

## Dipole-exchange modes in thin ferromagnetic films with strong out-of-plane anisotropies

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Dipole-exchange modes are calculated for ultrathin ferromagnetic films with strong out-of-plane surface anisotropies large enough to force the magnetization out of plane. The direction of the magnetization is controlled by an in-plane static applied field. It is shown that dipolar-dominated spin-wave modes go soft for applied field strengths near where the magnetization begins to turn out of plane. For ultrathin films, however, the surface mode may contain a significant exchange energy that results in a nonzero minimum frequency at the applied field where the magnetization begins to turn out of plane. A potentially useful finding is a sensitive dependence of this minimum frequency on the exchange constant of the material.

### INTRODUCTION

The high-quality ultrathin magnetic layers and magnetic superlattices currently available provide excellent opportunities for the study of surface and interface magnetism. A particularly fascinating phenomenon, with many potential practical applications, is a spontaneous out-of-plane magnetization in thin ferromagnetic films and multilayers believed to originate from strong surface or interface anisotropies.<sup>1-6</sup> This has been observed in single thin films of Fe on Cu and Co on Au when the film thicknesses are less than 10 Å and is strongly dependent on the substrate material and interface quality.<sup>1</sup> Superlattices of Co/Ag and Co/Pd also demonstrate this behavior when the thickness of the nonmagnetic layers is larger than that of the magnetic layers.<sup>7</sup>

Magnetic anisotropy can originate from spin-orbit coupling (which, in turn, is also affected by lattice strains and stresses) as well as dipolar interactions caused by, for example, surface roughness or inhomogeneities in the bulk material.<sup>8-11</sup> The reduced symmetry of the surface region of thin films also gives rise to the possibility of surface anisotropies that differ from the bulk even in perfect crystals.<sup>12</sup> The microscopic origin of observed surface anisotropies is not, however, completely understood at present.

Exchange interactions and anisotropy fields at the surface of the crystal can also strongly influence spin-wave energies in wavelength regions where both dipolar and exchange interactions in the spin system are important.<sup>13</sup> This wavelength region is in the  $10^{-5}$ – $10^{-6}$  cm range, and these excitations are readily studied by Brillouin light-scattering techniques, especially the surface waves which are strongly localized to the surfaces.<sup>14</sup>

For magnetic films with out-of-plane anisotropies strong enough to overcome the demagnetizing field and yield an out-of-plane magnetization, it is possible to vary the magnitude of the out-of-plane component of the magnetization by applying an in-plane magnetic field. There then exists a critical switching field below which the magnetization has an out-of-plane orientation and above which the magnetization lies in plane.<sup>1</sup> Also, in direc-

tions normal to the film plane and with thicknesses on the order of only a few atomic layers, one expects all the moments to be parallel across the film thickness since the energy costs of forming a small domain wall in this direction are very high.<sup>15</sup> The equilibrium orientation of the magnetization is then determined primarily by long-range dipolar interactions, and the pure dipolar spin-wave modes go soft near the critical switching field.<sup>1</sup>

In very thin films, however, the component of the spin wave's wave vector normal to the film can be relatively large, which means a significant exchange energy is still carried by the wave. This energy will be on the order of  $Aq^2$ , where  $A$  is the exchange constant of the material and  $q$  is the wave vector of the excitation. At the critical switching field, it is possible that this exchange energy will result in a nonzero frequency for the spin wave. The purpose of this paper is to calculate the conditions under which this may occur and the expected dispersion curves for the dipole-exchange spin-wave modes near this switching field. This may be a particularly interesting region for experimental study also, since here microscopic surface conditions can have a significant effect on observed frequencies through the exchange interactions governing the mode.

In what follows the theory behind these calculations is described and followed by a discussion of example calculations for Fe and Co thin films. Finally, it is observed that near switching field, the spin-wave excitations are especially sensitive to surface exchange fields, and a simple model is examined which provides a description of these surface exchange fields.

### THEORY

The geometry is defined in Fig. 1. The unprimed coordinate system is that of the film with the  $x$  axis normal to the film plane. An applied field  $H_0$  is taken along the  $z$  axis. The saturation magnetization per unit volume,  $M_s$ , lies in the  $xz$  plane, and a primed coordinate system is defined such that  $M_s$  lies along the  $z'$  axis. The angle  $\theta$  is defined between the  $z'$  and  $z$  axes.

In order to calculate the static orientation of the mag-

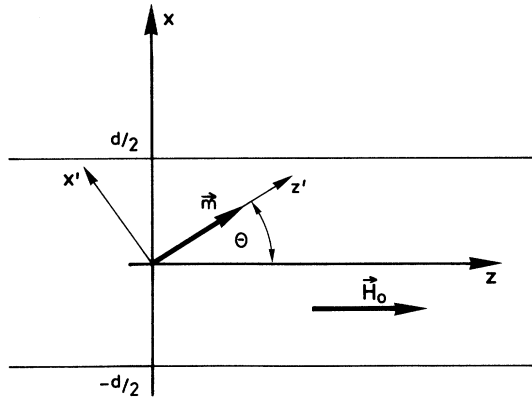


FIG. 1. Geometry: The unprimed coordinate system is for the film with the  $x$  axis normal to the film plane. An applied field  $H_0$  is taken along the  $z$  axis. The saturation magnetization  $M_s$  lies in the  $xz$  plane, and a primed coordinate system is defined such that  $M_s$  lies along the  $z'$  axis. The angle  $\theta$  is defined between the  $z'$  and  $z$  axes.

