# Magnetic instabilities in ultrathin ferromagnets

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We examine aspects of the dispersion relation of spin waves in ultrathin ferromagnetic films. When the easy axis is normal to the surfaces, and an external field is applied parallel to the surface, we find the state of uniform magnetization to be unstable, for magnetic fields in the near vicinity of that for which the magnetization just becomes parallel to the surface. We present results of a study of the temperature variation of the spin-wave dispersion relation, as a consequence of interactions between spin waves. From the structure of the result, we comment on the nature of the spin reorientation transition observed by Pappas, Kämper, and Hopster. It is noted that an earlier study of temperature-dependent renormalization of thin film parameters reported by Pescia and Pokrovsky overestimates the magnitude of the temperature corrections.

## I. INTRODUCTION

Currently, there is great interest in the magnetic properties of ultrathin ferromagnetic films grown on various nonmagnetic substrates. These films are one or, perhaps, very few atomic layers in thickness and thus provide a realization of truly-two-dimensional (2D) magnetic matter. Ultrathin films on Fe on Ag(100) (Ref. 1), Cu(100) (Ref. 2), and W(110) (Ref. 3) have been the subject of considerable activity.

If such films are truly two dimensional and if one assumes that they belong to the same universality class as the two-dimensional Heisenberg ferromagnet, then long-range magnetic order should be absent. In fact, such films exhibit long-range order. There is, in these materials, very strong uniaxial anisotropy normal to the surface. This anisotropy, counteracted by the influence of magnetic dipole couplings, may lead to an easy axis normal to the surface. The uniaxial anisotropy then leads to the long-range order in this instance.[4] The anisotropy constant may be such that the axis normal to the surface is a hard axis; the magnetization then lies in the plane. Yafet, Kwo, and Gyorgy[5] argue that in this case the presence of long-ranged dipolar couplings suppresses long-wavelength spin fluctuations sufficiently that long-range order is stabilized.

Pappas, Kämper, and Hopster observe that,[6] for an ultrathin film magnetized normal to the surface at low temperatures, the magnetization evolves into a state parallel to the surface at high temperatures. The experiments are carried out in zero external field. We refer to this phenomenon as spontaneous spin rotation. The purpose of this paper is to explore aspects of this phenomenon of spontaneous spin rotation and also to describe an instability of the state of uniform magnetization that can be induced by applying a magnetic field parallel to the surface. The origin of this instability is in the magnetic dipole interaction between moments. Its long-ranged character is essential; there is thus a relation between the instability discussed here and the domain structure discussed earlier by Yafet and Gyorgy[7] and subsequently observed by Allenspach, Stampanoni, and Bischoff[8]

Let $T_2$ be the ordering temperature of the film, and suppose $T \ll T_2$. Then spin waves are the elementary excitations of the film. We have studied the temperature dependence of the spin-wave dispersion in such films. The temperature variations have their origin in the interaction between the long-wavelength spin wave of interest here and thermally excited magnons in the film. We show how a theory of spontaneous spin rotations evolves from this analysis. We have also encountered an intriguing instability not noted in the earlier literature.

In our analysis we use a nearest-neighbor Heisenberg exchange Hamiltonian, supplemented with single-site anisotropy and dipolar coupling, recognizing this to be a phenomenological procedure for an itinerant-electron material such as Fe. Our attention is confined to the monolayer. We begin by summarizing various temperature and energy scales, since their (assumed) relative magnitudes enter our considerations importantly. There is $\hbar\Omega_M$, with $\Omega_M$ the maximum spin-wave frequency. Since exchange is very strong compared with either anisotropy or dipolar coupling, $\hbar\Omega_M \cong 4JS$ for the film with spins $S$ and exchange strength $J$. We also define $T_M$ by the relation $\hbar\Omega_M = k_B T_M$, with $k_B$ Boltzmann's constant. The ordering temperature $T_2$ is controlled by the weak anisotropy,[4] and so $T_2 < T_M$. The spin-wave regime explored here is the region $T < T_2$. Clearly, $\hbar\Omega_M$ is large compared with $k_B T$, and so quantum statistics must be used to treat the short-wavelength spin waves and their influence on the film's thermal properties. In earlier work, Pescia and Pokrovsky[9] examined temperature-dependent renormalizations of the parameters which characterize ultrathin films. In their work, classical statistical mechanics was used to describe all spin fluctuations in the film, including those of short wavelength with frequency in the range $\hbar\Omega_M$. We shall see that this led them to overestimate substantially the magnitude of the renormalization effects in the domain $T < T_2$. Finally, in general, there is a gap in the spin-wave spectrum we call $\hbar\Omega_0(0)$. This is controlled only by the combination of an-

