Spin-wave propagation on imperfect ultrathin ferromagnetic films

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(Received 24 February 1992; revised manuscript received 23 September 1992)

The effects of localized imperfections on spin-wave propagation in very thin ferromagnetic films are examined. The imperfections are assumed to be localized to a few lattice sites and cause local changes in anisotropy and exchange fields. These imperfections may be due to thickness variations or other geometrical imperfections. We find that in very thin films the lifetime of long-wavelength spin waves is relatively insensitive to scattering from even large numbers of imperfections, and therefore cannot explain large observed linewidths observed in Brillouin light-scattering experiments. On the other hand, we find that a band of long-wavelength spin-wave modes can exist in an inhomogeneous film with a distribution of effective anisotropy fields. It is possible to have large bands with bandwidths on the order of 10 GHz in rough ultrathin films due to the sensitive dependence of effective anisotropy fields on thickness.

I. INTRODUCTION

Magnetometry and Kerr measurements on molecularbeam-epitaxy (MBE)-grown ultrathin films of Co and Fe have indicated a spontaneous magnetization for a wide range of temperatures, but the various experimentally determined magnetic parameters may vary widely from sample to sample.^{1,2} Although it is generally supposed that roughness, inhomogeneities, impurities, and other imperfections are extremely important in determining the magnetic behavior of ultrathin ferromagnetic films, surprisingly little theoretical work has been done toward understanding the effects of imperfections on spin waves in very thin ferromagnetic films.^{3,4}

A particularly interesting finding has been the observation of a remarkably broad spin-wave peak in Brillouin light-scattering spectra taken on ultrathin ferromagnetic films.⁵ In thick films consisting of several hundred atomic layers, linewidths are typically very small—on the order of 0.5 GHz, which is less than the instrumental linewidth.⁶ But for films just a few atomic layers thick, linewidths on the order of 10 GHz or more are observed.^{5,7} The purpose of this paper is to examine the effects of imperfections on long-wavelength spin waves in order to estimate experimentally observable linewidths and frequency shifts.

We will consider two possible sources of linewidth broadening:

1. Lifetime shortening due to scattering from imperfections. Two types of imperfection would be occasional bumps, where an extra atom sits on top of the film, or occasional holes, where an atom is missing. A spin wave traveling with wave vector \mathbf{q} would be scattered by an imperfection into another state \mathbf{q}' . The linewidth of a mode \mathbf{q} observed by light-scattering techniques should correspond to the rate this mode scatters into all possible states q'. The linewidth of the mode is then proportional to the local changes in anisotropies or exchange interactions which produce the scattering. To calculate this, we develop a time-dependent perturbation theory for non-Hermitian systems.

2. The formation of new spin-wave states due to local thickness variations. This could occur, for example, on a rough film which has a distribution of regions with different thicknesses, each of which have different anisotropies. This kind of structure would create new spin-wave states, even when each region extends only over a few lattice sites, and could thus broaden a spectrum if the energies of these states lie near that of the state belonging to the perfect film.

In what follows we consider each of these mechanisms in detail and present some estimates of the consequent linewidths one might expect in possible light-scattered experiments. We also emphasize the approximations in our approach. In the second treatment, we consider only dipolar interactions and ignore possible complications due to exchange interactions. This puts limits on the validity of our theory, of course, but we expect that at low frequencies and for long-wavelength excitations the dipolar interactions will dominate over short-ranged exchange interactions.

In order to provide a background for what follows, it is useful to review some of the main features of spin waves in ultrathin films. Long-wavelength spin waves in ultrathin ferromagnetic films have been discussed by several authors, so here we will only review the relevant results. An ultrathin film may consist of several atomic layers, and we define "ultrathin" as being thin enough that only the lowest-order mode is appreciably populated at room temperatures. For Co and Fe this means films which are 1-3 atomic layers thick. The dispersion relation for such films can be shown to have the form⁸

$$\omega/\gamma = [H_0 + A_1 + Dq^2 + 4\pi M_s(f - qd/2)][H_0 + A_2 + Dq^2 + 2\pi M_s qd\sin^2\phi] .$$
⁽¹⁾