netization as a function of applied field strength, a free energy is first defined. Using  $K_1$  as a first-order volume anisotropy energy per unit volume,  $K_2$  as a second-order anisotropy energy per unit volume, and  $k_s$  as a surface anisotropy energy per unit area, the free energy  $F$  per unit area  $\sigma$  reads

$$F/\sigma = \int dx [-H_0 M_s \cos(\theta) + 2\pi M_s^2 \sin^2(\theta) - K_1 \cos^2(\theta) - K_2 \cos^4(\theta)] + 2k_s \sin^2(\theta), \quad (1)$$

where the second term in the integrand is due to demagnetizing fields. The last term is the surface energy due to surface anisotropies from both surfaces of the film. If the film thickness is much smaller than the width of a domain wall, the magnetization across the film can be treated as approximately uniform. In this case  $\theta$  is independent of  $x$ , and for a film of thickness  $d$  the energy per unit volume is

$$F/V = -H_0 M_s \cos(\theta) + 2\pi M_s^2 \sin^2(\theta) - K_1 \cos^2(\theta) - K_2 \cos^4(\theta) + 2k_s \sin^2(\theta)/d. \quad (2)$$

In this paper the convention  $k_s < 0$  means an out-of-plane orientation is preferred and  $k_s > 0$  means an in-plane orientation is preferred.

The minimum of  $F/V$  with respect to  $\theta$  gives the equilibrium direction of the magnetization defined by  $\theta$ . One solution is always that  $\theta=0$ . In the case where  $K_2$  is zero, a simple expression for the other possible solution is obtained for  $(-2k_s/d) > (2\pi M_s^2 + K_1)$ :

$$\cos\theta = H_0 M_s / (-4k_s/d - 4\pi M_s^2 - 2K_1). \quad (3)$$

Clearly, as  $d$  becomes small, this value of  $\theta$  approaches  $\pi/2$ .

An example plot of  $F/V$  is given in Fig. 2 as a function of applied field. The values used are appropriate for Co

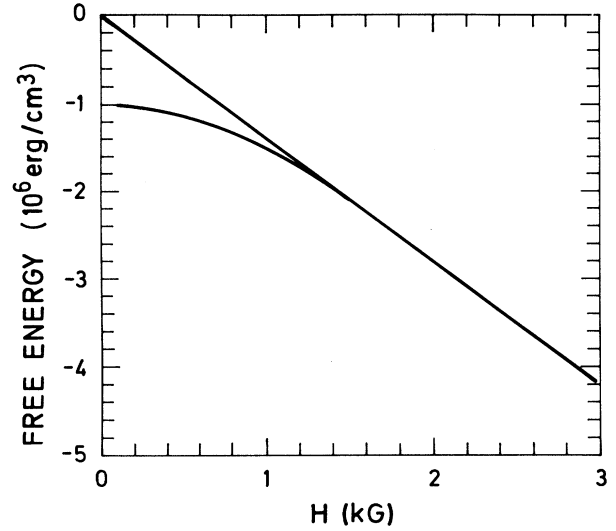


FIG. 2. Free energies per unit volume,  $F/V$ , for in-plane and out-of-plane orientations of  $M_s$ . Here the parameters are appropriate to Co with  $K_1=K_2=0$ ,  $4\pi M_s=17.6$  kG,  $d=6$  Å, and  $k_s=0.4$  erg/cm<sup>2</sup>. The lower curve is  $F/V$  using  $\theta$  defined by Eq. (3), and the upper curve is  $F/V$  for  $\theta=0$ .

( $4\pi M_s=17.6$  kG) with  $d=6$  Å. This thickness corresponds to roughly three atomic layers, and the distinction between surface and volume anisotropies is not meaningful. For simplicity,  $K_1=K_2=0$ , and the value  $k_s=-0.4$  erg/cm<sup>2</sup> is chosen in order to put the critical switching field near 1.4 kG. The lower curve in Fig. 2 is  $F/V$  using  $\theta$  defined by Eq. (3), and the upper curve is  $F/V$  for  $\theta=0$ . Thus Eq. (3) defines a lower-energy state. Near the "critical" field, the difference between the two possible states is very small, and one should question the stability of these orientations to fluctuations in the spin system. Furthermore,  $+\theta$  and  $-\theta$  out-of-plane orientations are equally likely. The net demagnetizing energy can then be reduced with the formation of a domain structure in the  $yz$  plane. The following theory, however, assumes that  $\theta$  is everywhere uniform and is thus only applicable for very small  $\theta$  or large domains.

Next, the dynamics of the system are studied. The approach here is the same as that used several times before in studies of the dipole-exchange region.<sup>13,16-18</sup> It is important to note, however, that in defining effective fields for the equations of motion, the surface energy term becomes an effective bulk anisotropy field. The validity of this assumption can be tested by solving the dipole-exchange spin-wave problem, in one case with these effective bulk fields and no surface anisotropy terms, and in another case without effective bulk fields but with surface anisotropy terms. This has been done by Rado for the case of in-plane orientation of the magnetization, and good agreement was found for the ferromagnetic resonance mode in very thin films.<sup>19</sup> This has also been done for the present case, and again good agreement between the two representations is found for the surface mode with applied fields above the critical switching field.

Since the surface anisotropy fields are described by effective bulk anisotropy fields, changes in spin-wave frequencies from different surface anisotropies at the top and bottom surfaces of the film cannot be considered in this theory. This is a significant limitation of the present theory, as compared to in-plane theories, and has important consequences for the boundary conditions as will be discussed below.