 

isotropy, external field if present, and the dipolar couplings. Thus, it is the smallest energy in the problem. We have $\hbar\Omega_0(0) \ll k_B T$.

## II. CALCULATIONS

In earlier papers[10,11] we have examined the behavior of spin waves in very thin films with dipolar and anisotropy effects modeled microscopically, and we have explored their thermodynamic properties at temperatures sufficiently low that the spin-wave dispersion relations are well approximated by their low-temperature form.

We first begin with a discussion of the behavior of long-wavelength spin waves in the ultrathin film. We suppose the film has uniaxial anisotropy $H_A$ normal to the surface, taken to be the $z$ direction. This arises from single-site anisotropy of the form $-KS_z^2$. Then $H_A = 2KS$, and when $H_A > 0$, this anisotropy acting along will render the axis normal to the surface an easy axis. Dipolar interactions lead to an effective anisotropy field $\tilde{H}_A = H_A - 4\pi M_0 c_1$, with $M_0$ the magnetization per unit volume of the bulk material and $c_1$ a number close to unity, dependent on the microscopic structure of the film. (In macroscopic theory, $c_1 = 1$, while for the 2D square lattice, $c_1 = 0.762$). The requirement for the surface normal to be an easy axis is $\hat{H}_A > 0$. When $\tilde{H}_A < 0$, the magnetization lies in plane.

Now we imagine, as in the experimental arrangement used by Dutcher *et al.*,[12] that there is an external magnetic field $H$ applied parallel to the surface of the film in the $x$ direction. If $H_A > 0$, the magnetization lies in the $x$-$z$ plane, canted with respect to the film normal by the angle $\theta$. One has[11] $\sin\theta = H/\tilde{H}_A$, when $H < \tilde{H}_A$, and $\theta = \pi/2$, for $H > \tilde{H}_A$.

In our earlier work,[10,11] and that of others,[12] it was assumed that this picture applied for all external fields $H$. In fact, we shall see shortly that there is a most interesting instability in the system for fields in the very near vicinity of $\tilde{H}_A$. This instability in fact was encountered as an anomaly in our earlier study of spin-wave dispersion in ultrathin films, but was erroneously dismissed as a consequence of limited numerical accuracy;[13] very considerable accuracy was indeed required to generate the results in this paper, but we now appreciate that the anomaly is real.

We have obtained the long-wavelength form of the spin-wave dispersion relation in the ultrathin film for the model described above. We use the method described by Yafet, Kwo, and Gyorgy[5] to evaluate the dipole sums in the limit of small wave vector $\mathbf{k}_\parallel$. The terms linear in $\mathbf{k}_\parallel$, which enter their discussion in a central manner, enter crucially in generating the instability for external magnetic fields near $\tilde{H}_A$.

Let $\phi$ be the angle between the wave vector $\mathbf{k}_\parallel$ and $x$-$z$ plane. We find the zero-temperature spin-wave dispersion relation to be, ignoring the small contribution of the dipolar contributions to the terms quadratic in the wave vector,

$$\Omega_0(\mathbf{k}_\parallel) = [A_1(\mathbf{k}_\parallel) A_2(\mathbf{k}_\parallel)]^{1/2} , \tag{1}$$

where

$$A_1(\mathbf{k}_\parallel) = H\sin\theta + \tilde{H}_A\cos^2\theta + \frac{\pi M_0 k_\parallel a_0}{\sqrt{2}}[1-\cos(2\phi)] + D_2 k_\parallel^2 , \tag{2a}$$

$$A_2(\mathbf{k}_\parallel) = H\sin\theta + \tilde{H}_A\cos(2\theta) - \sqrt{2}\pi M_0 k_\parallel a_0 \sin^2\theta + \frac{\pi M_0 k_\parallel a_0}{\sqrt{2}}[1+\cos(2\phi)]\cos^2\theta + D_2 k_\parallel^2 . \tag{2b}$$

In these expressions, $D_2$ is the exchange constant of the monolayer, equal to $D/2$ in our model, where $D$ is the bulk exchange constant (this assumes the exchange couplings within the film have the same magnitude as in the bulk).

