

Extrinsic contributions to the ferromagnetic resonance response of ultrathin films

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We develop a theory of the extrinsic contributions to the ferromagnetic resonance linewidth and frequency shift of ultrathin films. The basic mechanism is two magnon scattering by defects at surfaces and interfaces. In the presence of dipolar couplings between spins in the film, one realizes short wavelength spin waves degenerate with the ferromagnetic resonance (FMR) mode, provided the magnetization is parallel to the film surfaces. Defects on the surface or interface thus scatter the FMR mode into such short wavelength spin waves, producing a dephasing contribution to the linewidth, and a frequency shift of the resonance field. The mechanism described here is inoperative when the magnetization is perpendicular to the film.

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I. INTRODUCTION

There is currently great interest in the physical properties of ultrathin (very few atomic layer) ferromagnetic films, and magnetic multilayers formed from such spins. While very considerable attention is devoted to transport properties in such structures, because of applications to magnetic recording and data storage, in fact the microwave response of these systems is of great interest as well. Studies of the ferromagnetic resonance (FMR) spectrum are a rich source of information on the unique anisotropies found in these materials,^{1,2} and other physical properties as well. Furthermore, hybrid structures formed by depositing ferromagnetic films or multilayers on semiconducting substrates may form the basis for high-frequency microwave devices.³

Rather little attention has been directed toward the origin of the FMR linewidth in the ultrathin films. In bulk crystalline Fe, one observes an intrinsic contribution which varies linearly with the FMR frequency.^{4,5} That this should be the case is a prediction of the phenomenological Landau-Lifshitz equations. If this contribution has its microscopic origin in the coupling between spin motions, and the itinerant electrons in Fe, one may argue the linear frequency dependence follows from very general considerations in this low-frequency regime. In the ultrathin films, the analysis of FMR data^{1,6} shows the presence of a linear term, often with slope larger than that found in single-crystal Fe. In addition, extrapolation to zero frequency yields a rather substantive “zero-frequency linewidth,” with evident origin in surface or interface quality.^{1,6}

In this paper, we develop a theory of the extrinsic linewidth of ultrathin ferromagnetic films, based on the following picture. In the idealized FMR experiment, a uniform mode is excited whose wave vector \vec{k}_{\parallel} parallel to the surface is zero. For a simple film with magnetization \vec{M}_s parallel to the surface, the frequency of this mode (in the absence of anisotropy) is $\gamma[H_o(H_o + 4\pi M_s)]^{1/2}$, with γ the gyromagnetic ratio and M_s the magnetization. We show below, as discussed by earlier authors,⁷ that in the presence of dipolar couplings between spins, we have short wavelength spin waves with wavelength $k_{\parallel} \approx 10^5 \text{ cm}^{-1}$ degenerate with the

FMR mode, for ultrathin films of the ferromagnetic transition metals. Defects in the surface, modeled below, scatter energy from the uniform mode to these states, thus producing relaxation of dephasing character. We find, in the analysis presented here, a defect induced frequency shift as well.

The notion that such a two magnon process controls the extrinsic linewidth is by no means new. Indeed, in a seminal paper several decades ago, Sparks, Loudon, and Kittel⁸ developed a picture such as this to explain the origin of the extrinsic linewidth in yttrium iron garnet spheres. In their case, the surface defects had their origin in the grit used to polish the surface. We argue here that in ultrathin ferromagnetic films, the spin-wave dispersion is such that the two magnon mechanism is operative, provided the magnetization is parallel to the surface.

Our attention is confined to films sufficiently thin so that only a single spin-wave branch of acoustic character controls the magnetic response. In the ultrathin film limit, standing spin waves with nonzero wave vectors perpendicular to the film surface, $k_{\perp}^{(n)} = n\pi/d$ where $n \neq 1$ and d the film thickness, are upshifted by exchange to high frequencies well above those in the FMR range. It would be desirable to present the theory in more general form, valid for films or arbitrary thickness. Such an analysis will be very complex indeed, so we confine our attention to the ultrathin film limit.

We note that in a recent paper,¹⁰ McMichael, Stiles, Chen, and Egelhoff presented a brief, qualitative discussion of a two magnon contribution to the linewidth based on the same physical picture we employ as a basis. One finds no explicit results in their paper beyond a general expression for the scattering rate, however. Here we set forth a specific model of surface defects which may couple the FMR mode to the short-wavelength spin waves. Within this framework, we provide explicit predictions for the dependence of the extrinsic linewidth on the magnetic field at resonance, and for the defect induced frequency shift as well.

Quite recently Hurben and Patton⁹ presented a theory of extrinsic contributions to the linewidth of ferromagnetic films, also based on two magnon scattering induced by defects. Their theory is valid in what one may call the thick-film limit, where the modes degenerate with the FMR mode may be viewed as propagating, three-dimensional plane

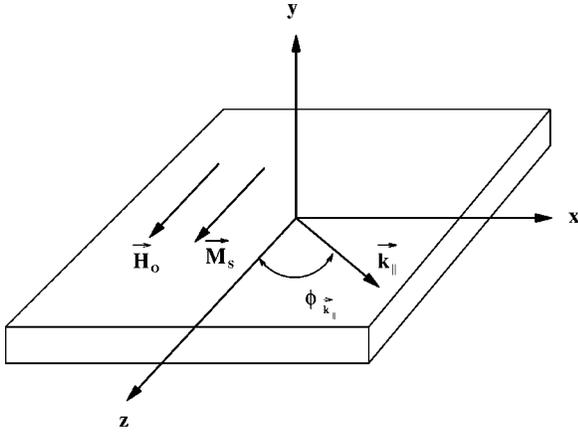


FIG. 1. Geometry of an ultrathin film of thickness d . The applied field \vec{H}_0 and the main component of the magnetization \vec{M}_s are lined up with the z axis, in plane. The angle $\phi_{\vec{k}_{\parallel}}$ is that between the magnon wave vector \vec{k}_{\parallel} and the z axis.

waves unaffected by the presence of surfaces. Such a picture is appropriate, say, for garnet films many microns in thickness, but not for the ultrathin films that are the focus of the present paper.

In Sec. II, we discuss the nature of the spin-wave dispersion in ultrathin films, and then introduce the mechanism that forms the basis for our analysis. Through means of an equation of motion method, in Sec. III we derive expressions for both the contribution to the linewidth and the frequency shift of the FMR resonance, from the two magnon process. Section IV is devoted to the development of a model of surface defects and their contribution to the matrix elements which control the two magnon mechanism. Section V presents our formulas for the two magnon contribution to the linewidth and frequency shift, and Sec. VI shows representative plots of the theoretically calculated extrinsic resonance linewidth and frequency shifts, as well as concluding remarks.

II. SPIN-WAVE DISPERSION IN ULTRATHIN FERROMAGNETIC FILMS; RELATION WITH TWO MAGNON MECHANISM

Our discussion will be directed toward an ultrathin ferromagnetic film, such as that illustrated in Fig. 1. The magnetization \vec{M}_s lies in the plane, parallel to the dc magnetic field \vec{H}_0 . We consider a spin wave which propagates in the plane of the film, with wave vector \vec{k}_{\parallel} which makes the angle $\phi_{\vec{k}_{\parallel}}$ with \vec{H}_0 and \vec{M}_s . As mentioned in Sec. I, the thickness d of the film is sufficiently thin that we need be concerned with only the low-lying acoustical spin-wave branch. The standing-wave modes are shifted upward by exchange in such thin films sufficiently that they play no role in the considerations that follow, as noted above.

