

## Dipolar interactions and the magnetic behavior of two-dimensional ferromagnetic systems

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A simple model is presented for spin-wave excitations and the low-temperature magnetization of ferromagnetic ultrathin films. The films are assumed to be quasi-two-dimensional arrays of fixed magnetic moments and both exchange and dipolar interactions are taken into account. Discrete and continuous models are used to examine the wave-vector dependence of the spin-wave frequency in the long-wavelength limit. This spin-wave theory can be used to calculate the temperature dependence of the saturation magnetization in the limit of small fluctuations. The role of dipolar interactions is examined for a single monolayer and for thin films consisting of two or three atomic layers.

### I. INTRODUCTION

Theoretical investigations of magnetic ordering in two dimensions suggested that exchange interactions alone were found to be insufficient to establish a stable ferromagnetic ground state.<sup>1-3</sup> Experimental evidence exists, however, for long-range magnetic order in several two-dimensional and quasi-two-dimensional systems.<sup>4-6</sup> Although the existence of magnetic anisotropies can lead to stable two-dimensional ordering, the importance of dipolar interactions in two-dimensional magnetic systems has also been recently recognized. The majority of this work has dealt with the influence of dipolar interactions on the Curie temperature of low-dimensional magnetic systems, and it has been argued that dipolar interactions help stabilize the two-dimensional order.<sup>7,8</sup>

The low-temperature properties of the magnetization can be examined with spin-wave theory, but when only short-ranged exchange interactions between spins are considered, one again has difficulties with stability in two dimensions. The spin-wave dispersion in three dimensions has a quadratic dependence on the wave vector, i.e.,  $\omega \sim Dq^2$ , where  $\omega$  is the frequency,  $D$  is the exchange constant, and  $\mathbf{q}$  is the wave vector. This leads to divergence problems at small wave vectors when this dispersion is used to evaluate the low-temperature magnetization in two dimensions.

The influence of dipolar interactions on the low-temperature behavior of the magnetization in two-dimensional systems has been shown to remove these divergence problems.<sup>9,10</sup> Furthermore, for ultrathin films consisting of several atomic layers, there exists a lower-frequency spin-wave mode which is thermally accessible at room temperatures and below. This mode is known as the Damon-Eshbach mode on thick films and the dipole-exchange "surface" mode on very thin films.<sup>11</sup> This mode exists for wavelengths in the  $10^{-5}$ -cm range for Fe and Co, and both short-range exchange interactions and long-range dipolar interactions are significant. As will be shown below, this mode is identical with the mode predicted for the monolayer and can be used to calculate the low-temperature magnetization of two-dimensional magnetic structures consisting of several atomic layers with a

relatively simple formalism.

The purpose of this paper is to present a simple macroscopic argument which gives the dispersion relation for the dipole-exchange spin-wave mode of a two-dimensional ferromagnetic system. The resulting expression is shown to be identical with that obtained from a microscopic treatment in the limit of long wavelengths. In particular, the low temperature magnetization can be calculated according to spin-wave theory without divergence problems.<sup>9</sup> Also, since this mode is a low-energy, long-wavelength excitation, the spins at the surfaces of the film precess in phase with the spins in the center of the film, so that the amplitude of the mode is very nearly independent of position in the film. This is an important point when studying this long-wavelength spin-wave mode on a thin film consisting of several atomic layers, since then the magnetic properties at and near the surfaces can shift the spin-wave frequency in the same manner as bulk anisotropies.

In what follows the dispersion relation for the long-wavelength spin-wave mode is first derived from a discrete model and the limiting form for small wave vectors is examined. Next, a simple continuum model is presented which gives the same dispersion law for long wavelengths and the results are compared to those for thin films consisting of several atomic layers. Finally, the low-temperature behavior of the magnetization in a two-dimensional structure calculated from spin-wave theory is discussed.

### II. DISCRETE MODEL

The geometry is such that a square array of magnetic spins lie in the  $y$ - $z$  plane and the spin-wave propagation vector  $\mathbf{q}$  makes an angle  $\phi$  with the  $z$  axis. The saturation magnetization  $M_s$  and a static applied magnetic field  $H_0$  lie in the  $z$  direction.

The following treatment is a limiting case of an approach developed by Benson and Mills, who used a similar model to investigate the spin-wave modes of a thin film.<sup>12</sup> Erickson and Mills have also recently applied this model to thin-film systems consisting of only a few atomic layers, with an emphasis on the effects of anisotropies

on the magnetic ground state.<sup>13</sup>

One begins with the Hamiltonian defined by

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{dip}} + \mathcal{H}_z \quad (1)$$

where the exchange terms are given by

$$\mathcal{H}_{\text{ex}} = \frac{1}{2} \sum_{jj'} J_{jj'} \mathbf{S}(\mathbf{j}) \cdot \mathbf{S}(\mathbf{j}') . \quad (2)$$

The exchange integral between spins at sites  $\mathbf{j}$  and  $\mathbf{j}'$  is denoted by  $J_{jj'}$ . The energy due to the presence of the static applied field is given by

$$\mathcal{H}_z = g\mu_B \sum_{\mathbf{j}} \mathbf{H}_0 \cdot \mathbf{S}(\mathbf{j}) . \quad (3)$$