Suppose $\tilde{H}_A > 0$, and so in zero field we have a film with magnetization normal to the surface. Then let $H$ be well above $\tilde{H}_A$, and so $\theta = \pi/2$. The external field has forced the film into the in-plane state. Then[14]

$$A_1^{>}(\mathbf{k}_\parallel) = H + \frac{\pi M_0 k_\parallel a_0}{\sqrt{2}}[1-\cos(2\phi)] + D_2 k_\parallel^2 \tag{3a}$$

and

$$A_2^{>}(k_\parallel) = (H - \tilde{H}_A) - \sqrt{2\pi} M_0 k_\parallel a_0 + D_2 k_\parallel^2 . \tag{3b}$$

Note that $A_2^{>}(\mathbf{k}_\parallel)$ has a minimum at $k_\parallel \neq 0$ when $k_\parallel = k_\parallel^{(m)} = \pi M_0 a_0/\sqrt{2}D_2$. At the minimum,

$$A_2^{>}(k_\parallel^{(m)}) = (H - \tilde{H}_A) - \frac{\pi^2 M_0^2 a_0^2}{2D_2} . \tag{4}$$

As $H$ is lowered toward $\tilde{H}_A$ from above, $A_2^{>}(k_\parallel^{(m)})$ vanishes at a field we call the upper critical field $H_c^{>}$, given by

$$H_c^{>} = \tilde{H}_A + \frac{\pi^2 M_0^2 a_0^2}{2D_2} . \tag{5}$$

The spin-wave frequency is pure imaginary when $\tilde{H}_A < H < H_c^{>}$, and so *in the field regime* $\tilde{H}_A < H < H_c^{>}$ *the uniformly magnetized in-plane state is unstable.* We suggest that the uniform state breaks up into linear domains, whose linear dimensions are the order of $1/k_\parallel^{(m)}$. Typical parameters ($M_0 \sim 1.5k$ G, $D_2 \sim 1.5\times 10^9$ G cm$^2$, $a_0 \sim 3\times 10^{-8}$ cm) give $k_\parallel^{(m)} \sim 10^5$ cm$^{-1}$ and $H_c^{>} - \tilde{H}_A \approx 10$ G.

Now suppose $H < \tilde{H}_A$, so that the magnetization is canted and $0 < \theta < \pi/2$. If, in fact, $\theta$ is very close to but just slightly less than $\pi/2$, then, to good approximation,

$$A_2^{<}(\mathbf{k}_\parallel) \cong 2(\tilde{H}_A - H) - \sqrt{2}\pi M_0 k_\parallel a_0 + D_2 k_\parallel^2 . \tag{6}$$

We again have an off-center minimum in $A_2^{<}(k_\parallel)$ at $k_\parallel^{(m)}$, but now

$$A_2^{<}(\mathbf{k}_\parallel^{(m)}) = 2(\tilde{H}_A - H) - \frac{\pi^2 M_0^2 a_0^2}{2D_2} . \tag{7}$$

*The uniform state is now unstable above a lower critical field*

$$H_c^< = \tilde{H}_A - \frac{\pi^2 M_0^2 a_0^2}{4D_2} \ . \tag{8}$$

Thus, in the regime $H_c^< < H < H_c^>$, the uniform state is unstable with respect to breakup into a linear pattern whose length scale is set by the inverse of $k_\parallel^{(m)}$. In light-scattering experiments such as those reported in Ref. 12, one should observe anomalous elastic scattering of light in this field regime. Since $k_\parallel^{(m)}$ is $10^5$ cm$^{-1}$, analysis of its angular distribution can provide information on the nature of the magnetic structure present in the film, in principle. We have theoretical studies of the magnetic structure of the film in this field regime underway. Examination of the eigenvector of the "soft spin wave" suggests that the film acquires an out-of-plane component of the magnetization when $H_c^< < H < H_c^>$. The domain structure should thus be similar to that discussed by Yafet and Gyorgy.[7]