The magnetic torque equations are written in the mag-

netization frame as  $(\partial/\partial t)\mathbf{m}' = \gamma\mathbf{m}' \times \mathbf{H}'$ , where  $\mathbf{m}'$  is the magnetization in the primed frame and  $\mathbf{H}'$  is the effective field acting in the primed frame. The nonretarded ( $\nabla \times \mathbf{h}' = 0$ ) case is considered where a magnetostatic potential  $\phi$  can be defined such that  $\mathbf{h}' = -\nabla\phi$ . The resulting electromagnetic equations from  $\nabla \cdot \mathbf{B}' = 0$  are also written in the magnetization's frame. This gives a system of three equations, which in matrix form are

$$\begin{bmatrix} i\omega/\gamma & H_0 \cos\theta - 4\pi M_s \sin^2\theta + R_y + \frac{2A}{M_s} q^2 & iM_s q_y \\ -(H_0 \cos\theta - 4\pi M_s \sin^2\theta + R_x + \frac{2A}{M_s} q^2) & i\omega/\gamma & -iM_s q'_x \\ 4\pi i q'_x & 4\pi i q_y & q^2 \end{bmatrix} \begin{bmatrix} m'_x \\ m'_y \\ \phi \end{bmatrix} = 0. \quad (4)$$

Here  $A$  is the exchange constant defined by  $2JS^2/a$  for a bcc crystal with lattice constant  $a$ . The exchange integral is  $J$  and the spin number is  $S$ . The anisotropy fields are contained in  $R_x$  and  $R_y$ :

$$R_x = -\frac{2}{M_s} \left[ \frac{2k_s}{d} + K_1 \right] (2 \sin^2\theta - 1) + 4 \frac{K_2}{M_s} (1 - 4 \sin^2\theta) \cos^2\theta, \quad (5)$$

$$R_y = \frac{2}{M_s} \left[ -\frac{2k_s \sin^2\theta}{d} + K_1 \cos^2\theta \right] + 4 \frac{K_2}{M_s} \cos^4\theta. \quad (6)$$

For propagation perpendicular to  $H_0$ , solutions for  $m'_x$ ,  $m'_y$ , and  $\phi$  are assumed to have the form

$$e^{iq_x x} e^{i(q_y y - \omega t)}. \quad (7)$$

Outside the film the magnetization is zero and the potential is assumed to decay exponentially away from the surface according to  $\exp(-\alpha x)$  for  $x > 0$  and  $\exp(\alpha x)$  for  $x < 0$ . The potential outside the material obeys Laplace's equation, which in turn requires  $\alpha = |q_y|$ .

Finally, it is noted that  $q^2$  and  $q_y$  must be the same in both the primed and unprimed systems, while the  $x$  component satisfies, for  $q_z = 0$ ,

$$q'_x = q_x \cos\theta, \quad (8)$$

$$q'_z = q_x \sin\theta. \quad (9)$$

The complete solution to the thin-film problem requires the application of appropriate boundary conditions. These must include the usual conditions on the continuity of the normal  $\mathbf{B}$  and tangential  $\mathbf{h}$  fields. Examination of Eq. (4), however, reveals that the determinant of this matrix will vanish for six different  $q_x$ . The general solution to the film problem then requires the superposition of six different partial waves of the form

given in (7), plus two waves in the regions outside the film geometry. Thus four more boundary conditions are required.

The Rado-Weertman boundary conditions provide the four extra conditions.<sup>11</sup> These are of the form

$$A \sum_{j=1}^6 \frac{\partial}{\partial x} m'_{xj} = 0, \quad (10)$$

$$A \sum_{j=1}^6 \frac{\partial}{\partial x} m'_{yj} = 0. \quad (11)$$

The subscript  $j$  indicates the partial wave associated with the wave number  $q_{xj}$ . Note that these are conditions on the primed magnetization, but are to be evaluated at the unprimed coordinate  $x = \pm d/2$ .

Although surface anisotropy is assumed to result in an out-of-plane magnetization, no anisotropy terms can be included in the boundary conditions given by Eqs. (10) and (11). This is because the assumption that the magnetization is uniform across the sample requires the static torques on both bulk and surface spins to vanish for the same angle  $\theta$ . The appearance of surface anisotropy terms in Eqs. (10) and (11) would imply that surface torques exist which are different from the bulk, thus leading to a nonuniform magnetization across the sample as the magnetization leaves the direction of the static field  $H_0$ . Hence the assumption of a uniform magnetization does not allow the inclusion of surface pinning terms due to surface anisotropies. Also, the question of spin canting cannot be addressed within this assumption. For very thin films near the switching point, however, it is believed that only small canting angles are likely, and these would have a negligible effect on the spin-wave frequencies.<sup>19</sup>

Finally, the boundary conditions on the tangential  $\mathbf{h}$  and normal  $\mathbf{B}$  fields are easily formulated in terms of the magnetic potential  $\phi_j$ :

$$\sum_{j=1}^6 [4\pi m'_{xj} \cos\theta - (iq_{xj} \pm q_y) \phi_j] = 0. \quad (12)$$

The plus sign is for the surface at  $x = d/2$ , and the minus sign is for  $x = -d/2$ .