$g$  is the gyromagnetic ratio and  $\mu_B$  the Bohr magnetron. Finally, the dipole-dipole interaction energy is given by

$$\mathcal{H}_{\text{dip}} = \frac{1}{2} g^2 \mu_B^2 \sum_{jj'} \left[ \frac{\mathbf{S}(\mathbf{j}) \cdot \mathbf{S}(\mathbf{j}')}{r_{jj'}^3} - 3 \frac{[\mathbf{r}_{jj'} \cdot \mathbf{S}(\mathbf{j})][\mathbf{r}_{jj'} \cdot \mathbf{S}(\mathbf{j}')] }{r_{jj'}^5} \right] . \quad (4)$$

The vector  $\mathbf{r}_{jj'}$  points from a spin at site  $\mathbf{j}$  to a spin at site  $\mathbf{j}'$ , and the distance between two spins is given by  $r_{jj'}$ .

Explicitly,

$$\mathbf{r}_{jj'} = \hat{\mathbf{y}}a(j_y - j'_y) + \hat{\mathbf{z}}a(j_z - j'_z) . \quad (5)$$

$\hat{\mathbf{y}}$  and  $\hat{\mathbf{z}}$  unit vectors in the  $y$  and  $z$  directions, the lattice spacing is given by  $a$ , and  $j_y$  and  $j_z$  are integers.

The equations of motion are next formed by commuting  $\mathbf{S}(\mathbf{j})$  with the Hamiltonian of Eq. (1) and linearizing under the assumptions that the transverse oscillations of  $\mathbf{S}(\mathbf{j})$  are small and  $\langle S_z(\mathbf{j}) \rangle \approx S$ . Note that in the long-wavelength limit, the spin operators behave as classical vectors. Finally, the translational invariance of the system in the  $y$  and  $z$  directions is used to write  $S_x(\mathbf{j})$  and  $S_y(\mathbf{j})$  in the form

$$S_x(\mathbf{j}) = A \exp[i(\omega t - \mathbf{q}_{\parallel} \cdot \mathbf{r}_{\mathbf{j}})] , \quad (6)$$

$$S_y(\mathbf{j}) = B \exp[i(\omega t - \mathbf{q}_{\parallel} \cdot \mathbf{r}_{\mathbf{j}})] . \quad (7)$$

$A$  and  $B$  are constants, the position vector  $\mathbf{r}_{\mathbf{j}} = \mathbf{r}_{\mathbf{j}, j'=0}$ , and the wave vector  $\mathbf{q}_{\parallel}$  is defined as a two-dimensional vector, appropriate to a two-dimensional square array of spins:

$$\mathbf{q}_{\parallel} = \hat{\mathbf{y}}q_y + \hat{\mathbf{z}}q_z . \quad (8)$$

The resulting equations of motion are then

$$\frac{1}{g^2 \mu_B^2} \omega A = iS \sum_{j'} \left[ \frac{1}{g\mu_B} \delta_{jj'} H_0 - \frac{1}{2} \left[ \frac{1}{g^2 \mu_B^2} J_{jj'} - \frac{1}{r_{jj'}^3} + 3 \frac{(\mathbf{r}_{jj'} \cdot \hat{\mathbf{y}})^2}{r_{jj'}^5} \right] e^{i(j'-j) \cdot \mathbf{q}_{\parallel}} + \frac{1}{2} \left[ \frac{1}{g^2 \mu_B^2} J_{jj'} - \frac{1}{r_{jj'}^3} + 3 \frac{(\mathbf{r}_{jj'} \cdot \hat{\mathbf{z}})^2}{r_{jj'}^5} \right] \right] B , \quad (9)$$

$$\frac{1}{g^2 \mu_B^2} \omega B = iS \sum_{j'} \left[ -\frac{1}{g\mu_B} \delta_{jj'} H_0 + \frac{1}{2} \left[ \frac{1}{g^2 \mu_B^2} J_{jj'} - \frac{1}{r_{jj'}^3} \right] e^{i(j'-j) \cdot \mathbf{q}_{\parallel}} - \frac{1}{2} \left[ \frac{1}{g^2 \mu_B^2} J_{jj'} - \frac{1}{r_{jj'}^3} + 3 \frac{(\mathbf{r}_{jj'} \cdot \hat{\mathbf{z}})^2}{r_{jj'}^5} \right] \right] A . \quad (10)$$

Cubic symmetry has been assumed, which means that dipolar terms of the form  $(\mathbf{r}_{jj'} \cdot \hat{\mathbf{y}})(\mathbf{r}_{jj'} \cdot \hat{\mathbf{z}})$  cancel in the sums.