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Here H_0 is the external applied field, D is an exchange constant in field units, d is the thickness of the film, M_S is the saturation magnetization, \mathbf{q} is the wave vector of the excitation, and ϕ is the angle between **q** and H_0 . Effective anisotropy fields are represented by A_1 and A_2 , and are combinations of in-plane and out-of-plane anisotropies. Also, $\gamma = g\mu_B / \hbar$, where g is the g factor of the material, μ_B is the Bohr magnetron, and \hbar is Planck's constant. The factor f arises due to an interesting feature of these modes in that the dipolar demagnetizing fields for an ultrathin film differ from those for a thick film. For a monolayer, j = 1.078. The dipolar fields in two dimensions appear to be especially important in stabilizing the two-dimensional ferromagnetic order at finite temperatures by changing the manner in which ω goes to zero as q goes to zero.

II. IMPURITY SCATTERING

We define our geometry such that the film lies in the yz plane and assume that both the magnetization and an external applied field lie in the z direction. The magnetization in each layer of a film is assumed to consist of a small dynamic part $\mathbf{m}(y,z,t)$ and a static part $\widehat{\mathbf{z}}M_S$. The dynamic part exists in the xy plane and varies in time according to $\exp(-i\omega t)$. We now want to examine the equations of motion, and study the effects of localized impurities on the allowed ω 's and \mathbf{m} 's using perturbation theory.

The magnetization obeys the Bloch equations of motion, which also relate \mathbf{m} to dipolar fields \mathbf{h} . In general, one has equations of motion for \mathbf{m} in each atomic layer, but a great simplification can be made for the lowest-energy mode of an ultrathin film by noting that the relative phases between the precessing magnetic moments varies slowly from atomic layer to atomic layer. For films that are only a few atomic layers thick, the \mathbf{m} and \mathbf{h} fields can be assumed to be independent of position in the direction normal to the atomic planes. Plane-wave solutions for \mathbf{m} and \mathbf{h} are assumed, and have the forms

$$\mathbf{m}(\mathbf{r}) = \mathbf{m}e^{i\mathbf{q}\cdot\mathbf{r}} \tag{2}$$

and

$$\mathbf{h}(\mathbf{r}) = \mathbf{h}e^{i\mathbf{q}\cdot\mathbf{r}} \,. \tag{3}$$

Here \mathbf{q} and \mathbf{r} lie in the film plane.

This problem has been solved in Ref. 8, and so we only describe the calculation and present the relevant results here. First, Bloch's equations are solved using solutions of the form shown in Eqs. (2)-(3), and the exchange boundary conditions are applied. The resulting equations provide an expression relating **m** and **h**. Next, the magnetostatic form of Maxwell's equations are solved and the electromagnetic boundary conditions are applied. These equations are then used to derive a second expression relating **m** and **h**. Finally, we eliminate **h** between the two expressions and obtain the following eigenvalue equation for **m**:

$$\frac{d}{dt}\mathbf{m} = -i\omega\mathbf{m} = \gamma \underline{L}\mathbf{m} , \qquad (4)$$

where \underline{L} is given by

$$\underline{L} = \begin{bmatrix} 0 & L_1 \\ -L_2 & 0 \end{bmatrix} .$$
(5)

The fields L_1 and L_2 are defined as

$$L_1 = Dq^2 + H_0 + A_1 + 2\pi M_s q d \sin^2 \phi , \qquad (6)$$

$$L_2 = Dq^2 + H_0 + A_2 + 4\pi M_s \left[f - \frac{qd}{2} \right] . \tag{7}$$

The positive eigenvalue of Eq. (4) is then given by $\omega/\gamma = (L_1L_2)^{1/2}$, which can be written as in Eq. (1) using Eqs. (6) and (7). Some care must be taken when solving for the eigenvectors of Eq. (4), however, since the matrix **L** is not Hermitian.⁹ To find the correct eigenvectors, we define right and left vectors \mathbf{m}_r and \mathbf{m}_l such that

$$-i\frac{\omega}{\gamma} = \mathbf{m}_{l}^{*} \underline{L} \,\mathbf{m}_{r} \,\,. \tag{8}$$