The frequencies of the allowed modes are determined numerically by solving the equations of motion [Eq. (4)] for the wave vectors  $q_{xj}$  for a given frequency  $\omega$ . Then, writing  $m'_{xj} = a_j \phi_j$  and  $m_{yj} = b_j \phi_j$ , where the subscripts indicate the associated  $q_{xj}$  value, the equations of motion are solved for  $a_j$  and  $b_j$  in terms of  $\phi_j$ . The boundary conditions [Eqs. (10)–(12)] have the final form

$$\sum_{j=1}^6 \frac{(H_{12} q_{xj} \cos \theta - i q_y \omega / \gamma)}{(H_{12} H_{21} - \omega^2 / \gamma^2)} q_{xj} \phi_j = 0, \quad (13)$$

$$\sum_{j=1}^6 \frac{(H_{21} q_y + i q_{xj} \cos \theta \omega / \gamma)}{(H_{12} H_{21} - \omega^2 / \gamma^2)} q_{xj} \phi_j = 0, \quad (14)$$

$$\sum_{j=1}^6 \left[ 4\pi M_s \frac{(H_{12} q_{xj} \cos \theta - i q_y \omega / \gamma)}{(H_{12} H_{21} - \omega^2 / \gamma^2)} \cos \theta - (i q_{xj} \pm q_y) \right] \phi_j = 0. \quad (15)$$

Here

$$H_{12} = H_0 \cos \theta - 4\pi M_s \sin^2 \theta + R_y + \frac{2A}{M_s} q^2, \quad (16)$$

$$H_{21} = H_0 \cos \theta - 4\pi M_s \sin^2 \theta + R_x + \frac{2A}{M_s} q^2. \quad (17)$$

With a fixed  $q_y$  and a guess for  $\omega$ , the  $q_{xj}$  are found and the boundary conditions are constructed. An iterative process is then used in order to find an  $\omega$  that satisfies the boundary conditions according to a numerical error condition.

Before proceeding to the results of this numerical procedure, it is useful to examine the determinant of the equations of motion (4) at the field where the magnetization begins to turn out of plane. For  $q_z = 0$  and  $K_2 = 0$ , one finds

$$\begin{aligned} \frac{\omega^2}{\gamma^2} = & \left[ H_0 + \frac{2K_1}{M_s} + 2 \frac{A}{M_s} q^2 \right] \\ & \times \left[ H_0 + \frac{2K_1}{M_s} + \frac{4k_s}{M_s d} + 4\pi M_s + 2 \frac{A}{M_s} q^2 \right] \\ & + \frac{16\pi k_s (q_y / q)^2}{d}. \end{aligned} \quad (18)$$

In the case where  $q_x \ll q_y$ , which describes the dipolar-dominated mode,  $\omega^2$  will be positive only if

$$Aq^2 \approx Aq_y^2 > 2\pi M_s^2. \quad (19)$$

This states simply that the ‘‘dipolar’’ mode will become soft at the switching field unless the exchange energy contained in the mode is greater than the demagnetizing energy.

## DISCUSSION

It is illuminating to trace the evolution of the spin-wave spectrum as the exchange constant  $A$  is increased

from zero. With  $A = 0$ , the spin-wave modes are completely governed by dipolar interactions. As is well known, a dipolar surface wave does not exist in a perpendicularly magnetized film.<sup>13</sup> When the magnetization lies in-plane, however, a surface mode can exist. The evolution of this dipolar surface mode as the magnetization swings out of plane in films with strong out-of-plane surface anisotropies can be studied as a limited case of the above formalism.<sup>20</sup> Doing so, one finds that a dipolar mode does exist which is strongly localized to a surface of the film for some orientation angles  $\theta \neq 0$ , but is bulklike near  $\theta = 0$ . In the limit of  $q_x = 0$ , this dipolar mode coincides with the ferromagnetic resonances mode of the film.

The surface mode with and without exchange is shown as a function of applied field in Fig. 3 for a 6-Å-thick Co film (using  $4\pi M_s = 17.6$  kG) with a  $k_s$  value of  $-0.4$  erg/cm<sup>2</sup> and  $q_y = 17.3 \times 10^5$  cm<sup>-1</sup>. Here  $\gamma = g\gamma_0$ , where  $\gamma_0 = 8.79 \times 10^6$  Hz/Oe. The  $g$  factor for Co is taken as 2.2. The dipole-exchange surface mode, as calculated from the full theory of the previous section, is represented by the solid line. The exchange constant is  $A = 2.85 \times 10^{-6}$  erg/cm, and the dashed line is the corresponding mode for  $A = 0$ . For this thickness the bulk modes have extremely large exchange energies and thus are not visible on this scale.

Note that the  $A = 0$  mode goes soft near the critical field, and there exists a range of fields between about 1.41 and 1.53 kG where there is no surface mode. This is due to the influence of the last term in Eq. (18), which depends both on the perpendicular anisotropy and the propagation wave vector of the mode. Note that when