The dispersion relation  $\omega(\mathbf{q})$  for the spin-wave excitation on a two-dimensional square lattice is easily found and takes the remarkably simple form (where  $\gamma = g\mu_B / \hbar$  and  $\hbar$  is Planck's constant)

$$\omega = \gamma \sqrt{(H_0 + \Sigma_{\text{ex}} + \Sigma_{d1})(H_0 + \Sigma_{\text{ex}} + \Sigma_{d2})} . \quad (11)$$

The exchange contribution to the energy is

$$\Sigma_{\text{ex}} = \frac{1}{2} \sum_{\mathbf{j}} J_{jj'} [1 - \exp(i\mathbf{q}_{\parallel} \cdot \mathbf{r}_{jj'})] , \quad (12)$$

and the dipolar contributions are

$$\begin{aligned} \Sigma_{d1} = & [\sigma_z(\mathbf{q}_{\parallel}) - \sigma_z(0)] + [\sigma_y(\mathbf{q}_{\parallel}) - \sigma_y(0)] \\ & + 3[\sigma_z(0) - \sigma_y(\mathbf{q}_{\parallel})] , \end{aligned} \quad (13)$$

$$\Sigma_{d2} = [\sigma_z(\mathbf{q}_{\parallel}) - \sigma_z(0)] + [\sigma_y(\mathbf{q}_{\parallel}) - \sigma_y(0)] + 3\sigma_z(0) . \quad (14)$$

The quantities  $\sigma_y$  and  $\sigma_z$  are defined by

$$\sigma_y(\mathbf{q}_{\parallel}) = \frac{1}{2} S \mu_B \sum_{j'} \frac{e^{ir_{jj'} \cdot \mathbf{q}_{\parallel}}}{r_{jj'}^5} (\mathbf{r}_{jj'} \cdot \hat{\mathbf{y}})^2 , \quad (15)$$

$$\sigma_z(\mathbf{q}_{\parallel}) = \frac{1}{2} S \mu_B \sum_{j'} \frac{e^{ir_{jj'} \cdot \mathbf{q}_{\parallel}}}{r_{jj'}^5} (\mathbf{r}_{jj'} \cdot \hat{\mathbf{z}})^2 . \quad (16)$$

From the structure of Eqs. (14)–(16), one sees that, as  $\mathbf{q}$  goes to zero, most of the dipolar terms cancel. The terms which do not cancel represent the demagnetizing fields that are expected for a thin planar structure. In particular, for  $qa \ll 1$ , the frequency of the spin-wave mode becomes

$$\omega = \gamma \sqrt{(H_0 + \Sigma_{\text{ex}})[H_0 + \Sigma_{\text{ex}} + 3\sigma_z(0)]}. \quad (17)$$

The quantity  $3\sigma_z(0)$  can be evaluated numerically using the methods of Ref. 12 and is the demagnetizing field for a square planar array of magnetic moments. In the next section, an expression identical to Eq. (17) will be derived using a continuum model and the macroscopic Maxwell electromagnetic equations.

Implicit in the derivation of Eq. (17) is the equality

$$\sigma_y(0) = \sigma_z(0). \quad (18)$$

This equality holds *only* when the lattice is highly symmetric. A distortion of the lattice from cubic symmetry invalidates Eq. (18) and leads to additional terms in the frequency. In fact, in the limit of very long wavelengths, one can use the techniques described in Ref. 12 to show that the sums for a rectangular lattice with lattice spacings  $a$  and  $a'$  are

$$\sigma_y(q_{\parallel}=0) = \left[ \frac{4\pi^2}{9a'a^2} + \frac{32\pi^2}{3a'^3} \sum_{m,n} n^2 K_2(2\pi nma/a') \right] S\mu_B, \quad (19)$$

$$\sigma_z(q_{\parallel}=0) = \left[ \frac{4\pi^2}{9a'^2a} + \frac{32\pi^2}{3a^3} \sum_{m,n} n^2 K_2(2\pi nma'/a) \right] S\mu_B. \quad (20)$$

Here  $K_2$  are modified Bessel functions of order 2.

With  $a \neq a'$ , these terms are no longer equal and create a uniaxial anisotropy due to dipolar interactions. A feeling for the magnitude of these anisotropies can be obtained by examining the energy at  $q_{\parallel}=0$  given by Eq. (11) for small differences between  $a$  and  $a'$ . Calculations show that for differences between  $a$  and  $a'$  on the order of 2% or 3%, which is a possible value for monolayers grown on (110) substrates, the dipolar anisotropy energy is on the order of  $0.1M_s^2$ , which is roughly the same magnitude as bulk anisotropies. This value is one to two orders of magnitude smaller than typical surface and interface anisotropies, however.

In the long-wavelength limit of  $q_{\parallel}a \ll 1$ , the exchange contribution given by Eq. (12) has the usual form

$$\Sigma_{\text{ex}} \sim q_{\parallel}^2. \quad (21)$$

For moderate  $q_{\parallel}a$ , the exchange contribution dominates, and it is easy to see from the dispersion relation of Eq. (11) that

$$\omega \sim q_{\parallel}^2. \quad (22)$$

As  $q_{\parallel}a$  goes to zero, it can be shown that<sup>9</sup>

$$\omega \sim \sqrt{q_{\parallel}}. \quad (23)$$

This dependence on  $q_{\parallel}$  is the essential feature of the dispersion of Eq. (11) since it is these modes which will dominate the low-temperature behavior of the magnetization. It can also be shown from Eqs. (9) and (10) that in the limit of large wavelengths,  $B \gg A$  for small  $H_0$ . This

means that the modes are highly elliptical for small  $H_0$ , so that the precession of the spins is primarily out of the film plane.