To describe the spin waves, we write the magnetization in the form $\vec{M}(\vec{r}, t) = M_s \hat{z} + \vec{m}(\vec{r}, t)$ when a mode is excited, where $\vec{m}(\vec{r}, t) = m_x(\vec{r}, t)\hat{x} + m_y(\vec{r}, t)\hat{y}$. Since only the low-lying acoustical branch is of interest, we may phrase the discussion entirely in terms of the transverse magnetization components, averaged over the film profile. Thus we consider

$$m_{x,y}(x, z, t) = \int_0^d m_{x,y}(x, y, z; t) \frac{dy}{d}. \quad (1)$$

We shall Fourier transform these amplitudes:

$$m_{x,y}(x, z; t) = \frac{1}{\sqrt{L^2 d}} \sum_{\vec{k}_{\parallel}} m_{x,y}(\vec{k}_{\parallel}; t) e^{i\vec{k}_{\parallel} \cdot \vec{r}_{\parallel}}, \quad (2)$$

where L^2 is the area of the film, $\vec{k}_{\parallel} = k_x \hat{x} + k_z \hat{z}$, and similarly for \vec{r}_{\parallel} . Note that $m_{x,y}(-\vec{k}_{\parallel}) = m_{x,y}(\vec{k}_{\parallel})^*$.

Of interest in our discussion of the spin wave dispersion in the film is the contribution to the spin-wave energy from dipolar fields generated by the spin motions. This may be written

$$H_d = -\frac{1}{2} \frac{1}{\sqrt{L^2 d}} \times \sum_{\vec{k}_{\parallel}} \vec{m}(\vec{k}_{\parallel}; t) \cdot \int e^{i\vec{k}_{\parallel} \cdot \vec{r}_{\parallel}} \vec{h}^{(d)}(x, y, z; t) dx dy dz, \quad (3)$$

where the integration is over the volume of the film. In what follows, we omit explicit reference to the time.

We decompose the dipolar field into two contributions: $\vec{h}_d^{(1)}$ from bulk magnetic charges and a second $\vec{h}_d^{(2)}$ from surface magnetic charges. We consider each in turn. We may write

$$\vec{h}_d^{(1)}(\vec{r}) = -\nabla \Phi_M^{(1)}(\vec{r}), \quad (4)$$

where, noting $-\nabla \cdot \vec{m}$ behaves as an effective magnetic charge density, the magnetic potential is

$$\begin{aligned} \Phi_M^{(1)}(\vec{r}) &= -\int \frac{d^3 r'}{|\vec{r} - \vec{r}'|} \nabla' \cdot \vec{m}(\vec{r}') \\ &= -\int \frac{1}{|\vec{r} - \vec{r}'|} \frac{\partial m_x}{\partial x'}(x', z') dx' dy' dz'. \end{aligned} \quad (5)$$

Thus

$$\Phi_M^{(1)}(\vec{r}) = -\frac{i}{\sqrt{L^2 d}} \sum_{\vec{k}_{\parallel}} k_x m_x(\vec{k}_{\parallel}) \int \frac{e^{i\vec{k}_{\parallel} \cdot \vec{r}'}}{|\vec{r} - \vec{r}'|} dx' dy' dz'. \quad (6)$$

The integral in Eq. (6) is readily evaluated. In the thin-film limit $k_{\parallel} d \ll 1$, the contribution to the dipole field (averaged over the film thickness) from bulk magnetic charges is then found to be

$$\begin{aligned} \vec{h}_d^{(1)}(\vec{r}) &= -\frac{2\pi}{\sqrt{L^2 d}} \sum_{\vec{k}_{\parallel}} k_{\parallel} d \sin \phi_{\vec{k}_{\parallel}} m_x(\vec{k}_{\parallel}) e^{i\vec{k}_{\parallel} \cdot \vec{r}_{\parallel}} \\ &\quad \times (\sin \phi_{\vec{k}_{\parallel}} \hat{x} + \cos \phi_{\vec{k}_{\parallel}} \hat{z}). \end{aligned} \quad (7)$$

Of central importance to what follows is the linear variation of $\vec{h}_d^{(1)}$ with wave vector in Eq. (7). The fact that we have short-wavelength modes degenerate with the FMR mode in the ultrathin film follows from this result.

The second contribution to the dipolar field and its contribution to the spin-wave energy arises from fields generated by surface magnetic charges. In a ferromagnet, if \hat{n} is the outward surface normal, then $\hat{n} \cdot \vec{M}$ is an effective magnetic surface charge density. Thus, when the spin wave is excited, we have the surface charge density $+m_y(\vec{k}_\parallel) e^{i\vec{k}_\parallel \cdot \vec{r}_\parallel}$ on the upper surface, and $-m_y(\vec{k}_\parallel) e^{i\vec{k}_\parallel \cdot \vec{r}_\parallel}$ on the lower. In the thin-film limit $k_\parallel d \ll 1$, the dominant contribution to the dipole field from this source has the form

$$\vec{h}_d^{(2)}(\vec{r}) = -\frac{4\pi}{\sqrt{L^2 d}} \hat{y} \sum_{\vec{k}_\parallel} \left(1 - \frac{k_\parallel d}{2}\right) m_y(\vec{k}_\parallel) e^{i\vec{k}_\parallel \cdot \vec{r}_\parallel}, \quad (8)$$

where we retain the contribution linear in \vec{k}_\parallel . When these fields are inserted into Eq. (3) and the resulting integration carried out, the contribution to the spin-wave excitation energy of the ultrathin film, in the notation introduced here, has the form

$$H_d = 2\pi \sum_{\vec{k}_\parallel} \left(1 - \frac{k_\parallel d}{2}\right) m_y^*(\vec{k}_\parallel) m_y(\vec{k}_\parallel) + \pi \sum_{\vec{k}_\parallel} k_\parallel d \sin^2 \phi_{\vec{k}_\parallel} m_x^*(\vec{k}_\parallel) m_x(\vec{k}_\parallel). \quad (9)$$

There are, of course, other contributions to the spin-wave energy. These are

(a) The Zeeman energy:

$$H_z = -H_o \int_V M_z(x, z) dx dy dz = -H_o M_s V + \frac{H_o}{2M_s} \int_V [m_x^2(x, z) + m_y^2(x, z)] dx dy dz, \quad (10)$$

where V is the volume of the film. The constant term is discarded, leaving

$$H_z = \frac{H_o}{2M_s} \sum_{\vec{k}_\parallel} [m_x^*(\vec{k}_\parallel) m_x(\vec{k}_\parallel) + m_y^*(\vec{k}_\parallel) m_y(\vec{k}_\parallel)], \quad (11)$$

when we use the variables introduced above.

(b) The exchange energy:

$$H_x = \frac{A}{M_s^2} \int_V [|\nabla m_x|^2 + |\nabla m_y|^2] dx dy dz = \frac{1}{2M_s} \sum_{\vec{k}_\parallel} Dk_\parallel^2 [m_x^*(\vec{k}_\parallel) m_x(\vec{k}_\parallel) + m_y^*(\vec{k}_\parallel) m_y(\vec{k}_\parallel)], \quad (12)$$

where the exchange stiffness is $D = 2A/M_s$.

(c) The surface anisotropy energy:

$$H_A = \frac{K_s}{M_s^2} \int_S m_y^2(x, z) dx dz = \frac{1}{2M_s} H_s \sum_{\vec{k}_\parallel} m_y^*(\vec{k}_\parallel) m_y(\vec{k}_\parallel), \quad (13)$$

where $H_s = 2K_s/M_s d$. Note H_s is here positive when the y direction is a hard axis.

Of course, in ultrathin films, an additional source of uniaxial anisotropy normal to the surface has origin in alteration of interlayer spacings. If, for example, the film is grown on a substrate whose lattice constant is larger than the bulk crystal form of the ultrathin film, there will be a decrease of interlayer spacing. Such effects just provide a supplement to H_s .