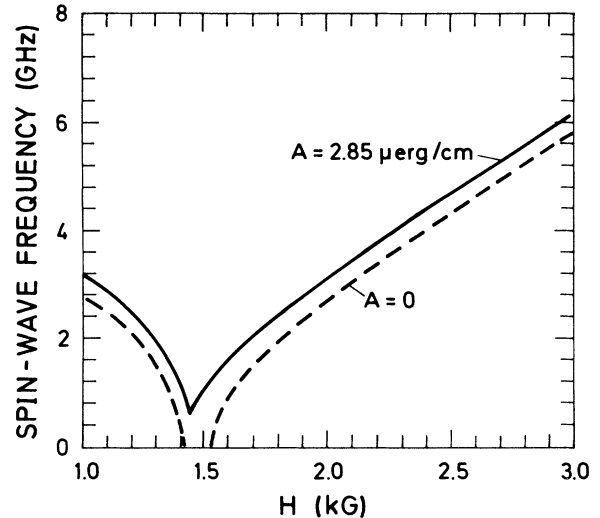


FIG. 3. Frequency as a function of applied field for the surface mode with and without exchange on a 6-Å-thick Co film is shown with a  $k_s$  value of  $-0.4$  erg/cm<sup>2</sup> at  $q_y = 17.3 \times 10^5$  cm<sup>-1</sup>. The solid line is for  $A = 2.85$  μerg/cm, and the dashed line is the same mode for  $A = 0$ . For this thickness the bulk modes have extremely large energies and are dominated by the exchange interaction. The important feature is that the surface mode has a sharp minimum at the critical field  $H_c = 1.43$  kG, but does not vanish as the  $A = 0$  mode does.

$q_y=0$ , the  $A=0$  mode goes soft exactly at the critical field. The most interesting feature, however, is that the  $A\neq 0$  surface mode has a sharp minimum at the critical field, but does not vanish as the  $A=0$  mode does. Then, according to Eq. (9), the exchange energy contained in the mode is larger than the demagnetizing energy.

The behavior of the surface mode for small  $H_0$  is also interesting. In Fig. 4 the magnetic potential  $\phi$  for  $A=0$  is profiled as a function of position within the film for the 6-Å-thick film of Fig. 3. For  $H_0=1$  G, where the magnetization is essentially perpendicular to the film, the curvature of  $\phi$  within the film indicates that this mode is bulklike. This is in agreement with previous results for perpendicularly magnetized ferromagnetic slabs where a surface mode does not exist in the dipolar limit. The larger fields of 10 and 100 G give the magnetization a significant in-plane component. Here the profile of  $\phi$  shows that the amplitude of the wave becomes larger at one surface and decays with distance into the film, which indicates that the mode is more surfacelike.

For applied field strengths above the critical switching field  $H_c$ , the surface mode is not strongly affected by exchange. Near  $H_c$  the surface mode is strongly influenced by exchange, as described before. This is also true for small applied field strengths where the magnetization has a large component perpendicular to the film plane since

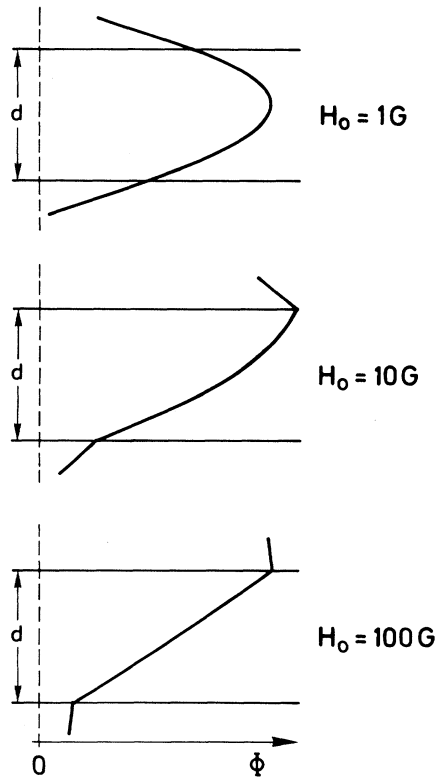


FIG. 4. Magnetic potential for the  $A=0$  mode for the 6-Å-thick Co film of Fig. 3 is profiled for different fields  $H_0$ . For small  $H_0$  the mode is clearly bulklike as seen by its curvature, while for larger fields the mode becomes strongly localized to a surface of the film.

then the mode is more bulklike with a larger variation in amplitude across the film thickness.

To see what happens in this region, first consider the case of perpendicular magnetization and  $A\neq 0$ . In this case a surface-localized mode exists in frequency regions where the  $q_{xj}$  are complex.<sup>13</sup> A surface-localized mode then exists only as a superposition of partial waves which are bulklike but strongly damped. Furthermore, this surface mode does not exist for all thicknesses or parallel wave vectors  $q_y$ . To study this surface-localized state as the magnetization tilts out of plane, it is therefore useful to examine the behavior of the  $q_{xj}$  as functions of  $H_0$  and  $\omega$ .

The regions of complex and imaginary  $q_x$  are shown in Fig. 5 for different values of  $\omega$  and  $q_y$  for the 6-Å-thick film of Fig. 3 with  $H_0=370$  G and also with  $H_0=381$  G. In this plot there are three regions: In region I four of the  $q_x$  are complex and two are purely imaginary. In region II all of the  $q_x$  are unequal and pure imaginary. In region III all of the  $q_x$  are unequal and complex. In region IV all of the  $q_x$  are again unequal, with four pure real  $q_x$  and two pure imaginary  $q_x$ . The solid line is the