### III. CONTINUUM MODEL

The long-wavelength nature of the modes described by Eq. (17) suggests that it should be possible to describe the spin-wave excitations with a semiclassical continuum model. Such a model is constructed as follows: First, the Landau-Lifshitz equations are written in terms of the time and spatially varying magnetization  $\mathbf{M}$ , the gyromagnetic factor  $\gamma$ , and an effective field  $\mathbf{H}_{\text{eff}}$ :

$$\dot{\mathbf{M}} = \gamma \mathbf{M} \times \mathbf{H}_{\text{eff}}, \quad (24)$$

where the effective field is defined by

$$\mathbf{H}_{\text{eff}} = \hat{z}H_0 + \frac{2A}{M_s} \nabla^2 \mathbf{M} + \mathbf{h}_d. \quad (25)$$

$\mathbf{h}_d$  is the dynamic demagnetizing field in the  $x$ - $y$  plane,  $M_s$  is the saturation magnetization, and the exchange constant  $A$  is defined for a simple cubic lattice by

$$\frac{2A}{M_s} \gamma = \frac{2JSa^2}{\hbar}, \quad (26)$$

where  $J$  is the exchange constant and  $S$  the spin number. The magnetization is assumed to consist of a static part in the  $z$  direction and a small fluctuating part  $\mathbf{m}$  in the  $x$ - $y$  plane. Both  $\mathbf{m}$  and  $\mathbf{h}_d$  are assumed to depend on the three-dimensional position vector  $\mathbf{x}$  according to  $\exp[i(\omega t - \mathbf{q}_{\parallel} \cdot \mathbf{x}_{\parallel})]$ , where

$$\mathbf{x} = \hat{x}x + \mathbf{x}_{\parallel}, \quad (27)$$

$$\mathbf{x}_{\parallel} = \hat{y}y + \hat{z}z. \quad (28)$$

Using Eq. (25) in Eq. (24) and linearizing, one has

$$i \frac{\omega}{\gamma} m_x = - \left[ H_0 + 2 \frac{A}{M_s} \left( q_{\parallel}^2 - \frac{\partial^2}{\partial x^2} \right) \right] m_y + h_y, \quad (29)$$

$$i \frac{\omega}{\gamma} m_y = \left[ H_0 + 2 \frac{A}{M_s} \left( q_{\parallel}^2 - \frac{\partial^2}{\partial x^2} \right) \right] m_x - h_x. \quad (30)$$

For films which are only one or two monolayers thick, the fields  $\mathbf{m}$  and  $\mathbf{h}_d$  are expected to either vary slowly or have negligible amplitudes across the film thickness  $d$  for the surface mode excitation. For long-wavelength excitations, it is then appropriate to define fields which are averages taken across the film thickness:<sup>14</sup>

$$\langle \mathbf{m} \rangle_{\text{film}} = \frac{1}{d} \int_{-d/2}^{d/2} dx \mathbf{m}, \quad (31)$$

$$\langle \mathbf{h}_d \rangle_{\text{film}} = \frac{1}{d} \int_{-d/2}^{d/2} dx \mathbf{h}_d. \quad (32)$$

These averages will now be calculated explicitly under the assumption that the magnetic excitations can be described by plane-wave solutions of the equations of motion. First, however, it is useful to note that the wavelengths of the spin-wave excitations of interest are long enough such that the magnetostatic approximation is val-

id and the dipolar fields  $\mathbf{h}_d$  obey  $\nabla \times \mathbf{h}_d = 0$ . It is then convenient to define a magnetic potential  $\psi$  such that  $\mathbf{h}_d = -\nabla\psi$ .

The plane-wave solutions for the magnetic potential  $\psi$  and magnetization  $\mathbf{m}$  inside a film with surfaces at  $x = \pm d/2$  are given by

$$\psi = (\psi_+ e^{\alpha x} + \psi_- e^{-\alpha x}) e^{iq_{\parallel} x_{\parallel}}, \quad (33)$$

$$\mathbf{m} = \left[ \hat{\mathbf{x}}(a_+ e^{\alpha x} + a_- e^{-\alpha x}) + \hat{\mathbf{y}}(b_+ e^{\alpha x} + b_- e^{-\alpha x}) \right] e^{iq_{\parallel} x_{\parallel}}. \quad (34)$$

Here  $\alpha$  is a decay constant which is as yet undetermined.