We now present results of our analysis of the temperature dependence of the spin-wave dispersion relation. We keep the instability just described in mind in this discussion. We have proceeded as follows. We carry out the Holstein-Primakoff transformation for the model monolayer described above. Linear terms in the annihilation and creation operators vanish if $\sin\theta = H/\tilde{H}_A$, as discussed earlier.[10] Continuing on, one encounters quadratic, cubic, and quartic terms, where we stop. We treat the quartic terms in lowest order of perturbation theory as described below to generate the temperature corrections discussed here. The cubic terms have their origin in the magnetic dipole interactions or when $\theta \neq 0$ or $\pi/2$ in the single-site anisotropy as well. We assume, as established for the bulk problem,[15] that the influence of the cubic terms taken to second order of perturbation theory is small. (Note, however, that the cubic terms taken to second order contribute to the same order in the basic expansion parameter $1/S$ as the quartic terms taken to first order.) The role of the cubic terms will be explored elsewhere,[16] where a full description of the calculations will be presented. Here we shall use the results to discuss the properties of the film.

Our earlier discussion[10] of spin waves at $T=0$, upon which the derivation of Eq. (1) is based, uses equations of motion for the annihilation and creation operators $a_{k_\parallel}^\dagger$ and $a_{-\mathbf{k}_\parallel}$, which are coupled in the presence of dipolar coupling and anisotropy. These equations of motion contain certain dynamical matrices $D_{++}^{(0)}(\mathbf{k}_\parallel)$, $D_{+-}^{(0)}(\mathbf{k}_\parallel)$, etc., from which the spin-wave dispersion is obtained after diagonalizing the appropriate $2\times 2$ matrix of these objects. We have developed a diagrammatic Green's-function method within which a self-energy matrix $\Sigma_{++}(\mathbf{k}_\parallel,\omega)$, $\Sigma_{+-}(\mathbf{k}_\parallel,\omega)$ is generated, in a manner similar to early versions of the theory of superfluidity in dilute Bose gases.[14] To lowest order in the quartic terms (the one-loop diagram and its partner, which gives exchange corrections), we generate the leading corrections to $D_{++}^{(0)}(\mathbf{k}_\parallel)$, $D_{+-}^{(0)}(\mathbf{k}_\parallel)$, etc., and evaluate these in the long-wavelength limit. The calculations are quite complex and, as remarked above, are described elsewhere.[16]

The result of this analysis, for values of $H$ sufficiently far removed from $\tilde{H}_A$ for the role of the linear terms in the spin-wave dispersion to be ignored, is that Eq. (1) continues to describe the spin-wave dispersion, but now $\tilde{H}_A$ and $D_2$ are replaced by temperature-dependent parameters $\tilde{H}_A(T)=\tilde{H}_A+\Delta H_A(T)$ and $D_2(T)=D_2+\Delta D_2(T)$. For the monolayer we thus arrive at a conclusion identical to that of Ref. 13, where it was demonstrated that at finite temperature, with small effects from cubic terms set aside, the $T=0$ formula for the spin-wave dispersion still applies to leading order in temperature corrections.

We find the results

$$\frac{\Delta\tilde{H}_A(T)}{\tilde{H}_A} = -\frac{2}{N_\parallel S}\sum_{\mathbf{k}_\parallel}\left\{\frac{A(\mathbf{k}_\parallel)}{\Omega_0(\mathbf{k}_\parallel)}\right\} n(\mathbf{k}_\parallel) \tag{9}$$

and

$$\frac{\Delta D_2(T)}{D_2} = -\frac{a_0^2}{4N_\parallel S}\sum_{\mathbf{k}_\parallel} k_\parallel^2 \left\{\frac{A(\mathbf{k}_\parallel)}{\Omega_0(\mathbf{k}_\parallel)}\right\} n(\mathbf{k}_\parallel) \ , \tag{10}$$

where

$$A(\mathbf{k}_\parallel) = H\sin\theta + \tilde{H}_A(1-\tfrac{3}{2}\sin^2\theta) + \sqrt{2}\pi M_0 k_\parallel a_0[1-\tfrac{3}{2}\sin^2\theta - \tfrac{1}{2}\cos(2\phi)\sin^2\theta] + D_2 k_\parallel^2 \ .$$