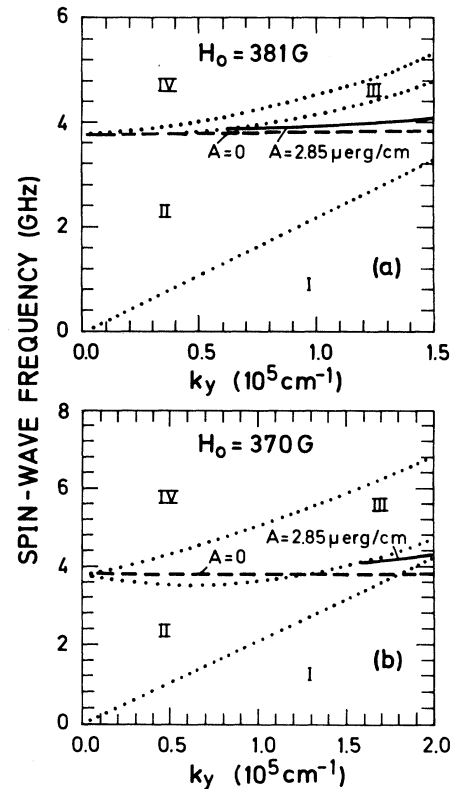


FIG. 5. Frequencies of the surface modes of Fig. 3 are shown as functions of  $q_y$  for two different applied fields. The dotted lines separate the different regions I–IV described in the text, the dashed line is the  $A=0$  mode, and the solid line is the  $A\neq 0$  surface mode. (a)  $H_0=381$  G and the  $A\neq 0$  mode merges into  $A=0$  mode in region II where all the  $q_{xi}$  are imaginary. (b)  $H_0=370$  G and the  $A\neq 0$  mode follows the border between regions II and III.

dipole-exchange surface mode, and the dashed line is the  $A=0$  dipolar surface mode.

For  $H_0=381$  G [Fig. 5(a)], the dipole-exchange mode merges into the  $A=0$  mode as  $q_y$  becomes small. When  $H_0$  is reduced to 370 [Fig. 5(b)], the dipole-exchange mode no longer joins the  $A=0$  mode, but instead follows the border separating regions II and III. It is interesting to note that an examination of the equations of motion shows that there are only four unique  $q_{xi}$  for points along this line. It is then not possible to satisfy the full set of dipole and exchange boundary conditions.

The behavior of the surface mode is now examined near  $H_c$ . The magnitude of the frequency near  $H_c$  depends on the magnitude of the exchange constant, the parallel component of the propagation wave vector  $q_y$ , the magnetization, and also the surface anisotropy. In Fig. 6 the frequency of the spin waves at the critical switching field is shown as a function of  $A$ . The lower curve is calculated by the theory presented above for the 6-Å-thick Co film of Fig. 4. Below approximately 2  $\mu\text{erg/cm}$ , the surface mode goes soft. As  $A$  increases, the frequency of the surface mode also increases.

The theory presented above assumes a uniform demagnetizing field of  $4\pi M_s$ . In thin films where out-of-plane switching is expected to occur, however, these are no longer valid assumptions.<sup>21</sup> As an approximation, one can write the average demagnetizing field for a slab geometry as  $4\pi d_m M_s$ , where  $d_m$  is a reduction factor for thin slabs that is found by numerically summing the dipole-dipole terms for the local field at a given lattice site due to all neighboring magnetic moments.<sup>21</sup>

The higher-frequency curve in Fig. 6 illustrates the

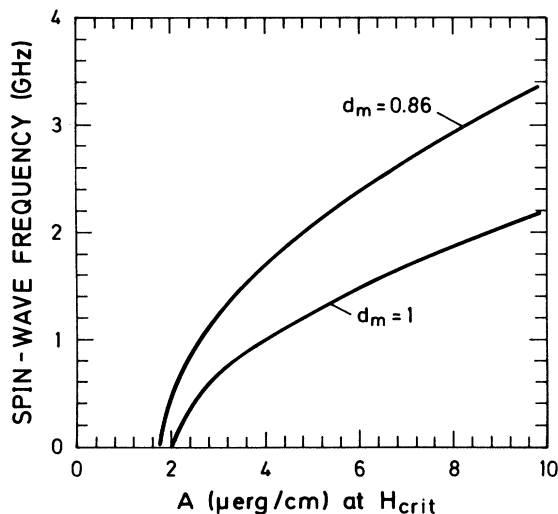


FIG. 6. Frequency of the surface mode at the critical switching field is shown as a function of the exchange constant for  $d=6$  Å,  $4\pi M_s=17.6$  kG, and  $g=2.2$  (Co values). The lower curve is calculated for a demagnetizing field of  $4\pi M_s$ , and the upper curve is calculated for a demagnetizing field of  $4\pi d_m M_s$ , where  $d_m$  is calculated from the numerical results of Ref. 22. Here,  $d_m=0.86$ .

effects of reduced demagnetizing fields on the spin-wave frequency at the critical switching field. Here  $d_m=0.86$  and is calculated from the results of Ref. 22. The effect is to increase the frequency of the surface mode and to lower the minimum  $A$  at which the surface mode goes soft. The critical switching field is also reduced since a reduced demagnetizing field makes it easier to turn the magnetization out of plane.

Finally, the question of how to properly represent the reduced symmetry of the surface through the boundary conditions is again examined. In the discussion preceding Eqs. (11)–(13), it was noted that in replacing the surface anisotropy fields by effective bulk fields, and assuming the surface magnetic moments to be aligned in the same direction as the bulk moments, the anisotropy-induced “pinning” at the surface could not be described in a consistent manner. On the other hand, it is possible that higher-order exchange terms in the boundary conditions, which would reflect the reduced symmetry of the surface through surface exchange fields that differ from the bulk exchange fields, may have a significant effect near the critical switching field.