When all these terms are combined, we have a Hamiltonian which may be written

$$H = \frac{1}{2M_s} \sum_{\vec{k}_\parallel} \{H_x(\vec{k}_\parallel) m_x^*(\vec{k}_\parallel) m_x(\vec{k}_\parallel) + H_y(\vec{k}_\parallel) m_y^*(\vec{k}_\parallel) m_y(\vec{k}_\parallel)\}, \quad (14)$$

where

$$H_x(\vec{k}_\parallel) = H_o + 2\pi M_s k_\parallel d \sin^2 \phi_{\vec{k}_\parallel} + Dk_\parallel^2, \quad (15)$$

and

$$H_y(\vec{k}_\parallel) = B_o + H_s - 2\pi M_s k_\parallel d + Dk_\parallel^2, \quad (16)$$

where $B_o = H_o + 4\pi M_s$.

It should be remarked that we could include other forms of anisotropy in our model Hamiltonian, such as the fourfold in plane anisotropy. This would complicate various formulas considerably, if \vec{H}_o is not applied parallel to the easy axis. We shall assume, in the interest of simplicity, that H_o is along the easy axis. Then the influence of the fourfold anisotropy is then simply to replace H_o by $H_o + H_a$, where H_a is the effective in plane anisotropy field. If the field H_o is applied along the hard axis, and H_o is sufficiently large for the magnetization to align along the hard axis, then H_o is replaced by $(H_o - H_a)$ everywhere.

If $(-\gamma)$ is the gyromagnetic ratio, then in our model, the spin-wave frequency is given by

$$\Omega(\vec{k}_\parallel) = \gamma [H_x(\vec{k}_\parallel) H_y(\vec{k}_\parallel)]^{1/2}. \quad (17)$$

Now we may discuss the basis for the two magnon scattering process that is the central mechanism explored in the present paper, and which was discussed as well in Ref. 10.

First, in an FMR experiment, the mode with wave vector $\vec{k}_\parallel = 0$ is excited. Its frequency is given by the well-known expression

$$\Omega_{FM} = \gamma \sqrt{H_o(H_o + H_s + 4\pi M_s)} \quad (18)$$

for a film with magnetization parallel to the surface.

Consider the wave-vector dependence of the spin wave dispersion relation in Eq. (17). We keep terms through those quadratic in the wave vector, noting that for the metallic films of interest the contribution of the dipolar energy quadratic in the wave vector is small. Thus

$$\Omega^2(\vec{k}_\parallel) = \Omega_{FM}^2 - 2\pi\gamma^2 M_s k_\parallel d (H_o - [B_o + H_s] \sin^2 \phi_{\vec{k}_\parallel}) + \gamma^2 (B_o + H_s + H_o) Dk_\parallel^2. \quad (19)$$

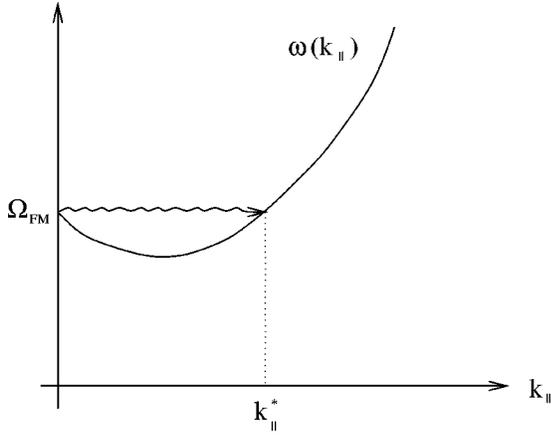


FIG. 2. Graphical representation of the scattering process from a uniform mode to a mode degenerate with it, of wave vector k_{\parallel}^* , which can occur if $\sin^2 \phi_{\vec{k}_{\parallel}} < H_o / (H_s + B_o)$.

In the ultrathin film limit, we see from Eq. (19) that the dipolar energy generates a term in the dispersion relation *linear* in the wave vector. As noted some time ago by Erickson and Mills,⁷ for a range of propagation angles the initial slope is negative. Furthermore, these authors discussed circumstances where the negative slope can actually drive $\Omega^2(\vec{k}_{\parallel})$ negative, producing an instability of the uniformly ferromagnetic state. What is important here is that exchange leads to the positive term quadratic in wave vector, and thus at finite wave vectors, we have modes degenerate with the uniform FMR mode. We encounter the negative slope, and thus finite wave-vector modes degenerate with the FMR mode when

$$\sin^2 \phi_{\vec{k}_{\parallel}} < \frac{H_o}{B_o + H_s}. \quad (20)$$

Some numbers are of interest. Typically, if we have Fe films in mind, FMR measurements employ dc fields $H_o \ll B_o$. Then setting H_s aside for the moment, the value of k_{\parallel} of the modes degenerate with the FMR frequency is $k_{\parallel} \approx 2\pi M_s d / D$. For Fe, $D = 2.5 \times 10^{-9}$ G cm², so for a film 30-Å thick, we have $k_{\parallel} \approx 10^6$ cm⁻¹. Macroscopic theory, such as that used here, is adequate to describe such modes.

After the FMR mode is excited, defects on the surface scatter the $\vec{k}_{\parallel} = 0$ FMR modes into the finite wave-vector states just described, in a manner similar to the mechanism described in Ref. 8. We illustrate the scattering process schematically, in Fig. 2, with the dispersion relation in Eq. (19) in mind. In this paper, we develop the theory of this two magnon scattering, with attention to specific models of matrix elements which control the scattering. As we shall see in Sec. III, and mentioned above, these scatterings introduce a frequency shift of the mode as well.

In earlier analyses of FMR linewidth data in ultrathin films, data at a small number of frequencies is fitted to a functional form which contains a term linear in frequency, and a zero-frequency residual linewidth. The linear term is assumed to be the contribution from damping of the spin motion within the body of the film, as described by the Landau-Lifshitz equation, though the slope is found often to be larger than that appropriate to the bulk material. The re-

sidual “zero-field” linewidth is argued to correlate with interface quality.¹ We shall see here that the mechanism we explore has a more complex variation with applied dc field, or FMR frequency.

The spin-wave dispersion relation just quoted is valid when the magnetization and \vec{H}_o both lie in the plane of the film, and are parallel to each other. If the magnetization is tipped out of the plane, the phase space within which the negative slope occurs decreases, and eventually $\Omega(\vec{k}_{\parallel})$ increases with \vec{k}_{\parallel} for all values of the wave vector. Then the mechanism examined here is rendered inoperative. This is noted in Ref. 10, and in fact in this paper, data are presented which show that the FMR linewidth indeed decreases markedly as \vec{M}_s is tipped out of the plane.

One may examine the nature of the spin-wave dispersion relation for the case where \vec{H}_o is applied out of the plane. Suppose \vec{H}_o makes the angle ϕ_H with the plane of the film. Then the magnetization will be canted out of plane, at the angle ϕ_M . Given ϕ_H , one finds ϕ_M by solving

$$\sin(\phi_H - \phi_M) = \frac{(H_s + 4\pi M_s)}{2H_o} \sin(2\phi_M). \quad (21)$$

The spin-wave dispersion relation then assumes the form

$$\begin{aligned} \Omega^2(\vec{k}_{\parallel}) = & \Omega_{FM}^2(\phi_H, \phi_M) - 2\pi\gamma^2 M_s k_{\parallel} d \\ & \times \{ [\cos^2 \phi_M - \sin^2 \phi_M \cos^2 \phi_{\vec{k}_{\parallel}}] [H_o \cos(\phi_H - \phi_M) \\ & - (H_s + 4\pi M_s) \sin^2 \phi_M] - \sin^2 \phi_{\vec{k}_{\parallel}} \\ & \times [H_o \cos(\phi_H - \phi_M) + (H_s + 4\pi M_s) \cos(2\phi_M)] \} \\ & + \gamma^2 D k_{\parallel}^2 \{ 2H_o \cos(\phi_H - \phi_M) + (H_s + 4\pi M_s) \\ & \times (1 - 3\sin^2 \phi_M) \}, \end{aligned} \quad (22)$$

where

$$\begin{aligned} \Omega_{FM}^2(\phi_H, \phi_M) = & \gamma^2 [H_o \cos(\phi_H - \phi_M) \\ & - (H_s + 4\pi M_s) \sin^2 \phi_M] [H_o \cos(\phi_H - \phi_M) \\ & + (H_s + 4\pi M_s) \cos(2\phi_M)]. \end{aligned} \quad (23)$$