Here $N_\parallel$ is the number of unit cells in the film [the lattice is arranged so we have an atom at the origin and four neighbors at $(a_0/2)(\pm\hat{\mathbf{x}}\pm\hat{\mathbf{y}})$], and

$$n(\mathbf{k}_\parallel) = \{\exp[\hbar\Omega_0(\mathbf{k}_\parallel)/k_B T]-1\}^{-1}$$

is the Bose-Einsein function.

We may also calculate, within standard spin-wave theory (no renormalization effects), the temperature variation of the film magnetization $M_0(T)$. We find $M_0(T)/M_0 = 1-\Delta(T)$, where $\Delta(T)$ has a form identical to the right-hand side of Eq. (9), except the factor of 2 is missing. Thus, at least initially, we find that $H_A(T)$ decrease with temperature twice as fast as the film magnetization. This will be important for what follows.

Consider the case where the external field $H$ vanishes and $\tilde{H}_A > 0$; so $\theta = 0$. Then $\Omega_0(\mathbf{k}_\parallel) = \tilde{H}_A + D_2 k_\parallel^2$, and the right-hand sides of Eqs. (9) and (10) are readily evaluated to give, in the limit $\hbar\Omega_0(0) \ll k_B T$,

$$\frac{\tilde{H}_A(T)}{\tilde{H}_A} = 1 - \frac{a_0^2 k_B T}{4\pi\hbar D_2 S}\ln\left[\frac{k_B T}{\hbar\Omega_0(0)}\right] \tag{11}$$

and

$$\frac{D_2(T)}{D_2} = 1 - \frac{a_0^4}{32\pi S}\left[\frac{k_B T}{\hbar D_2}\right]^2 \zeta(2) \ . \tag{12}$$

Here $\zeta(2)$ is the Riemann zeta function of argument 2. We turn next to a discussion of the implication of these results.

## III. RESULTS AND DISCUSSION

We first compare our results with those of Pescia and Pokrovsky.[9] These authors begin with a model Hamiltonian with dipole-dipole interactions of strength $\Omega$ (analogous to our parameter $M_0$ in the spin-wave dispersion), uniaxial anisotropy of strength $\lambda$ (analogous to our $H_A$), and exchange couplings of strength $\Gamma$ (analogous to our $D_2$). They generate temperature dependences of these $T=0$ parameters by means of a renormalization-group analysis.

We find our $H_A$ and $M_0$ (as they enter the equations of motion for spin waves) to be renormalized in precisely the same manner, and so in the end the renormalized spin-wave dispersion relation may be expressed in terms of the single effective anisotropy field $\tilde{H}_A(T)$. Pescia and Pokrovsky find the same functional dependence on temperature for their two parameters $\lambda$ and $\Omega$, but in fact different numerical prefactors enter into each [see the first two entries in their Eq. (2)]. We have not reproduced their calculation; unfortunately, few details appear in their paper. We regard this difference between the two analyses as technical in nature, and we offer no further comments on this point. The temperature dependence of their renormalization corrections to $\lambda$ and $\Omega$ differs from ours; in place of the factor $\ln[k_BT/\hbar\Omega(0)]$ in our Eq. (11), they have $\ln(L/a)$, with $a$ the lattice constant and $L$ the characteristic length scale which describes spin fluctuations.

A result equivalent to theirs follows from our Eq. (9) (with $H=\theta=0$) if we treat all spin fluctuations, including the high-energy short-wavelength fluctuations, in a classical manner. This is done by replacing the Bose-Einstein function $n(\mathbf{k}_\parallel)$ by its classical limit everywhere, $k_BT/\hbar\Omega_0(\mathbf{k}_\parallel)$. When this is done and the Brillouin zone is replaced by a circle with radius $k_M$, we find Eq. (11) to be replaced by

$$\frac{\tilde{H}_A(T)}{\tilde{H}_A}=1-\frac{a_0^2k_BT}{2\pi\hbar D_2S}\ln[k_M\xi^<]\ , \tag{13}$$

where $\xi^<=[D_2/\Omega_0(0)]^{1/2}/2$ is the correlation length which entered our earlier discussion of the nature of spin fluctuations in ultrathin films at low temperatures [see Eq. (38a) of Ref. 9, and suppose the external field is absent].