We also require that the modes are normalized across the film thickness as follows:

$$1 = \sum_{j=1}^{N_L} \int d^2 r \mathbf{m}_l^* \cdot \mathbf{m}_r , \qquad (9)$$

where the sum runs over the atomic layers, 1 to N_L , and the integration is over the sample area W^2 . The resulting right and left eigenvectors for the $+\omega$ eigenvalue are

$$\mathbf{m}_{r}(\mathbf{q}) = \frac{1}{W\sqrt{2N_{L}L_{2}}} \begin{bmatrix} i\sqrt{L_{1}} \\ \sqrt{L_{2}} \end{bmatrix} e^{i\mathbf{q}\cdot\mathbf{r}} , \qquad (10)$$

$$\mathbf{m}_{l}(\mathbf{q}) = \frac{1}{W\sqrt{2N_{L}L_{1}}} [i\sqrt{L_{2}}, \sqrt{L_{1}}]e^{i\mathbf{q}\cdot\mathbf{r}} .$$
(11)

Our goal is to describe the effects of an impurity on the eigenmodes given by Eqs. (10) and (11) We define an impurity as a local change \underline{L}' from the effective field \underline{L} , where the components of \underline{L}' are defined as Δ_1 and Δ_2 such that

$$\underline{L}' = \begin{bmatrix} 0 & \Delta_1 \\ -\Delta_2 & 0 \end{bmatrix} a^2 \delta(\mathbf{r}_i) .$$
 (12)

The impurity is located at position r_i and assumed to extend over an area a.²

We proceed as in standard time-dependent perturbation theory. The dynamic magnetization at any time is defined as $\Psi(t)$ and written as a sum over allowed states **q** with time-dependent coefficients $a_q(t)$. These are defined separately for the right and left eigenvectors:

$$\Psi_r(t) = \sum_{\mathbf{q}} a_{\mathbf{q}}(t) e^{-i\omega t} \mathbf{m}_r(\mathbf{q}) , \qquad (13)$$

$$\Psi_l(t) = \sum_{\mathbf{q}} b_{\mathbf{q}}(t) e^{-i\omega t} \mathbf{m}_l(\mathbf{q}) .$$
(14)

Writing the perturbed equations of motion for the right eigenvector as

$$\frac{d}{dt}\Psi_{r} = \gamma(\underline{L} + \underline{L}')\Psi_{r} , \qquad (15)$$

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we arrive at the following equation for a_q (we drop explicit reference to the time dependence of the coefficients for simplicity):

$$\frac{d}{dt}a_{\mathbf{q}} = \gamma \sum_{\mathbf{q}'} a_{\mathbf{q}'} e^{it[\omega(\mathbf{q}') - \omega(\mathbf{q})]} M_{lr}(\mathbf{q} - \mathbf{q}') , \qquad (16)$$

where the perturbation is contained in the matrix element M_{lr} defined by

$$M_{lr}(\mathbf{q}-\mathbf{q}') = \sum_{j=1}^{N_L} \int d^2 \mathbf{r} \, \mathbf{m}_l^*(\mathbf{q}') \underline{L}' \mathbf{m}_r(\mathbf{q}) \,. \tag{17}$$

An equation similar to Eq. (16) can be derived for the b_q , with the difference that the matrix element M_{rl} appears in place of M_{lr} :

$$M_{rl}(\mathbf{q}-\mathbf{q}') = \sum_{j=1}^{N_L} \int d^2 r \, \mathbf{m}_r(\mathbf{q}) \cdot [\mathbf{m}_l^*(\mathbf{q}')\underline{L}'] \,. \tag{18}$$

In our problem, L' is such that $M_{rl} = M_{lr}$.