Substituting these expressions into the equations of motion [Eqs. (29) and (30)] and integrating across the film thickness, one obtains

$$i \frac{\omega}{\gamma} (a_+ + a_-) + \left[ H_0 - \frac{2A}{M_s} (q_{\parallel}^2 - \alpha^2) \right] (b_+ + b_-) + iq_y M_s (\psi_+ + \psi_-) = 0, \quad (35)$$

$$\left[ H_0 - \frac{2A}{M_s} (q_{\parallel}^2 - \alpha^2) \right] (a_+ + a_-) - i \frac{\omega}{\gamma} (b_+ + b_-) + \alpha M_s (\psi_+ - \psi_-) = 0. \quad (36)$$

The behavior of the magnetization at the surfaces of the film is described with exchange boundary conditions of the Rado-Weertman form.<sup>15</sup> This allows the inclusion of surface anisotropies which can act to "pin" the surface spins. For simplicity, an out-of-plane directed uniaxial surface anisotropy  $k_s$  is defined:

$$\pm A \left[ \frac{\partial}{\partial x} m_y \right]_{x=\pm d/2} = k_s (m_y)_{x=\pm d/2}. \quad (37)$$

Using the forms for  $\mathbf{m}$  given by Eq. (33), these boundary conditions provide four new equations from which two equations can be derived which involve  $(a_+ + a_-)$  and  $(b_+ + b_-)$ . These two equations are substituted into the averaged equations of motion [Eqs. (35) and (36)], yielding

$$i \frac{\omega}{\gamma} (a_+ + a_-) + \left[ H_0 + \frac{2A}{M_s} q_{\parallel}^2 \right] (b_+ + b_-) + iq_y M_s (\psi_+ + \psi_-) = 0, \quad (38)$$

$$\left[ H_0 + \frac{2A}{M_s} q_{\parallel}^2 - \alpha \frac{k_s}{M_s} \coth \left[ \frac{\alpha d}{2} \right] \right] (a_+ + a_-) - i \frac{\omega}{\gamma} (b_+ + b_-) + \alpha M_s (\psi_+ - \psi_-) = 0. \quad (39)$$

The hyperbolic cotangents are now expanded in powers of  $\alpha d$ , and terms up to first order are kept. The resulting expressions are

$$i \frac{\omega}{\gamma} (a_+ + a_-) + \left[ H_0 + \frac{2A}{M_s} q_{\parallel}^2 \right] (b_+ + b_-) + iq_y M_s (\psi_+ + \psi_-) = 0, \quad (40)$$

$$\left[ H_0 + \frac{2A}{M_s} q_{\parallel}^2 - \frac{2k_s}{M_s d} \right] (a_+ + a_-) - i \frac{\omega}{\gamma} (b_+ + b_-) + \alpha M_s (\psi_+ - \psi_-) = 0. \quad (41)$$

Equations (40) and (41) are the Landau-Lifshitz equations of motion with exchange boundary conditions, averaged over the magnetic film thickness. This same procedure is next applied to the Maxwell equation of motion,  $\nabla \cdot \mathbf{B} = 0$ , and the electromagnetic boundary conditions. The macroscopic  $\mathbf{B}$  field is here defined appropriate to an ultrathin-film geometry:

$$\mathbf{B} = \mathbf{h}_d + 4\pi f \mathbf{m}. \quad (42)$$

$f$  is a demagnetization factor for a thin-film geometry. In the thick-film limit,  $f$  is 1, and in the monolayer limit, the calculated value of  $f$  is 0.5392. It is noted that averaged values for cases in between have been calculated by an alternative method and are tabulated in Ref. 14. The important point for what follows, however, is the identity<sup>12</sup>

$$f = \frac{3}{4\pi S \mu_B} \sigma_z(0). \quad (43)$$

It will be seen below that this identity connects the microscopic approach described in the previous section with the macroscopic continuum theory of this section.

In order to proceed, the fields outside the film need to be defined. In these regions, the magnetization is zero and the magnetic potential has the form

$$\psi_{<} = \psi_{<} e^{-\beta x} e^{iq_{\parallel} x_{\parallel}}, \quad (44)$$

for the region above the film, and

$$\psi_{>} = \psi_{>} e^{\beta x} e^{iq_{\parallel} x_{\parallel}}, \quad (45)$$

for the region beneath the film. The decay constant  $\beta$  is determined from  $\nabla \cdot \mathbf{B} = 0$  for the nonmagnetic regions. This gives  $\beta = |\mathbf{q}_{\parallel}|$ .

The electromagnetic boundary conditions require the normal components of  $\mathbf{B}$  and the tangential components of  $\mathbf{h}_d$  to be continuous across the surfaces at  $x = \pm d/2$ . This results in four equations involving  $\psi_{<}$ ,  $\psi_{>}$ ,  $\psi_+$ ,  $\psi_-$ ,  $a_+$ , and  $a_-$ . Another equation is obtained by averaging  $\nabla \cdot \mathbf{B} = 0$  across the film thickness, which relates  $\psi_+$ ,  $\psi_-$ ,  $a_+$ ,  $a_-$ ,  $b_+$ , and  $b_-$ . These five equations can be used to derive the following relations, where again expansions have been made to first order in  $\alpha d$ :

$$\alpha (\psi_+ - \psi_-) = 4\pi f \left[ \frac{a_+ + a_-}{1 + \beta d/2} \right], \quad (46a)$$

$$iq_y (\psi_+ + \psi_-) = 4\pi f \frac{q_y^2 d}{2\beta} \left[ \frac{b_+ + b_-}{1 + \beta d/2} \right]. \quad (46b)$$

