi_{\perp}\}$ and is identified as a domain phase. The circled region labeled MP is thought to be the multicritical point of this reorientation transition phase diagram.

in-plane remanence as determined from the imaginary parts of the susceptibility. The dashed line connects the temperatures where $\text{Re}\{\chi_{\perp}\}$ reaches its maximum value. We speculate that this marker tracks the reorientation transition boundary indirectly, through the dependence of an activation energy on the vanishing magnetic anisotropy. The greyed region consistently exhibits a simple exponential decay of $\text{Re}\{\chi_{\perp}\}$. This diagram is described in detail elsewhere.¹²

The focus in the present work is the greyed area identifying the condensing domain region in the strictly perpendicular 1–2 ML Fe coverage range. As shown in Figs. 2 and 3, χ does not possess a clear signature of the Curie temperature. The determination of $T_C \approx 325 \text{ K}$ for the reoriented, in-plane 2.2 ML Fe film suggests that T_C is less than 325 K for the 1.5 ML Fe film.¹² On the basis of the whole phase diagram, we expect $300 \text{ K} < T_C < 340 \text{ K}$. $\text{Re}\{\chi_{\perp}\}$ exhibits simple exponential decay and is greater than 1 SI unit well below, within and well above this range. Indeed, all of the out-of-plane films that we have studied have large perpendicular susceptibilities ($\text{Re}\{\chi_{\perp}\} \gg 1$) for several tens of kelvins above where remanence ceases and, indeed, above reasonable estimates of the Curie temperature. The exponential decay of χ is an approximation valid for $T \ll T_C$. This suggests a question: why is there no deviation from the exponential behavior with increasing T ?

A possible answer with theoretical basis is that the condensing-domain phase, which is technically paramagnetic, melts gradually and continuously into the completely disordered state with no obvious Curie temperature. Recent Monte Carlo simulations of the 2D dipolar Ising model show this behavior.¹⁹ Again, this model includes the usual exchange interaction and dipolar interactions between perpendicularly oriented moments. In the simulations, critical fluc-

tuations occur in the domain walls, rather than the interiors, because of the large local exchange energy. The divergence of the fluctuation–correlation length is then truncated when it grows comparable to the domain size. At this point, a transition in domain topology is expected, and the condensation with increasing temperature proceeds along with increasing disorder. The expected transition is analogous to the progression of transitions between smectic, nematic, and tetragonal liquid crystal phases, where domain walls unbind and domain positional order is lost (smectic→nematic) and finally orientational order is lost (nematic→tetragonal). The topological transformation does not interrupt the spatial condensation of the domain structure. Such a transformation is probably not observable in a spatially averaging measurement like the macroscopic magnetic susceptibility. The high temperature phase condenses and disorders gradually to the completely disordered state with increasing temperature with no sign of a distinct temperature, (T_C), separating domain and completely disordered phases.

This idea is compatible with Curie temperature determinations in other perpendicularly magnetized ultrathin films^{22,23} that possessed stronger perpendicular anisotropy than the 1.5 ML Fe/2 ML Ni film discussed here. The stronger anisotropy may stabilize a single-domain state to temperatures much closer to T_C : close enough to observe the reduction of $M(T)$ by critical fluctuations. The 1–2 ML Fe/2 ML Ni data may then be representative of a special region of the reorientation transition phase diagram. Because of the extremely weak perpendicular magnetic anisotropy so close to the reorientation transition, domains nucleate and unbind from defects at much lower temperatures relative to T_C than for the other out-of-plane films described in the literature.^{22,23}

Finally, we emphasize that there is a fundamental difference in the high temperature magnetic behavior of ultrathin films with in-plane versus out-of-plane magnetization, particularly when the $K_{\perp\text{eff}}$ is weak. From this work, perpendicularly magnetized films are domain-like at high temperatures. Their high temperature properties are well described by the intermediate temperature theory of Kashuba and Pokrovsky,¹⁵ and qualitatively consistent with the 2D *dipolar* Ising model, although neither model may be completely appropriate for the 1.5 ML Fe/Ni bilayer at high temperature. In contrast, in-plane magnetized films such as ultrathin Fe/W(110) are good realizations of 2D Ising magnets.^{32,33} The distinction between in-plane and perpendicularly magnetized films persists to temperatures well above T_C (or the expected T_C). In the case of the Fe/Ni films described here, this is confirmed by the measured anisotropy in χ at high temperatures.

VI. CONCLUSIONS

Perpendicularly magnetized, ultrathin films with weak $K_{\perp\text{eff}}$ disintegrate into domains at high temperature and as the reorientation transition is approached. Magnetic susceptibility measurements of perpendicularly magnetized, ultrathin fcc Fe films grown on a 2 ML Ni buffer indicate the presence of a domain phase and measure contrast from

domain-wall motion. The domain density of this phase increases exponentially with temperature in a manner quantitatively consistent with theory.¹⁵ At Fe thicknesses slightly lower than that for which the reorientation transition is first observed, where the perpendicular anisotropy is weakest, this domain phase exists for a broad range of temperature. This shows that high temperature models for perpendicularly magnetized, ultrathin films must explicitly include the dipolar interaction and 2D thermodynamics. Our experiments show qualitative and quantitative agreement with the 2D dipolar models presently available and underscore the need for high temperature theories more appropriate for real films.

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