Suppose H_o is applied perpendicular to the film, so $\phi_H = \pi/2$. There are then two cases to consider. If $H_o < (H_s + 4\pi M_s)$, we have $\phi_M < \pi/2$, i.e., the magnetization is canted at the angle $\phi_M^c = \sin^{-1}[H_o / (H_s + 4\pi M_s)]$, and the dispersion relation of spin-wave modes of Eq. (22) simplifies to

$$\begin{aligned} \Omega^2(\vec{k}_{\parallel}) = & \Omega_{FM}^2(\pi/2, \phi_M^c) + \gamma^2 (H_s + 4\pi M_s) \\ & \times (2\pi M_s k_{\parallel} d \sin^2 \phi_{\vec{k}_{\parallel}} + D k_{\parallel}^2) \left[1 - \left(\frac{H_o}{H_s + 4\pi M_s} \right)^2 \right]. \end{aligned} \quad (24)$$

If $H_o > (H_s + 4\pi M_s)$, then $\phi_M = \pi/2$, and the film is magnetized normal to its surface, and the dispersion relation becomes

$$\Omega^2(\vec{k}_{\parallel}) = \Omega_{FM}^2(\pi/2, \pi/2) + 2\gamma^2[H_o - (H_s + 4\pi M_s)] \times [\pi M_s k_{\parallel} d + Dk_{\parallel}^2]. \quad (25)$$

Notice that in both configurations corresponding to Eqs. (24) and (25) both terms linear in k_{\parallel} have positive coefficients, so there are no finite wave-vector modes degenerate with the ferromagnetic resonance mode. In what follows, our attention will be focused on the case where \vec{H}_o and \vec{M}_s are parallel to the film surfaces, and to each other.

III. DERIVATION OF EXPRESSIONS FOR LINEWIDTH AND FREQUENCY SHIFTS INDUCED BY SURFACE AND INTERFACE DEFECTS

In this section, our attention is confined to the case where \vec{H}_o and \vec{M}_s are both parallel to the film surface, and to each other. We shall treat the Hamiltonian in Eq. (14) as a quantum-mechanical, zero-order spin Hamiltonian, with $m_{x,y}(\vec{k}_{\parallel})$ regarded as an operator, and $m_{x,y}^*(\vec{k}_{\parallel})$ replaced by its Hermitian adjoint $m_{x,y}^+(\vec{k}_{\parallel})$. By adapting the continuum theory developed elsewhere¹¹ to the present problem, one deduces the commutation relation

$$[m_x(\vec{k}_{\parallel}), m_y^+(\vec{k}'_{\parallel})] = i\mu_o M_s \delta_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}}, \quad (26)$$

where μ_o is the magnetic moment of a magnetic ion in the film ($\mu_o = -\gamma\hbar$). The remaining operators commute among themselves.

Our approach will be to examine the equation of motion for the response functions

$$S_{\alpha\beta}(\vec{k}_{\parallel}, t) = i \frac{\theta(t)}{\hbar} \langle [m_{\alpha}(\vec{k}_{\parallel}, t), m_{\beta}^+(\vec{k}_{\parallel}, 0)] \rangle, \quad (27)$$

where the operators are in the Heisenberg representation, α and β range over x and y , and $\theta(t)$ is the Heaviside step function, equal to unity for $t > 0$ and zero when $t < 0$.

Of interest is the Fourier transform

$$S_{\alpha\beta}(\vec{k}_{\parallel}, \Omega) = \int_{-\infty}^{\infty} S_{\alpha\beta}(\vec{k}_{\parallel}, t) e^{i\Omega t} dt. \quad (28)$$

These functions, when considered as a function of frequency Ω , have poles when Ω equals the spin-wave frequencies in the system. In the presence of damping or scattering, the poles shift off the real axis. The imaginary part of the frequency is the linewidth, or inverse lifetime of the mode.

One begins with the equation of motion

$$i\hbar \frac{\partial}{\partial t} S_{\alpha\beta}(\vec{k}_{\parallel}, t) = \delta(t) \langle [m_{\beta}^+(\vec{k}_{\parallel}), m_{\alpha}(\vec{k}_{\parallel})] \rangle + i \frac{\theta(t)}{\hbar} \langle [[m_{\alpha}(\vec{k}_{\parallel}, t), H], m_{\beta}^+(\vec{k}_{\parallel}, 0)] \rangle. \quad (29)$$

A short calculation of the Fourier transforms defined in Eq. (28) for the perfect film described by the Hamiltonian in Eq. (14) provides the following expressions:

$$S_{xx}^{(0)}(\vec{k}_{\parallel}, \Omega) = \frac{\gamma^2 H_y(\vec{k}_{\parallel}) M_s}{\Omega^2(\vec{k}_{\parallel}) - \Omega^2}, \quad (30)$$

$$S_{yx}^{(0)}(\vec{k}_{\parallel}, \Omega) = \frac{i\gamma\Omega M_s}{\Omega^2(\vec{k}_{\parallel}) - \Omega^2}, \quad (31)$$

$$S_{xy}^{(0)}(\vec{k}_{\parallel}, \Omega) = -S_{yx}^{(0)}(\vec{k}_{\parallel}, \Omega), \quad (32)$$

and

$$S_{yy}^{(0)}(\vec{k}_{\parallel}, \Omega) = \frac{\gamma^2 H_x(\vec{k}_{\parallel}) M_s}{\Omega^2(\vec{k}_{\parallel}) - \Omega^2}. \quad (33)$$

Here $\Omega(\vec{k}_{\parallel})$ is the spin-wave dispersion relation given in Eqs. (17) and (19).

Our task is next to introduce the two magnon scattering terms into the Hamiltonian, and examine their influence on the structure of $S_{\alpha\beta}(\vec{k}_{\parallel}, \Omega)$. When this task is completed, we shall obtain expressions for both the lifetime and frequency shift produced by these processes.

In this section, we introduce the most general two magnon scattering term into the Hamiltonian, and then we analyze its contents. We begin with

$$V_2 = \frac{1}{2} \sum_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} m_x^+(\vec{k}'_{\parallel}) V_{xx}(\vec{k}'_{\parallel}, \vec{k}_{\parallel}) m_x(\vec{k}_{\parallel}) + \sum_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} m_x^+(\vec{k}'_{\parallel}) V_{xy}(\vec{k}'_{\parallel}, \vec{k}_{\parallel}) m_y(\vec{k}_{\parallel}) + \frac{1}{2} \sum_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} m_y^+(\vec{k}'_{\parallel}) V_{yy}(\vec{k}'_{\parallel}, \vec{k}_{\parallel}) m_y(\vec{k}_{\parallel}). \quad (34)$$

Section IV will be devoted to the construction of models for surface and interface defects. That discussion will lead to explicit forms for the matrix elements $V_{\alpha\beta}(\vec{k}'_{\parallel}, \vec{k}_{\parallel})$. For the moment, we address the issue of generating formal expressions for the two magnon contribution to the linewidth.

In the presence of such scattering, we need to examine response functions more general than those in Eq. (27). Thus we consider

$$S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; t) = i \frac{\theta(t)}{\hbar} \langle [m_{\alpha}(\vec{k}_{\parallel}, t), m_{\beta}^+(\vec{k}'_{\parallel}, 0)] \rangle. \quad (35)$$

When scattering processes such as those in Eq. (34) are introduced, wave vector is no longer conserved, so $S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; t)$ has nonzero off diagonal matrix elements in wave vector. We define the Fourier transform of $S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; t)$ as in Eq. (28).