Substituting these two relations into the averaged equations of motion given by Eqs. (40) and (41), equations of motion are obtained which are independent of  $\alpha$ :

$$i\frac{\omega}{\gamma}(a_+ + a_-) + \left[ H_0 + \frac{2A}{M_s}q_{\parallel}^2 + \frac{4\pi M_s f q_y^2 d/2}{\beta(1+\beta d/2)} \right] \times (b_+ + b_-) = 0, \quad (47)$$

$$\left[ H_0 + \frac{2A}{M_s}q_{\parallel}^2 - \frac{2k_s}{M_s d} + \frac{4\pi M_s f}{1+\beta d/2} \right] (a_+ + a_-) - i\frac{\omega}{\gamma}(b_+ + b_-) = 0. \quad (48)$$

The frequency of the long-wavelength mode, where  $\beta d = q_{\parallel} d \ll 1$ , is then given by

$$\frac{\omega}{\gamma} = \left[ \left[ H_0 + \frac{2A}{M_s}q_{\parallel}^2 - \frac{2k_s}{M_s d} + 4\pi M_s f (1 - \frac{1}{2}q_{\parallel} d) \right] \times \left[ H_0 + \frac{2A}{M_s}q_{\parallel}^2 + 2\pi M_s f q_{\parallel} d \sin^2 \phi \right] \right]^{1/2}. \quad (49)$$

Here the angle  $\phi$  is defined between  $\mathbf{q}_{\parallel}$  and the  $z$  axis. Equation (49) has several interesting features. First, note that the surface anisotropy has been averaged over the film and appears as an effective bulk anisotropy. Thus the surface anisotropy, which acts to “pin” the surface spins, here functions as a simple shift in the surface mode frequency. Second, the energy depends on the propagation direction through dipolar interactions and is largest for propagation in the  $y$  direction, perpendicular to the applied field.

The most interesting feature of Eq. (49), however, is the behavior as  $q_{\parallel} d$  goes to zero. We set  $k_s$  to zero for simplicity. The directional dependence disappears, and one has

$$\omega = \gamma \left[ \left[ H_0 + \frac{2A}{M_s}q_{\parallel}^2 \right] \left[ H_0 + \frac{2A}{M_s}q_{\parallel}^2 + 4\pi f M_s \right] \right]^{1/2}. \quad (50)$$

Comparison of Eqs. (17) and (50) shows that the discrete and continuous models give the same dispersion law for long wavelengths. Note that this correspondence rests on the identification of  $f$  with the  $q_{\parallel} a = 0$  dipole sum  $\sigma$  [Eq. (43)].

Finally, it is interesting to compare Eq. (49) to the dispersion relation derived by Yafet, Kwo, and Gyorgy.<sup>9</sup> In order to agree with the expression given in Ref. 9, the terms  $f(1 - \frac{1}{2}q_{\parallel} d)$  and  $f q_{\parallel} d \sin^2 \phi$  must be replaced with  $(f - \frac{1}{2}q_{\parallel} d)$  and  $q_{\parallel} d \sin^2 \phi$ . This difference comes from the dependence of the dipole sums on  $q_{\parallel}$  and shows the inadequacy of the simple approximation of Eqs. (42) and (43). But since these expressions are only valid in the long-wavelength region anyway, the error involved in this approximation is quite small and the continuum approximation presented here is very good.

#### IV. DISCUSSION

The frequency of Eq. (50) is shown in Fig. 1 as a function of  $q_{\parallel}$ . Here the value  $A/2\pi M_s^2 = 2.31 \times 10^{-13}$  is used. This value is chosen appropriate to bulk Co and is used only to illustrate certain general features, although

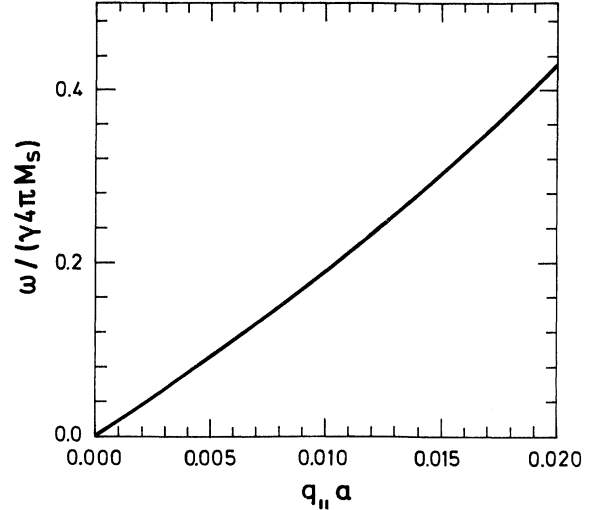


FIG. 1. Approximate dispersion relation for spin waves on the two-dimensional array.  $H_0$  is zero, and  $A/2\pi M_s^2 = 2.31 \times 10^{-13}$ . All anisotropies are set to zero. For large wave vectors, the behavior is quadratic in  $q_{\parallel}$ , while for small wave vectors the frequency is linear in  $q_{\parallel}$ .

it is noted that this ratio could change significantly for a monolayer.  $H_0$  is set to zero, and the frequencies and wave vectors are in unitless reduced forms. For small  $q_{\parallel}$  the frequency increases linearly with  $q_{\parallel}$ , while for larger  $q_{\parallel}$  the frequency is quadratic in  $q_{\parallel}$ . For finite  $d$ , the frequency increases as  $\sqrt{q_{\parallel}}$  at long wavelengths. As shown in Ref. 9, this behavior allows one to calculate the temperature dependence of the saturation magnetization without the problem of singularities in the two-dimensional sum over states.