Effective boundary conditions can be derived by requiring the surface magnetization to precess at the same frequency as the bulk magnetization. This approach is based on previous work by Sparks and has been applied by several authors.<sup>18,22</sup> The idea is that the equations of motion for the magnetization at the surface,

$$\frac{\partial}{\partial t} \mathbf{m} = \gamma \mathbf{m} \times \mathbf{H}_s, \quad (20)$$

should give the same frequency as the bulk equations of motion:

$$\frac{\partial}{\partial t} \mathbf{m} = \gamma \mathbf{m} \times \mathbf{H}_b. \quad (21)$$

Here  $\mathbf{m}$  is the magnetization (assumed to be a continuous function of position),  $\mathbf{H}_b$  is the effective field acting in the bulk,  $\mathbf{H}_s$  is the effective field acting at the surface, and  $\gamma$  is the gyromagnetic ratio. This requirement can be expressed by writing Eq. (20) as

$$\frac{\partial}{\partial t} \mathbf{m} = \gamma \mathbf{m} \times \mathbf{H}_b + \gamma \mathbf{m} \times (\mathbf{H}_s - \mathbf{H}_b) \quad (22)$$

and requiring

$$\mathbf{m} \times (\mathbf{H}_s - \mathbf{H}_b) = 0 \quad (23)$$

at the surfaces. Equation (23) then defines the effective exchange boundary conditions.

Equation (23) requires that the torques due to differences between the bulk and surface effective fields vanish. These surface torques can arise from differences between bulk and surface anisotropies, and also from differences between bulk and surface effective exchange fields. In order to calculate the surface exchange fields, the crystal structure of the surface is examined and an effective exchange field is derived in the mean-field limit. This is a well-known procedure, and only the results are presented below.<sup>13</sup>

In the bulk of a cubic crystal, this calculation gives the usual  $A \nabla^2 \mathbf{m}$  term for the exchange field. But the number

of nearest neighbors is less at the surface than in the bulk, and this results in the appearance of terms first order in  $a(\partial/\partial x)\mathbf{m}$  in the surface exchange fields. The difference between the surface and bulk exchange fields for (110) bcc crystal faces at the surfaces is given by

$$(\mathbf{H}_s - \mathbf{H}_b)_{\text{ex}} = A \left[ \mp \left[ \cos\theta \frac{\partial}{\partial x'} + \sin\theta \frac{\partial}{\partial z} \right] - \frac{a}{2\sqrt{2}} \left[ \cos^2\theta \frac{\partial^2}{\partial x'^2} + \frac{1}{2} \frac{\partial^2}{\partial y'^2} + \sin\theta \cos\theta \frac{\partial^2}{\partial y' \partial z'} + \sin^2\theta \frac{\partial^2}{\partial z'^2} \right] \right] \mathbf{m}'(x), \quad (24)$$

where the negative sign on the first term is chosen for the top surface of the material and the positive sign for the bottom surface of the material.

For bcc (100) surfaces, the difference between surface and bulk fields is given by

$$(\mathbf{H}_s - \mathbf{H}_b)_{\text{ex}} = A \left[ \mp \left[ \cos\theta \frac{\partial}{\partial x'} + \sin\theta \frac{\partial}{\partial z'} \right] - \frac{a}{4} \nabla^2 \right] \mathbf{m}'(x). \quad (25)$$

Again, the same  $\mp$  sign convention holds for the top and bottom surfaces. Both Eqs. (24) and (25) have been multiplied by the quantity

$$n_s = \frac{1}{a^2} \quad \text{and} \quad \frac{\sqrt{2}}{a^2}, \quad (26)$$

for (100) and (110) surfaces, respectively, where  $n_s$  is the number of surface spins per unit area for the bcc lattice, thus forming surface field densities.

The effect of these new terms on the boundary conditions is examined in Fig. 7. Here the solid line is without the second-order terms in the boundary conditions, the dashed curve is for (100) surfaces, and the dotted curve is for (110) surfaces. The parameters are for Co and are the same as those used in Fig. 3. For fields below and above the switching field, the second-order terms in both the (100) and (110) cases have a negligible effect compared to the first-order term. Near the switching field, however, these terms produce a noticeable difference in the spin-wave frequency. The different surfaces, however, produce nearly the same frequencies.

In conclusion, the strong dependence of the spin-wave frequency at the switching field on the exchange constant and out-of-plane anisotropy makes this a particularly interesting area for experimental study. Recent measurements by Dutcher *et al.*<sup>1</sup> suggest that the spin-wave frequencies in this region are accessible to light-scattering experiments, and the measurement of the switching field and the spin-wave dispersion curve near the switching field for these thin films may give unique information about the exchange and anisotropy constants. A major

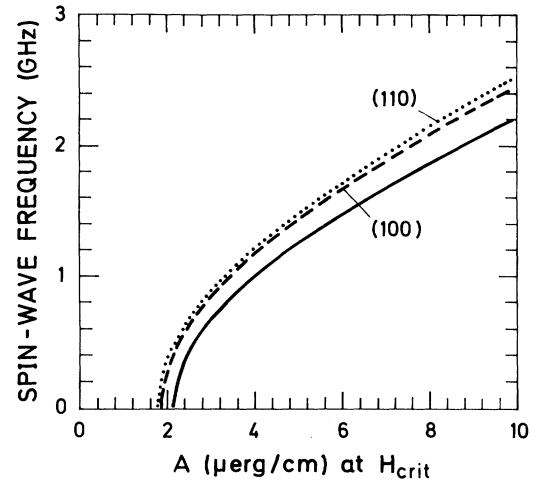


FIG. 7. Surface-mode frequency with second-order terms in the boundary conditions [Eqs. (24) and (25)]. The film is the same as that used in Fig. 3 (6-Å-thick Co film). The solid line is without the second-order terms in the boundary conditions, and the dashed (dotted) curve is for (100) [(110)] surfaces.

difficulty, however, is the possibility of domain formation in the film as the magnetization begins to turn out of plane. Inhomogeneities in the surface anisotropy, for example, could lead to variations in the equilibrium orientation of the magnetization that depend on position in the  $yz$  plane. A 1% change in  $k_s$  would lead to changes as large as 10% in the critical switching field for the examples presented here and could thus tend to "wash" out the minimum in the spin-wave frequency as determined by a light-scattering experiment. Measurements of this sort would thus require films with well-defined surfaces free of defects and roughness on length scales of several micrometers.