The result in Eq. (13) is equivalent to the result found by Pescia and Pokrovsky, save for the difference in numerical prefactors mentioned above. Their application of classical statistical mechanics to all spin fluctuations, including the large number of spin fluctuations with excitation energy $\hbar\Omega_M \gg k_BT$, has led them to overestimate substantially the magnitude of the renormalization effects in these systems.

We are not the first to note that quantum theoretic methods are required in the analysis of spin fluctuation effects in ultrathin films at low temperatures. For example, in their study of the two-dimensional spin-$\frac{1}{2}$ antiferromagnet, Chakravarty, Halperin, and Nelson[18] note the necessity of using quantum theoretic methods in such problems and have developed a renormalization-group scheme appropriate for the problem addressed in their paper.

Pescia and Pokrovsky also examine the temperature renormalization of their exchange parameter $\Gamma$, to find a correction very similar to that displayed in our Eq. (13), and which thus differs qualitatively from the behavior given in our Eq. (12). For the ratio $D_2(T)/D_2$, we obtain a result equivalent to theirs if we ignore the contribution from the exchange diagram to the proper self-energy. There is a partial cancellation between the leading exhange contribution to the proper self-energy and the "one-loop" (Hartree) contribution which leads to a small residue proportional to $T^2$, rather than $T\ln[k_BT/\hbar\Omega_0(0)]$ [or to $T\ln(k_M\xi^<)$ if the influence of quantum statistics is ignored]. This cancellation is well known in the theory of spin-wave renormalization in bulk magnetic materials. Exchange contributions to the energy are not included within classical renormalization-group analyses, unfortunately.

We conclude, then, that at least at temperatures well below $T_2$, the two-dimensional ordering temperature, the analysis of Pescia and Pokrovsky leads to overestimates of the magnitude of the temperature renormalizations. It would be of great interest to see this problem reexamined, within the framework of a quantum theoretic scheme such as that in Ref. 16. We believe that at low temperatures our spin-wave renormalization scheme provides the correct leading terms.

As we remarked above, our analysis shows that at low temperatures, $[\tilde{H}_A(T)/H_A]$ decreases more rapidly than the reduced magnetization $M_0(T)/M_0$. [Recall our mention of the factor of two in Eq. (9)]. There is then the possibility that $\tilde{H}_A(T)$, initially positive at low temperatures, may vanish at a temperature $T_R$ below $T_2$; $\tilde{H}_A(T)$ will then be negative for $T>T_R$. We conclude by exploring the implications of this behavior within the framework of the concepts discussed above.

We suppose that the external field $H=0$ and that the spin-wave dispersion is described by Eq. (1) with $\tilde{H}_A$ replaced by $\tilde{H}_A(T)$, $M_0$ by $M_0(T)$, in the linear terms in $k_\parallel$, and $D_2$ by $D_2(T)$. (Our analysis proves the correctness of the first and third requirement at low temperatures.) Then, for $T<T_R$, $\tilde{H}_A(T)$ is positive and $\theta=0$. Everywhere for $T<T_R$ the spin waves are stable, and at $k_\parallel=0$ we have a gap proportional to $\tilde{H}_A(T)$. The gap thus vanishes as $T_R$ is approahced from below; long-range order will be absent at $T_R$, and critical scattering of light such as that reported earlier[12] will be realized as $T_R$ is approached.