While Eqs. (50) and (17) agree in the limit of  $q_{\parallel} \sim 0$ , it is useful to know how well they agree for larger  $q_{\parallel}$ . Another important question is how well the approximation works for films which are thicker than one monolayer. First, the question of larger  $q_{\parallel}$  is addressed.

The validity of Eqs. (50) and (17) for larger  $q_{\parallel}$  can be determined from Table I. In Table I the frequency of the spin-wave mode calculated from the full discrete model [Eq. (11)] is shown for two directions of propagation:  $\phi = 90^\circ$  ( $\mathbf{q}_{\parallel} = \hat{y}q_y$ ) and  $\phi = 0$  ( $\mathbf{q}_{\parallel} = \hat{z}q_z$ ). The method used to evaluate the sums of Eqs. (12) and (13) is described in Ref. 12. For comparison, the frequency as calculated from the continuous model [Eq. (50)] is also shown. The parameters are the same as in Fig. 1, and in order to facilitate comparison with the continuous model,  $\Sigma_{\text{ex}} = (2A/M_s)q_{\parallel}^2$ , which is valid in the long-wavelength limit.

For larger  $q_{\parallel}$  the two models are in good agreement since the exchange energy dominates. As  $q_{\parallel}$  goes to zero, however, the frequencies calculated from the discrete and continuous models diverge for propagation in the  $y$  direction, perpendicular to the saturation magnetization. The frequency dependence on propagation direction is well known from studies of magnetostatic spin waves in thick films and is due to the dipolar fields generated by the fluc-

TABLE I. Comparison of the frequencies calculated from the discrete model [Eq. (11)] and the continuum model [Eq. (50)]. The frequencies of the spin-wave mode are shown for various wavelengths for propagation parallel to the  $z$  axis ( $\phi=0$ ) and perpendicular to the  $z$  axis ( $\phi=\pi/2$ ). The applied field  $H_0$  is set to zero,  $A/2\pi M_s^2=2.31 \times 10^{-13}$ , and there are no anisotropies. The last column is the difference between the frequencies for the two propagation directions in GHz units using parameters appropriate to bulk Co:  $4\pi M_s=17.6$  kG,  $A=2.85 \times 10^{-6}$  erg/cm, and  $\gamma=1.936 \times 10^{-6}$  Hz/Oe.

$q_{\parallel}a$	Eq. (50)		Eq. (11)	$\hbar\omega(\phi=90^\circ) - \hbar\omega(\phi=0)$ ( $10^{-6}$ eV)
	$\frac{\omega}{4\pi M_s \gamma}$	$\frac{\omega}{4\pi M_s \gamma}$	$\frac{\omega}{4\pi M_s \gamma}(\phi=90^\circ)$	
$10^{-1}$	6.032 71	6.032 71	6.045 60	2.889
$10^{-2}$	0.185 38	0.185 38	0.189 34	0.888
$10^{-3}$	0.017 66	0.017 66	0.021 14	0.780
$10^{-4}$	0.001 77	0.001 77	0.004 07	0.515
$10^{-5}$	0.000 18	0.000 18	0.001 17	0.222

tuating in-plane component of the magnetization. The in-plane dipolar fields alternate in sign every half wavelength and oppose each other for propagation directions away from the saturation magnetization.<sup>16</sup> The extreme case is for propagation perpendicular to the saturation magnetization. Note, however, that the difference in energies for propagation perpendicular and parallel to the saturation magnetization is quite small since the precession of the magnetization is mostly out of plane.

The last column in Table I shows the difference in energies of a spin wave propagating in the  $\phi=0^\circ$  and  $90^\circ$  directions for a thin film with Co parameters ( $4\pi M_s=17.6$  kG,  $A=2.85 \times 10^{-6}$  erg/cm, and  $\gamma/2\pi=3.078 \times 10^6$  Hz/Oe). There is no applied field. These energy differences are very small: In terms of experimentally measurable quantities, the energy difference is too small to be measured by light-scattering methods and also too small to have a significant effect on the thermodynamics. To a good approximation, the spin-wave energies can be thought of as isotropic with respect to propagation direction.

Next, the effects of film thickness are examined. A good description of the spin-wave modes in thin films can be obtained with a continuum theory that includes both exchange and dipolar effects in a semiclassical electromagnetic boundary value problem. Details of this model are given by several authors and are not repeated here. Instead, the results of such a calculation are presented for various film thicknesses in order to study the evolution of the thin-film surface mode as the film thickness approaches that of a monolayer.