The preceding analysis also shows that the surface mode near the switching field is unusually sensitive to surface exchange fields. This in turn might be of interest especially in cases where polarization and diffusion effects across interfaces are thought to exist. A simple generalization of the boundary conditions of Eqs. (24) and (25), for example, would also allow the study of surface exchange constants that differ from the bulk values.

Finally, the collective spin-wave excitations of multilayers and superlattices with easy axes normal to the layer planes are the subject of a subsequent paper. Collective excitations on these kinds of structures are especially interesting since they often reveal unique properties not observed for excitations in the individual constituents.

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- <sup>1</sup>J. R. Dutcher, J. F. Cochran, I. Jacob, and W. F. Egelhoff, Jr., Phys. Rev. B **39**, 10430 (1989); J. R. Dutcher, B. Heinrich, J. F. Cochran, D. A. Steigerwald, and W. F. Egelhoff, Jr., J. Appl. Phys. **63**, 3464 (1988).
- <sup>2</sup>S. T. Purcell, B. Heinrich, and A. S. Arrott, J. Appl. Phys. **64**, 5337 (1988); F. J. A. den Broeder, D. Kuiper, A. P. van de Mosselaer, and W. Hoving, Phys. Rev. Lett. **60**, 2769 (1988).
- <sup>3</sup>J. J. Krebs, B. T. Jonker, and G. A. Prinz, J. Appl. Phys. **63**, 3467 (1988).
- <sup>4</sup>B. Heinrich, K. B. Urquhart, A. S. Arrott, J. F. Cochran, K. Myrtle, and S. T. Purcell, Phys. Rev. Lett. **59**, 1756 (1987); Z. S. Shan, D. J. Sellmyer, S. S. Jaswal, Y. J. Wang, and J. X. Shen, Phys. Rev. Lett. **63**, 449 (1989).
- <sup>5</sup>M. J. Pechan and I. K. Schuller, Phys. Rev. Lett. **59**, 142 (1987).
- <sup>6</sup>F. J. A. den Broeder, H. C. Donkersloot, H. J. G. Draaisma, and W. J. M. de Jonge, J. Appl. Phys. **61**, 4317 (1987).
- <sup>7</sup>J. V. Harzer, B. Hillebrands, and G. Güntherodt, J. Appl. Phys. (to be published).
- <sup>8</sup>T. Mizoguchi and G. S. Cargill III, J. Appl. Phys. **50**, 3570 (1979).
- <sup>9</sup>A. P. Malozemoff, Phys. Rev. B **35**, 3679 (1987).
- <sup>10</sup>H. J. G. Draaisma and W. J. M. de Jonge, J. Appl. Phys. **64**, 3610 (1988).
- <sup>11</sup>G. T. Rado and J. R. Weertman, J. Phys. Chem. Solids **11**, 315 (1959).
- <sup>12</sup>J. G. Gay and R. Richter, Phys. Rev. Lett. **56**, 2728 (1986).
- <sup>13</sup>R. E. De Wames and T. Wolfram, Prog. Surf. Sci. **2**, 233 (1972); R. E. De Wames and T. Wolfram, J. Appl. Phys. **41**, 987 (1970).
- <sup>14</sup>See, for example, the review articles by P. Grünberg and also M. H. Grimsditch in *Light Scattering in Solids V*, edited by M. Cardona and G. Güntherodt (Springer-Verlag, Berlin, 1989).
- <sup>15</sup>D. L. Mills, Phys. Rev. B **39**, 12306 (1989); G. T. Rado, J. Appl. Phys. **61**, 4262 (1987).
- <sup>16</sup>B. Hillebrands, Phys. Rev. B **37**, 9885 (1988); **41**, 530 (1990).
- <sup>17</sup>J. Dutcher, J. F. Cochran, B. Heinrich, and A. S. Arrott, J. Appl. Phys. **64**, 6095 (1988).
- <sup>18</sup>R. L. Stamps and R. E. Camley, Phys. Rev. B **35**, 1919 (1987).
- <sup>19</sup>G. T. Rado, Phys. Rev. B **40**, 407 (1989).
- <sup>20</sup>When  $A=0$  in the equations of motion, only two unique values for  $q_x$  can be found. The boundary-value problem is then formulated with only the electromagnetic conditions for the continuity of normal component of  $\mathbf{B}$  and the tangential component of  $\mathbf{h}$ .
- <sup>21</sup>H. J. G. Draaisma and W. J. M. de Jonge, J. Appl. Phys. **64**, 3610 (1988); J. R. Dutcher, B. Heinrich, J. F. Cochran, D. A. Steigerwald, and W. F. Egelhoff, Jr., *ibid.* **63**, 3464 (1988).
- <sup>22</sup>M. Sparks, J. Appl. Phys. **11**, 1018 (1970); R. C. Moul and M. G. Cottam, J. Phys. C **16**, 1307 (1983).