The coefficients a_q and b_q are approximated using standard perturbation theory with the requirement that at time t = 0 only state a_{q_0} , b_{q_0} is occupied and that the perburbation L' is turned on slowly. Since we are interested in the lifetime of state q_0 , we seek the solutions to

$$\frac{d}{dt}a_{\mathbf{q}_0} = \frac{1}{\Gamma_a}a_{\mathbf{q}_0} \tag{19}$$

and

$$\frac{d}{dt}b_{\mathbf{q}_0} = \frac{1}{\Gamma_b}b_{\mathbf{q}_0} \,. \tag{20}$$

The quantity $b_{\mathbf{q}_0}^* a_{\mathbf{q}_0}$, which measures the magnitude of the fluctuation in the magnetization with energy $\omega(\mathbf{q}_0)$, thus decays in time at the rate

$$\frac{1}{\Gamma} = \operatorname{Re}\left[\frac{1}{\Gamma_a} + \frac{1}{\Gamma_b}\right],\qquad(21)$$

where Re denotes the real part.

To second order in the perturbation, we find

$$\frac{1}{\Gamma} = 2\pi\gamma^2 \sum_{\mathbf{q}\neq\mathbf{q}_0} |\boldsymbol{M}_{lr}|^2 \delta[\omega(\mathbf{q}) - \omega(\mathbf{q}_0)] , \qquad (22)$$

where the summation is over all states other than the initial \mathbf{q}_0 state. This expression can be evaluated by first converting to an integral over \mathbf{q} with an appropriate density of states. The difficulty is that the resulting integral is best solved numerically when the exact density of states is used because of the directional dependence of the frequency on the propagation angle ϕ . In what follows, however, we will make a simple estimate by ignoring the angular dependence on ϕ . Such an approximation is unwarranted in a bulk sample, but is reasonable in ultrathin films, as can be seen from Eq. (1).

For this approximation, we assume that $qd \ll 1$, and calculate the resulting density of states $\rho(\omega)$:

$$\rho(\omega) = \frac{\sqrt{L_1 L_2}}{2\gamma D(L_1 + L_2)} .$$
⁽²³⁾

If the total number of spins is N, then $W = Na^2$ for a simple cubic lattice. Using Eqs. (10)–(12), (17), and (22), we find that the deay rate for n impurities is

$$\frac{1}{\Gamma} = \frac{a^2}{8} \left[\frac{n}{N} \right] \frac{\gamma}{D} \frac{(L_1'L_2 + L_2'L_1)}{(L_1 + L_2)\sqrt{L_1L_2}} .$$
(24)

For simplicity, consider the special case when the perturbation is isotropic so that $L'_1 = L'_2 = L'$. We define the change in energy represented by the perturbation as

$$\Delta \omega = \gamma L' , \qquad (25)$$

so that the decay rate is written as

$$\frac{1}{\Gamma} = \frac{a^2}{8} \left[\frac{n}{N} \right] \frac{(\Delta \omega)^2}{\gamma D} \frac{(2Dq^2 + 4\pi M_s f)}{\sqrt{Dq^2(Dq^2 + 4\pi M_s f)}} .$$
(26)

Thus the lifetime of the spin wave is inversely proportional to the fractional number of impurities on the film, and directly proportional to the strength of the exchange interaction.

For a numerical estimate of Eq. (26), we use parameters appropriate to bulk Co: $\gamma = 17.6 \times 10^6$ rad/Oe, $\gamma D = 0.0056$ Hz cm⁻², $4\pi Ms = 17.6$ kG, and $a = 2 \times 10^{-8}$ cm. For simplicity, f is set equal to 1. A large effective anisotropy field due to a variation in thickness, for example, could be on the order of 10 kG with a corresponding change in frequency of $\Delta \omega = 10$ GHz. Even with a large fraction of impurities of the order of 0.01, the decay rate will be on the order of

$$\frac{1}{\Gamma} \approx 0.0001 \text{ GHz} . \tag{27}$$

In units of field, $1/\gamma \Gamma \approx 1$ G, which is on the same order of magnitude as the scattering of the ferromagnetic resonance mode from surface pits.¹⁰ But this linewidth is much smaller than that observed or even observable with Brillouin light scattering.