Well above $T_R$, $\tilde{H}_A(T)$ is negative, so that $\theta=\pi/2$. We then have, in Eq. (1),

$$A_1(\mathbf{k}_\parallel)=\frac{\pi M_0(T)k_\parallel a_0}{\sqrt{2}}[1-\cos(2\phi)]+D_2k_\parallel^2 \tag{14}$$

and

$$A_2(\mathbf{k}_\parallel)=|\tilde{H}_A(T)|-\sqrt{2}\pi M_0(T)k_\parallel a_0+D_2k_\parallel^2\ . \tag{15}$$

Now, as $T$ is lowered toward $T_R$ from above, *there is a temperature region just above $T_R$ where the state of uni-*

*form magnetization is unstable,* for reasons identical to those encountered earlier in our discussion of field-induced canting at $T \equiv 0$. For temperatures near $T_R$, we may write $|\tilde{H}_A(T)| = |(d\tilde{H}_A/dT)_{T_R}|(T - T_R)$, and then upon following reasoning identical to that given earlier, we find the width $\Delta T$ of the unstable region is

$$\frac{\Delta T}{T_R} = \frac{\pi^2}{2}\frac{M_0^2 a_0^2}{D_2}\frac{1}{T_R|(d\tilde{H}_A/dT)|_{T_R}|} . \tag{16}$$

Pappas, Kämper, and Hopster,[6] in their study of spontaneous spin rotation, find behaviors consistent with the above scenario. As they approach $T_R$ from below, the order parameter drops to zero continuously as $T_R$ approached, a behavior consistent with a spin excitation spectrum whose gap vanishes at $T_R$. They do see a temperature region within which there is apparently no long-range order in the film. We suggest that this is the regime where $|\tilde{H}_A(T)|$ is so small that the dipolar instability has set in. Then, with increasing temperature, the spin-wave spectrum is stable at all $\mathbf{k}_\parallel$ and the gap grows with increasing temperature, as does the magnetization parallel to the surface.

It is difficult to estimate $\Delta T$ from Eq. (16), since we do not know $(d\tilde{H}_A/dT)_{T_R}$ for these films. Crude reasoning suggests that $T_R(d\tilde{H}_A/dT)_{T_R}$ has a magnitude comparable to the low-temperature value of $\tilde{H}_A$ itself, which we guess to be $\approx 10^3$ G for the films, which at $T=0$ are quite close to the thickness at which the perpendicular configuration is unstable. Then, if we take $M_0 \cong 1.5\times 10^3$ G, $a_0 \simeq 3\times 10^{-8}$ cm, and $D_2 \cong 10^{-9}$ G cm$^2$ (again a parameter whose value is not known), then Eq. (16) gives $(\Delta T/T_R) \sim 5\times 10^{-3}$. The best quality data displayed in the paper by Pappas, Kämper, and Hopster would seem to be that for the six-atomic-layer Fe film on Cu(100), and the gap in temperature where long-range order appears absent appears perhaps an order of magnitude larger than this estimate. (We are referring to the lower panel in Fig. 1, which describes the sample as having three atomic layers of Fe. It was noted later[19] that in fact six layers of Fe were present on this particular sample.) In our picture the order parameter drops to zero as $T \to T_R$ from below and again as $T \to T_R + \Delta T$ from above. The true width of the unstable region may be difficult to extract from the data reliably. Also, the film may be inhomogeneous, and so the effective value of $T_R$ may vary from region to region on the film.

The film may also have the character of an island film, with islands of size $L$ coupled loosely; spins within an island could be coupled tightly. One would then replace the lattice constant $a_0$ in Eq. (16) by $L$, and if $L \cong 10a_0$, the value of $\Delta T$ would be comparable to experiment. If this conjecture is correct, then there should be a corresponding increase in the gap $(H_c^> - H_c^<)$ within which field-induced spin rotations produce a spatial instability. This could be checked experimentally.

The discussion in this paper has centered on the consequences of the fact that, in two dimensions, dipolar couplings can lead to anomalous negative dispersion in spin waves. In the thin film, this leads to the instabilities we have discussed. It is interesting to note that in a very different context such negative dispersion has been measured directly.[20] Oxygen atoms on metal surfaces have large time-dependent electric dipole moments as they vibrate normal to the surface; there are thus strong dipole-dipole couplings betwen the oxygen atoms. Electron-energy-loss studies of surface phonons of the $c(2\times 2)$ O overlayer on Ni(100) provide clear evidence for the presence of negative dispersion at small $\mathbf{k}_\parallel$ very similar to that explored here, although in this system this leads to no instabilities of the sort explored here.

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