We begin by generating equations of motion for these response functions. When we calculate $[dm_{\alpha}(\vec{k}_{\parallel}, t)/dt]$, we add phenomenological damping as described by the Landau-Lifshitz equation to the terms generated by the spin Hamiltonian.

We then find the following set of equations for S_{xx} and S_{yx} :

$$\begin{aligned}
& i\Omega S_{xx}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega) - \gamma \tilde{H}_y(\vec{k}_{\parallel}) S_{yx}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega) \\
&= \gamma M_s \sum_{\vec{k}''_{\parallel}} V_{xy}^*(\vec{k}''_{\parallel}, \vec{k}_{\parallel}) S_{xx}(\vec{k}''_{\parallel}, \vec{k}'_{\parallel}; \Omega) \\
&+ \gamma M_s \sum_{\vec{k}''_{\parallel}} V_{yy}^*(\vec{k}''_{\parallel}, \vec{k}_{\parallel}) S_{yx}(\vec{k}''_{\parallel}, \vec{k}'_{\parallel}; \Omega), \quad (36)
\end{aligned}$$

$$\begin{aligned}
& \gamma \tilde{H}_x(\vec{k}_{\parallel}) S_{xx}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega) + i\Omega S_{yx}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega) \\
&= \gamma M_s \delta_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} - \gamma M_s \sum_{\vec{k}''_{\parallel}} V_{xx}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) S_{xx}(\vec{k}''_{\parallel}, \vec{k}'_{\parallel}; \Omega) \\
&- \gamma M_s \sum_{\vec{k}''_{\parallel}} V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) S_{yx}(\vec{k}''_{\parallel}, \vec{k}'_{\parallel}; \Omega). \quad (37)
\end{aligned}$$

We have introduced

$$\gamma \tilde{H}_{x,y}(\vec{k}_{\parallel}) = \gamma H_{x,y}(\vec{k}_{\parallel}) - ig\Omega, \quad (38)$$

where $g \equiv G/\gamma M_s$ and G is the Gilbert damping constant, which appears in the Landau-Lifshitz equation of motion. To place Eqs. (36) and (37) in the form given, we have employed identities imposed on the matrix elements in Eq. (34) by the requirement that the Hamiltonian be Hermitian. For example, $V_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}) = V_{\alpha\beta}^*(-\vec{k}_{\parallel}, -\vec{k}'_{\parallel})$, and $V_{\alpha\alpha}(\vec{k}'_{\parallel}, \vec{k}_{\parallel}) = V_{\alpha\alpha}(-\vec{k}_{\parallel}, -\vec{k}'_{\parallel})$.

The two functions $S_{yy}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega)$ and $S_{xy}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega)$ obey equations rather similar in structure to Eqs. (36) and (37). In the interest of brevity, we omit displaying their explicit form.

We assume now that the defects responsible for the scattering are arranged on the surface in a random manner. We average all quantities over an ensemble of realizations of random defects. When this is done, the correlation function $S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega)$ becomes diagonal in wave vector. The averaging process restores translational invariance, on the average. If we denote this averaging process by angular brackets, then

$$\begin{aligned}
\langle S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega) \rangle &= \delta_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} \langle S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}_{\parallel}; \Omega) \rangle \\
&\equiv \delta_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} \bar{S}_{\alpha\beta}(\vec{k}_{\parallel}; \Omega). \quad (39)
\end{aligned}$$

For any particular film, we may write

$$S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega) = \delta_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} \bar{S}_{\alpha\beta}(\vec{k}_{\parallel}; \Omega) + \Delta S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega), \quad (40)$$

where $\Delta S_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}; \Omega)$ owes its existence to the specific arrangements of defects on the film under consideration.

In a related problem whose formal structure is very similar to that explored here, Huang and Maradudin¹² have utilized a projection operator method to obtain a closed equation for $\bar{S}_{\alpha\beta}(\vec{k}_{\parallel}, \Omega)$, in the limit where the amplitude of the roughness associated with the random defects may be assumed small. In essence, the averaged propagators obey a Dyson equation with a self-energy of matrix form. The self-energy matrix is quadratic in the two magnon matrix elements. In our case we find, after a straightforward application of this procedure, that the $\bar{S}_{\alpha\beta}(\vec{k}_{\parallel}, \Omega)$ obey

$$\begin{aligned}
& [i\Omega + \Sigma_{yx}(\vec{k}_{\parallel}, \Omega)] \bar{S}_{xx}(\vec{k}_{\parallel}, \Omega) + [-\gamma \tilde{H}_y(\vec{k}_{\parallel}) \\
&+ \Sigma_{yy}(\vec{k}_{\parallel}, \Omega)] \bar{S}_{yx}(\vec{k}_{\parallel}, \Omega) = 0, \quad (41)
\end{aligned}$$

and

$$\begin{aligned}
& [-\gamma \tilde{H}_x(\vec{k}_{\parallel}) + \Sigma_{xx}(\vec{k}_{\parallel}, \Omega)] \bar{S}_{xx}(\vec{k}_{\parallel}, \Omega) \\
&- [i\Omega - \Sigma_{xy}(\vec{k}_{\parallel}, \Omega)] \bar{S}_{yx}(\vec{k}_{\parallel}, \Omega) = -\gamma M_s, \quad (42)
\end{aligned}$$

where the matrix $\Sigma(\vec{k}_{\parallel}, \Omega)$ may be written in the form

$$\Sigma_{\alpha\beta}(\vec{k}_{\parallel}, \Omega) = \sum_{\vec{k}''_{\parallel}} \frac{\gamma^2 M_s^2}{[\gamma^2 \tilde{H}_x(\vec{k}''_{\parallel}) \tilde{H}_y(\vec{k}''_{\parallel}) - \Omega^2]} N_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}), \quad (43)$$

where

$$\begin{aligned}
N_{xx}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) &= \gamma \tilde{H}_y(\vec{k}''_{\parallel}) |V_{xx}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})|^2 + \gamma \tilde{H}_x(\vec{k}''_{\parallel}) |V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})|^2 \\
&- 2\Omega \text{Im}[V_{xx}^*(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})], \quad (44)
\end{aligned}$$

$$\begin{aligned}
N_{yy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) &= \gamma \tilde{H}_y(\vec{k}''_{\parallel}) |V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})|^2 + \gamma \tilde{H}_x(\vec{k}''_{\parallel}) |V_{yy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})|^2 \\
&- 2\Omega \text{Im}[V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) V_{yy}^*(\vec{k}_{\parallel}, \vec{k}''_{\parallel})], \quad (45)
\end{aligned}$$

$$\begin{aligned}
N_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) &= \gamma \tilde{H}_y(\vec{k}''_{\parallel}) V_{xx}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) \\
&+ \gamma \tilde{H}_x(\vec{k}''_{\parallel}) V_{yy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) \\
&- i\Omega \{V_{xx}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) V_{yy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) \\
&- V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) V_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})\}, \quad (46)
\end{aligned}$$

and

$$N_{yx}(\vec{k}_{\parallel}, \vec{k}''_{\parallel}) = N_{xy}(\vec{k}_{\parallel}, \vec{k}''_{\parallel})^*. \quad (47)$$

We have employed the identity, required by Hermiticity, $V_{\alpha\alpha}(\vec{k}_{\parallel}, \vec{k}'_{\parallel})^* = V_{\alpha\alpha}(\vec{k}'_{\parallel}, \vec{k}_{\parallel})$.