III. THICKNESS VARIATIONS AND ROUGHNESS

Variations in film thickness, i.e., roughness, can change the effective anisotropy fields experienced by longwavelength spin-wave excitations in a thin film. We will show that even in the limit that the effective anisotropy fields vary over length scales much shorter than the wavelength of the spin wave, these variations can lead to new allowed spin-wave modes. Consequently, a distribution of anisotropies along a thin film may create a band of spin-wave frequencies.

We will consider only long-wavelength spin-wave excitations in the dipolar limit and assume that exchange interactions are negligible. The film lies in the yz plane as before, and the magnetization is directed along the z axis.

We will model these variations with appropriately weighted magnetic permeabilities using an effectivemedium approach. In order to form macroscopic magnetic permeabilities, we must perform an average over several lattice sites of the true microscopic anisotropies. We do this by assigning effective uniaxial anisotropy fields which are uniform over several tens or hundreds of lattice sites. For the thin films considered here, the effective anisotropy fields are therefore constant across the film thickness, but may vary as one moves across the film plane. This approximation should be valid when the wavelength of the spin wave is much larger than the period of the variations.

For simplicity, we consider the idealized case of anisotropy variations in the y direction only. The variations are shown schematically in Fig. 1, and are defined such that an effective uniaxial anisotropy field H_1 acts in regions of width d_1 and an effective uniaxial anisotropy field H_2 acts in regions of width d_2 , etc.

Using arguments similar to those in Refs. 11 and 12, one can show that the frequency of the long-wavelength mode in ultrathin film is given by

$$\mu_{xx} = 0 , \qquad (28)$$

where μ_{xx} is the xx component of the magnetic permeability tensor given by $1+\chi$, where χ is the susceptibility defined by $\mathbf{m} = \chi \mathbf{h}$ for a uniform film. In the limit of very long wavelengths, μ_{xx} for the uniform material is given by

$$\mu_{xx} = 1 + \frac{4\pi M_s H_i}{H_i^2 - \omega^2 / \gamma^2} , \qquad (29)$$

where

$$H_i = H_0 + A$$
 . (30)

The anisotropy field is represented by A, and for simplicity is taken to be the same for both in-plane and out-ofplane directions.

We now calculate the effective-medium permeabilities for a simple example of a film with only two different anisotropies. The fields in a stripe are denoted by the subscripts "1" and "2," which correspond to anisotropy fields A_1 and A_2 , respectively. As in Refs. 11 and 12, we assume that the magnetic **b** and **h** fields are approximately constant across each stripe in the film. The x component of the average magnetic field, $\langle \mathbf{b} \rangle$, is defined as

$$\langle b \rangle_{x} = f_{1}b_{1x} + f_{2}b_{2x}$$
, (31)



FIG. 1. Schematic representation of a thin film with different regions of effective anisotropy fields. The true film structure is replaced by a film of uniform thickness but with regions of different lengths where the effective anisotropy fields are different.

where the filling factors are defined as

$$f_1 = \frac{d_1}{d_1 + d_2} , \qquad (32)$$

$$f_2 = \frac{d_2}{d_1 + d_2} \ . \tag{33}$$

We now define the effective permeabilities $\langle \mu_{xx} \rangle$ and $\langle \mu_{xy} \rangle$ such that

$$\langle b \rangle_{x} = \langle \mu_{xx} \rangle \langle h \rangle_{x} + \langle \mu_{xy} \rangle \langle h \rangle_{y} , \qquad (34)$$

where the components of $\langle \mathbf{h} \rangle$ are the average magnetic induction fields. Under the assumption that the fields are essentially constant within each stripe, continuity of tangential **h** requires that

$$h_{1y} = h_{2y} = \langle h \rangle_y , \qquad (35)$$

$$h_{1x} = h_{2x} = \langle h \rangle_x . \tag{36}$$

From Eq. (34) one then finds

$$\langle \mu_{xx} \rangle = f_1 \mu_{xx}^{(1)} + f_2 \mu_{xx}^{(2)},$$
(37)

where $\mu_{xx}^{(1)}$ and $\mu_{xx}^{(2)}$ are the permeabilities associated with each individual region of the form given by Eqs. (29)-(31).