In Fig. 2 the frequency of the surface mode is plotted as a function of the in-plane propagation angle  $\phi$ , defined as the angle between  $q_{\parallel}$  and the  $z$  axis. The three curves are calculated according to the semiclassical electromagnetic boundary value problem described above and represent the surface modes for three different thickness films: one, five, and ten monolayers (ML). The reduced wave vector  $q_{\parallel}a$  is held at  $10^{-2}$  and  $H_0=0$ . For ease of comparison, the demagnetizing factor is set to 0.5392 for all cases shown. In reality, however, this factor increases with increasing thickness, which in turn raises the frequency of the spin-wave mode. The 1-ML case is the isotropic spin-wave mode calculated from Eq. (50). As the

thickness decreases, the directional dependence of the frequency also decreases, and the frequency approaches that of Eq. (50). It is noted that the small thickness approximation of Eq. (49) agrees with the results from the "full" boundary-condition problem to within a few percent for the thicknesses shown here.

The energy of the spin wave is smallest for propagation in the  $\phi=0$  direction and largest for propagation in the  $\phi=90^\circ$  direction. As described above, this difference is very small for the monolayer film. The precision of the magnetization becomes less elliptic as the film thickness increases, however, and the directional dependence of the frequency increases correspondingly.

Because dipolar interactions modify the long-wavelength behavior of  $\omega(q_{\parallel})$ , spin-wave theory can be used to calculate the magnetization as a function of tem-

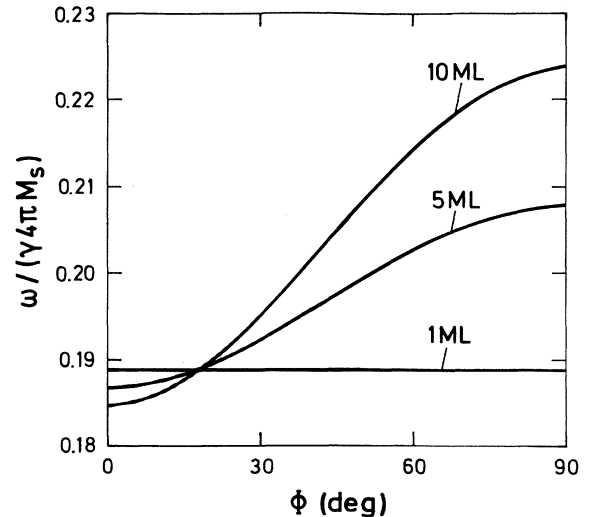


FIG. 2. Frequency as a function of propagation direction  $\phi$  for films of various thicknesses. The solid lines are calculated from a semiclassical continuum model, and the dashed line is calculated for the limiting case of a monolayer. The dependence of the frequency on  $\phi$  increases with increasing thickness and is due to the higher-order terms neglected in the approximations leading to Eq. (52).

perature without having to deal with divergences at  $q_{\parallel}=0$ . This can be seen by examining the expression for the magnetization as a function of temperature,  $M(T)$ , defined by

$$\frac{M(T)}{M(0)} = 1 - \frac{\Delta M(T)}{M(0)}. \quad (51)$$

For low temperatures,  $\Delta M(T)$  is proportional to a two-dimensional sum over spin-wave states.<sup>9</sup> The integration is taken over the first Brillouin zone and in the limit  $q_{\parallel} \rightarrow 0$ , the integrand does not diverge.

A dispersion relation of the form  $\omega = Dq_{\parallel}^2 + H_0$  without dipole interactions will result in a temperature dependence for the magnetization that is much too sensitive to the magnitude of  $H_0$  since the Curie temperature in this case would become vanishingly small as the field goes to zero. This is because  $H_0$  creates a "gap" in the spin-wave dispersion for  $q_{\parallel} \sim 0$ , which in turn makes it more difficult to thermally excite spin waves and essentially stabilizes the system. The same effect can also be achieved by bulk or surface anisotropies. The effect of the dipolar fields, however, is to modify how  $\omega$  goes to zero as  $q_{\parallel}$  goes to zero, which in turn determines the sensitivity of  $M(T)$  to external applied fields and anisotropies.

## V. CONCLUSIONS

The spin-wave dispersion for a magnetic monolayer has been derived from a microscopic argument and shown to have a simple form that can also be obtained

from a simple macroscopic model. The formalism was shown to be applicable to ultrathin films consisting of two or three atomic layers for the low-energy spin waves, since these spin waves are governed by the magnetic properties of the film averaged over several lattice sites.

The most important property of these spin-wave modes is their dependence on wave vector at long wavelengths due to the presence of long-range dipolar interactions. This means that the thermodynamic properties of monolayers and ultrathin films are strongly affected by anisotropies and external fields, since both create gaps in the spin-wave dispersion curves at long wavelengths. Furthermore, lattice distortions which reduce the symmetry of the system can also contribute slightly to the magnetic anisotropies through dipolar interactions. It is thus necessary to distinguish between the effects of dipolar interactions and magnetocrystalline anisotropies when interpreting experimental data from low-dimensional magnetic systems.

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