It is a simple matter to solve for $\bar{S}_{xx}(\vec{k}_{\parallel}, \Omega)$ and $\bar{S}_{yx}(\vec{k}_{\parallel}, \Omega)$ from Eqs. (41) and (42). Thus

$$\bar{S}_{xx}(\vec{k}_{\parallel}, \Omega) = \frac{\gamma M_s [\gamma \tilde{H}_y(\vec{k}_{\parallel}) - \Sigma_{yy}(\vec{k}_{\parallel}, \Omega)]}{[\gamma \tilde{H}_x(\vec{k}_{\parallel}) - \Sigma_{xx}(\vec{k}_{\parallel}, \Omega)] [\gamma \tilde{H}_y(\vec{k}_{\parallel}) - \Sigma_{yy}(\vec{k}_{\parallel}, \Omega)] + [i\Omega + \Sigma_{yx}(\vec{k}_{\parallel}, \Omega)] [i\Omega - \Sigma_{xy}(\vec{k}_{\parallel}, \Omega)]}. \quad (48)$$

We may generate expressions for \bar{S}_{yy} and \bar{S}_{xy} in a similar manner. Since the full form of these propagators is not required for what follows, we omit quoting their form in the interest of brevity.

We can simplify a number of features in Eq. (48). Since we assume the influence of the two magnon scattering on various quantities is small, save in the near vicinity of the spin-wave pole in the denominator, we may ignore $\Sigma_{yy}(\vec{k}_{\parallel}, \Omega)$ in the numerator. Also, we may replace $\tilde{H}_y(\vec{k}_{\parallel})$ in the numerator by simply $H_y(\vec{k}_{\parallel})$, setting aside the contribution from the damping.

Finally, our derivation of Eqs. (41) and (42) retains only terms in the equation of motion quadratic in the matrix elements $V_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel})$. Thus when we expand the factors in the denominator, we may discard products such as $\Sigma_{xx}(\vec{k}_{\parallel}, \Omega)\Sigma_{yy}(\vec{k}_{\parallel}, \Omega)$, which are fourth order. It is meaningful to retain such terms only if the individual self-energy matrix elements $\Sigma_{\alpha\beta}(\vec{k}_{\parallel}, \Omega)$ are each calculated complete through fourth order.

With these approximations, we may write

$$\bar{S}_{xx}(\vec{k}_{\parallel}, \Omega) = \frac{\gamma^2 H_y(\vec{k}_{\parallel}) M_s}{\Omega^2(\vec{k}_{\parallel}) - \Omega^2 - i\gamma g \Omega [H_x(\vec{k}_{\parallel}) + H_y(\vec{k}_{\parallel})] - \Sigma(\vec{k}_{\parallel}, \Omega)}, \quad (49)$$

where

$$\Sigma(\vec{k}_{\parallel}, \Omega) \equiv \sum_{\vec{k}''_{\parallel}} \frac{\gamma^2 M_s^2 N(\vec{k}_{\parallel}, \vec{k}''_{\parallel})}{\gamma^2 \tilde{H}_x(\vec{k}''_{\parallel}) \tilde{H}_y(\vec{k}''_{\parallel}) - \Omega^2}, \quad (50)$$

where $\Omega^2(\vec{k}_{\parallel})$ is the spin-wave dispersion relation defined in Eq. (19), and

$$N(\vec{k}_{\parallel}, \vec{k}'_{\parallel}) \equiv \gamma H_x(\vec{k}_{\parallel}) N_{yy}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}) + \gamma H_y(\vec{k}_{\parallel}) N_{xx}(\vec{k}_{\parallel}, \vec{k}'_{\parallel}) - 2\Omega \text{Im}[N_{xy}(\vec{k}_{\parallel}, \vec{k}'_{\parallel})]. \quad (51)$$

The real part of $\Sigma(\vec{k}_{\parallel}, \Omega)$ contains information about the two magnon scattering induced frequency shift of the spin wave, and the imaginary part their contribution to the linewidth. The expression for the quantity $N(\vec{k}_{\parallel}, \vec{k}'_{\parallel})$ is lengthy, and is derived readily from Eqs. (44)–(47).

Our interest is in the FMR mode, with wave vector $\vec{k}_{\parallel} = 0$. Thus we set $\vec{k}_{\parallel} = 0$ everywhere. Furthermore, in the calculation of $N(\vec{k}_{\parallel}, \vec{k}'_{\parallel})$, the factors of $\tilde{H}_x(\vec{k}_{\parallel})$, $\tilde{H}_y(\vec{k}_{\parallel})$ may be set to $H_x(0)$ and $H_y(0)$, since for the wave vector regime of interest, the finite wave-vector corrections are quantitatively small; they play a crucial role in the denominator of Eq. (50), of course, since they are responsible for rendering finite wave-vector spin waves degenerate with the FMR mode.

Then we have

$$\bar{S}_{xx}(\vec{k}_{\parallel} = 0, \Omega) = \frac{\gamma^2 [B_o + H_s] M_s}{\Omega_{FM}^2 - \Omega^2 - i\gamma g \Omega [B_o + H_s + H_o] - \Sigma(0, \Omega)}. \quad (52)$$

After considerable algebra, we find the numerator $N(0, \vec{k}''_{\parallel})$ in Eq. (50) may be expressed in terms of a positive definite quantity:

$$N(0, \vec{k}''_{\parallel}) = \gamma^2 [(B_o + H_s) V_{xx}(0, \vec{k}''_{\parallel}) + H_o V_{yy}(0, \vec{k}''_{\parallel}) + i[H_o(B_o + H_s)]^{1/2} [V_{xy}(0, \vec{k}''_{\parallel}) - V_{xy}^*(0, \vec{k}''_{\parallel})]]^2. \quad (53)$$

As noted above, the imaginary part of $\Sigma(0, \Omega)$ controls the two magnon contribution to the linewidth. The real part of $\Sigma(0, \Omega)$ provides us with the defect induced frequency shift of the resonance mode. If we keep only the imaginary part for the moment, and assume the Lorentzian which appears may be replaced by a suitably weighted Dirac delta function, then we have our final form for the spin-wave propagator:

$$\bar{S}_{xx}(\vec{k}_{\parallel} = 0, \Omega) = \frac{\gamma^2 [H_s + B_o] M_s}{\Omega_{FM}^2 - \Omega^2 - i\gamma g \Omega [B_o + H_s + H_o] - i\Gamma}, \quad (54)$$

with

$$\Gamma \equiv \frac{\pi \gamma^2 M_s^2}{2\Omega_{FM}} \sum_{\vec{k}''_{\parallel}} N(0, \vec{k}''_{\parallel}) \delta[\Omega(\vec{k}''_{\parallel}) - \Omega]. \quad (55)$$

This concludes our formal derivation of the two magnon scattering contribution to the linewidth. The influence of the frequency shift will be discussed below. Our next task, addressed in Sec. IV, is to make models of surface and interface defects, and their contribution to the matrix elements $V_{\alpha\beta}(\vec{k}_{\parallel}, \vec{k}'_{\parallel})$. This will lead us to find expressions for the linewidth, and frequency shift.

IV. MODEL OF SURFACE AND INTERFACE DEFECTS, AND THEIR CONTRIBUTION TO TWO MAGNON MATRIX ELEMENTS

In this section, we present a model of defects on the film surface, or at an interface, and examine their contribution to the matrix element for two magnon scattering. As noted earlier, in their very brief discussion, the authors of Ref. 10 provided no estimate of the magnitude of the matrix elements, and their relation to the morphology of specific surface features. Because of this, they gave no assessment, even qualitative, of the specific predictions of this mechanism. In our view, a central issue is the magnitude and field dependence of the two magnon scattering contributions. For this, explicit models are required.

We may expect island formation in the ultrathin films, and also depressions in the surface as well. We note that in a very interesting synchrotron study of interface roughness, Idzerda and his colleagues have quantified the length scale of the ‘‘magnetic roughness’’ present in ultrathin films.¹³

We assume that on the surface we have defects both in the form of bumps or islands, and also we have depressions or pits. The size of these structures will be supposed small compared to the wavelength of both spin waves involved in the two magnon scattering event. Thus when deriving their contribution to the matrix element, we may assume the spin-wave amplitudes are constant in their vicinity. We also as-

sume all bumps are identical, with the same orientation throughout the film, and similarly for pits. Of course, this will not be the case in practice. In the end, one may account for the influences of variations in size, topology, and orientation by replacing parameters which refer to the linear dimensions of a surface defect by suitable ensemble averages.