The frequencies of the allowed spin-wave modes are given by setting $\langle \mu_{xx} \rangle$ to zero as in Eq. (28). One finds that the effective-medium permeability for this example gives one mode for each region, so that there are a total of two allowed modes. The frequencies of these modes depend on the anisotropy in each region, so that the closer the anisotropies are to one another, the closer the two frequencies will be.

The frequencies of the spin-wave modes for this example are shown in Fig. 2 as functions of f_1 . The frequency is shown in reduced units $\omega/\gamma 4\pi M_S$. The applied field is $H_0 = 2\pi M_S$, $H_1 = 4\pi M_S$, and $H_2 = 0.4\pi M_S$. The arrows show the frequencies of a spin wave for each material taken separately, denoted by ω_1 and ω_2 , respectively, and the solid lines are for the stripe geometry. As f_1 goes to 1, the frequency of the upper mode approaches ω_1 and



FIG. 2. Allowed propagation frequencies on a film with two alternating regions with different anisotropies. The frequencies are shown as a function of the filling factor f_1 , which determines the relative sizes of each region. When f_1 is 0 or 1 the film is uniform.

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the frequency of the lower mode approaches $\gamma(H_0 + H_2)$. As f_1 goes to 0, the frequency of the upper mode approaches $\gamma(H_0 + H_1)$, and that of the lower mode approaches ω_2 . The modes with frequencies near $\gamma(H_0 + H_1)$ or $\gamma(H_0 + H_2)$ are resonances which would make little contribution to a light-scattering intensity, since they occur only in the limit of an almost perfectly uniform film.

For values of f_1 well away from one and zero, both modes should make significant contributions to a lightscattering intensity and, as pointed out above, the difference between the frequencies depends on the difference between the anisotropies. Also, it simple to extend the above argument to the case of several regions of different anisotropies. One finds that a spin-wave mode exists for each different anistropy. Thus in order to achieve a broad band of frequencies, a distribution of anisotropy fields is required. In this case we would have

$$\langle \mu_{xx} \rangle = \sum_{i} f_{i} \mu_{xx}^{(i)} . \tag{38}$$

Here f_i is the filling factor for the *i*th region and is given by

$$f_i = \frac{d_i}{\sum_i d_i} , \qquad (39)$$

and $\mu_{xx}^{(i)}$ is the permeability of the *i*th region and d_i is the width of the *i*th region. It can be shown that variations of the local anisotropy fields in the z direction lead to similar results.

IV. CONCLUSIONS

We have examined the propagation of long-wavelength spin waves on ultrathin ferromagnetic films which have imperfections that cause local changes in the effective magnetic fields through variations in anisotropies. We found that for the case of nearly perfect films with widely spaced imperfections, the scattering of a spin wave with wave vector \mathbf{q} into other wave-vector states will lead to lifetimes on the order of 10^{-9} s. The corresponding lifetime corresponds to linewidths observed in light-scattering experiments, and would be much smaller than 1 GHz.

We have also examined the other extreme where anisotropy variations are clustered together inside regions much smaller than the wavelength of the spin wave. Here we studied the electromagnetic problem of quasiperiodic variations in the local anisotropy field by modeling the film with an effective magnetic permeability. We found that a broad of long-wavelength spin-wave states could be formed by a distribution of anisotropies. The width of the spin-wave band would be determined by the minimum and maximum values of the variations in the anisotropies.

Finally, we repeat that these calculations have ignored the effects of exchange interactions. The magnitude of this assumption is not clear, but recent theoretical work suggests that geometrical imperfections can have an extremely large effect on the propagation of exchangedominated spin waves in ultrathin films.¹³ A difficult but necessary step is to include both exchange and dipolar interactions into the description of spin-wave propagation on rough film films.

ACKNOWLEDGMENTS

This work was partially supported by the U.S. Army Research Office through Grant No. DAAL0391-G-0229. One of us (R.L.S.) also thanks the Alexander von Humboldt Foundation for support.

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