There are three contributions to the matrix element we consider: that from the perturbation to the Zeeman term, that from the perturbation to the dipolar energy, and finally that associated with the surface anisotropy. We consider each in turn.

A. Zeeman perturbation

As presented already in Eq. (10), the change in Zeeman energy due to the presence of a single bump is given by

$$\Delta H_z = \frac{H_o}{2M_s} \int_{V_b} dV [m_x^2(x, z) + m_y^2(x, z)], \quad (56)$$

with V_b the volume of the bump. Since the transverse magnetization associated with spin waves will not vary significantly over the region of the bump, if it is indeed small compared to the wavelength of the spin waves, the energy of a collection of bumps is approximated as

$$\Delta H_z^b = \sum_j \Delta H_z^j = \frac{H_o}{2M_s} V_b \sum_j [m_x^2(\vec{x}_j) + m_y^2(\vec{x}_j)], \quad (57)$$

where \vec{x}_j is the position of bump j . Using the representation of the magnetization in terms of Fourier components given in Eq. (2), one obtains

$$\begin{aligned} \Delta H_z^b = & \frac{H_o}{2M_s} \frac{V_b}{Ld} \sum_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} S^b(\vec{k}'_{\parallel} - \vec{k}_{\parallel}) [m_x^+(\vec{k}'_{\parallel}) m_x(\vec{k}_{\parallel}) \\ & + m_y^+(\vec{k}'_{\parallel}) m_y(\vec{k}_{\parallel})], \end{aligned} \quad (58)$$

where we have defined the following structure factor for the array of bumps

$$S^b(\vec{q}_{\parallel}) = \frac{1}{L} \sum_j e^{-i\vec{q}_{\parallel} \cdot \vec{x}_j}. \quad (59)$$

By similar arguments the change in the Zeeman energy due to ‘‘pits’’ or ellipsoidal depressions is

$$\begin{aligned} \Delta H_z^p = & -\frac{H_o}{2M_s} \frac{V_p}{Ld} \sum_{\vec{k}_{\parallel}, \vec{k}'_{\parallel}} S^p(\vec{k}'_{\parallel} - \vec{k}_{\parallel}) [m_x^+(\vec{k}'_{\parallel}) m_x(\vec{k}_{\parallel}) \\ & + m_y^+(\vec{k}'_{\parallel}) m_y(\vec{k}_{\parallel})], \end{aligned} \quad (60)$$

with $S^p(\vec{q}_{\parallel})$ an equivalent structure factor for pits.

B. Dipolar perturbation

First, we consider the change in the dipolar energy due to the presence of bumps on the surface of the film. We write the magnetization approximately as $\vec{M}(\vec{x}) = M_s \hat{z} + \vec{m}(\vec{x})$, with $\vec{m}(\vec{x}) = m_x(\vec{x}) \hat{x} + m_y(\vec{x}) \hat{y}$ the transverse component associated with the presence of the spin waves. Now we write $\vec{m}(\vec{x}) = \vec{m}_f(\vec{x}) + \sum_j \vec{m}_b^j(\vec{x})$, with $\vec{m}_b^j(\vec{x}) = 0$ outside the bump

(j) and $\vec{m}_f(\vec{x}) = 0$ outside the nominal film. Thus $\vec{m}_f(\vec{x})$ vanishes within the bump. The terms in the dipolar energy of the whole film of quadratic order in $\vec{m}(\vec{x})$ are then

$$\begin{aligned} H_D^b = & -\frac{1}{2} \int_V dV \vec{m} \cdot \vec{H}_D(\vec{m}) \\ = & -\frac{1}{2} \int_V dV \vec{m}_f \cdot \vec{H}_D(\vec{m}_f) \\ & -\frac{1}{2} \sum_j \int_{V_b^j} dV \vec{m}_b^j \cdot \vec{H}_D(\vec{m}_f) \\ & -\frac{1}{2} \sum_j \int_{V_f} dV \vec{m}_f \cdot \vec{H}_D(\vec{m}_b^j) \\ & -\frac{1}{2} \sum_i \sum_j \int_{V_b^i} dV \vec{m}_b^i \cdot \vec{H}_D(\vec{m}_b^j), \end{aligned} \quad (61)$$

with \vec{H}_D representing the demagnetizing fields of the different magnetization configurations considered. The first term quadratic in $\vec{m}_f(\vec{x})$ was already considered in the zeroth-order Hamiltonian appropriate to the perfectly smooth film. The second and third term are equal by virtue of the reciprocity theorem,¹⁴ and combined can be written as

$$\begin{aligned} H_D^{fb} = & -\sum_j \int_{V_b^j} dV \vec{m}_b^j \cdot \vec{H}_D(\vec{m}_f) \\ \simeq & -V_b \sum_j \vec{m}(\vec{x}_j) \cdot \vec{H}_D(\vec{m}_f; \vec{x}_j). \end{aligned} \quad (62)$$

The second term of Eq. (62) has been written with the approximation once again that the size of the bump is small with respect to the length scale of the variation of the spin wave field or $\vec{m}(\vec{x})$, i.e., the integral is easily approximated since the integrands are almost constant. The field appearing in Eq. (62) is evaluated just outside the film, at the position of the bump, and it is (to lowest order in $k_{\parallel}d$)

$$\begin{aligned} \vec{H}_D(\vec{m}_f; \vec{x}_j, y=d/2) = & \frac{1}{\sqrt{L^2d}} \sum_{k_{\parallel}} e^{i\vec{k}_{\parallel} \cdot \vec{x}_j} 2\pi(k_{\parallel}d) \\ & \times [m_y(\vec{k}_{\parallel}) - i\hat{k}_x^{\parallel} m_x(\vec{k}_{\parallel})] (\hat{y} - i\hat{k}_{\parallel}), \end{aligned} \quad (63)$$

i.e., the term H_D^{fb} will be of order $(k_{\parallel}d)$, therefore it will be neglected from the final analysis. We shall see, for example, that the final term in Eq. (61) dominates in the limit $k_{\parallel}d \ll 1$.

Now we turn to the fourth term in Eq. (61), for the case $i=j$, i.e., the ‘‘self-energy’’ terms:

$$H_D^{Sbj} = -\frac{1}{2} \int_{V_b^j} dV \vec{m}_b^j \cdot \vec{H}_D(\vec{m}_b^j). \quad (64)$$

In principle here one should calculate the energy due to a uniform magnetization within the bump. This is a difficult task, even for a structure of simple shape. We shall assume we may introduce appropriate demagnetizing factors N_x and

N_y for the structure. This will provide us with a contribution that is correct, to within factors of order unity. We shall appreciate later that this approximation will not influence our final result. Thus

$$H_D^{Sbj} = \frac{1}{2} V_b [N_x m_x^2(\vec{x}_j) + N_y m_y^2(\vec{x}_j)]. \quad (65)$$

Note that N_x and N_y are less than 4π ($N_x + N_y + N_z = 4\pi$).

Finally we address the terms which couple different bumps, i.e., the terms of the type

$$H_D^{ij} = - \int_{V_b^i} dV \vec{m}_b^i \cdot \vec{H}_D(\vec{m}_b^j). \quad (66)$$

The field of the bump (j) at the position of the bump (i) corresponds to that of a magnetic charge dipole within our approximation scheme [in the case of a semispherical bump of radius R the dipole is $\vec{p} = (2\pi/3)R^3\vec{m}(\vec{x}_j)$]. This field is weak in comparison with the self-field of bump (i), then it will be neglected.

Now we can summarize the results for bumps. The dominant dipolar contribution comes from terms like those of Eq. (65). Then, the main contribution of bumps to the change in dipolar energy is approximately

$$\Delta H_D^b = \frac{1}{2} V_b \sum_j [N_x m_x^2(\vec{x}^j) + N_y m_y^2(\vec{x}^j)]. \quad (67)$$

Using a Fourier representation of the quantities involved, the change in dipolar energy becomes

$$\begin{aligned} \Delta H_D^b = & \frac{1}{2} \frac{V_b}{Ld} \sum_{\vec{k}_\parallel, \vec{k}'_\parallel} S^b(\vec{k}'_\parallel - \vec{k}_\parallel) [N_x m_x^+(\vec{k}'_\parallel) m_x(\vec{k}_\parallel) \\ & + N_y m_y^+(\vec{k}'_\parallel) m_y(\vec{k}_\parallel)], \end{aligned} \quad (68)$$

where $S^b(\vec{k}'_\parallel - \vec{k}_\parallel)$ is the structure factor introduced above.

Now we consider the case of ‘‘pits’’ or depressions. These will prove to have a different contribution to the dipolar energy, as will be seen in the following.

First, the magnetization is written as

$$\vec{m}(\vec{x}) = \vec{m}_f(\vec{x}) + \sum_j \vec{m}_p^j(\vec{x}), \quad (69)$$

where $\vec{m}_f(\vec{x}) = 0$ outside the nominal film, and $\vec{m}_p^j(\vec{x}) = 0$ outside the pit, and also $\vec{m}_p^j(\vec{x}) = -\vec{m}_f(\vec{x})$ in the region of the pit, so that we have an empty region there. Notice that we have used superposition of magnetization configurations with overlap at the region of the pit.

Using our decomposition of $\vec{m}(\vec{x})$ one obtains an expression analogous to Eq. (61) for the dipolar energy quadratic in $\vec{m}(\vec{x})$. The interaction term becomes

$$H_D^{lp} = - \sum_j \int_{V_p^j} dV \vec{m}_p^j \cdot \vec{H}_D(\vec{m}_f) \approx V_p \sum_j \vec{m}(\vec{x}^j) \cdot \vec{H}_D(\vec{m}_f; \vec{x}_j). \quad (70)$$

Here the difference with the bumps, apart from the sign of \vec{m}_p , is that the demagnetizing field is evaluated inside the

film, which makes a large difference since the field inside the film is approximately $\vec{H}_D(\vec{m}_f; \vec{x}_j) \approx -4\pi m_y(\vec{x}_j)\hat{y}$, i.e.,

$$\vec{H}_D(\vec{m}_f; \vec{x}_j, y=d/2) \approx - \frac{1}{\sqrt{L^2 d}} \sum_{\vec{k}_\parallel} e^{i\vec{k}_\parallel \cdot \vec{x}_j} 4\pi m_y(\vec{k}_\parallel) \hat{y}. \quad (71)$$

Thus the interaction term for pits becomes approximately

$$H_D^{lp} \approx -4\pi \frac{V_p}{Ld} \sum_{\vec{k}_\parallel, \vec{k}'_\parallel} S^p(\vec{k}'_\parallel - \vec{k}_\parallel) m_y^+(\vec{k}'_\parallel) m_y(\vec{k}_\parallel), \quad (72)$$

with $S^p(\vec{q}_\parallel) \equiv (1/L) \sum_j e^{-i\vec{q}_\parallel \cdot \vec{x}_j}$ the structure factor of pits.

The treatment of the coupling terms between pits is similar as for bumps. Only the self-terms are important in the end, and it is easy to see that they are exactly the same as for bumps. Thus we approximate the self-terms by

$$\begin{aligned} \Delta H_D^{Sp} = & \frac{1}{2} \frac{V_p}{Ld} \sum_{\vec{k}_\parallel, \vec{k}'_\parallel} S^p(\vec{k}'_\parallel - \vec{k}_\parallel) [N_x m_x^+(\vec{k}'_\parallel) m_x(\vec{k}_\parallel) \\ & + N_y m_y^+(\vec{k}'_\parallel) m_y(\vec{k}_\parallel)]. \end{aligned} \quad (73)$$

Finally, adding all terms, the change in dipolar energy due to pits can be written as

$$\begin{aligned} \Delta H_D^p = & \frac{1}{2} \frac{V_p}{Ld} \sum_{\vec{k}_\parallel, \vec{k}'_\parallel} S^p(\vec{k}'_\parallel - \vec{k}_\parallel) [N_x m_x^+(\vec{k}'_\parallel) m_x(\vec{k}_\parallel) \\ & - (8\pi - N_y) m_y^+(\vec{k}'_\parallel) m_y(\vec{k}_\parallel)]. \end{aligned} \quad (74)$$

C. Magnetic surface anisotropy perturbation

In the ultrathin films of interest, surface anisotropy can be very strong. Variation in the direction of the anisotropy axis over the surface of a defect will lead to two magnon scattering, as emphasized by Heinrich.¹⁵ We consider this contribution here.

As far as surface magnetic anisotropy is concerned there is no difference between an ‘‘upward bump’’ or a ‘‘depression’’ of the same shape, so we will talk about defects, with both types in mind.

The change in surface anisotropy energy associated with the presence of a defect is given by

$$\Delta H_A = \frac{K_s}{M_s^2} \int_S dS [\vec{M}(\vec{x}) \cdot \hat{n}(\vec{x})]^2 - \frac{K_s}{M_s^2} \int_{\bar{S}} dS M_y^2(\vec{x}), \quad (75)$$

where $\hat{n}(\vec{x})$ is the normal to the surface of the defect at point \vec{x} , and \bar{S} is the projection of the surface S of the defect in the x - z plane.

Writing the magnetization as

$$\vec{M}(\vec{x}) = \vec{M}_{eq}(\vec{x}) \sqrt{1 - \left(\frac{\vec{m}(\vec{x})}{M_s} \right)^2} + \vec{m}(\vec{x}), \quad (76)$$

with $\vec{m}(\vec{x}) \perp \vec{M}_{eq}(\vec{x})$, it immediately satisfies the condition $\vec{M}^2(\vec{x}) = M_s^2$ if $\vec{M}_{eq}(\vec{x})^2 = M_s^2$, where $\vec{M}_{eq}(\vec{x})$ is the equilib-

rium magnetization configuration. In our case $\vec{M}_{eq}(\vec{x}) \simeq M_s \hat{z}$. Using Eq. (76), the terms to second order in $\vec{m}(\vec{x})$ in Eq. (75) become

$$\begin{aligned} \Delta H_A = & \frac{K_s}{M_s^2} \int_S dS [\hat{n}(\vec{x}) \cdot \hat{m}(\vec{x})]^2 - \frac{K_s}{M_s^2} \int_S dS \hat{n}_z^2(\vec{x}) \vec{m}^2(\vec{x}) \\ & - \frac{K_s}{M_s^2} \int_{\bar{S}} dS m_y^2(\vec{x}). \end{aligned} \quad (77)$$

Assuming the variation of $\vec{m}(\vec{x})$ is small within the region of the defect, then $\vec{m}(\vec{x})$ can be taken out of the integrals as $\vec{m}(\vec{x}_j)$, where \vec{x}_j is the position of the j th defect. Also, we assume the defect has symmetry such that $\int dS \hat{n}_x(\vec{x}) \hat{n}_y(\vec{x}) = 0$. Then this energy becomes

$$\Delta H_A^j = \frac{\bar{S}_d K_s}{M_s^2} \{ [f_x - f_z] m_x^2(\vec{x}_j) + [f_y - f_z - 1] m_y^2(\vec{x}_j) \}. \quad (78)$$

We have introduced the geometrical factors

$$f_\alpha = \frac{1}{\bar{S}_d} \int dS \hat{n}_\alpha^2, \quad (79)$$

where \bar{S}_d is the basal surface area of the defect, i.e., $f_x + f_y + f_z = S_d / \bar{S}_d$, with S_d the defect's surface area.

For what follows, it is important to note that if the topology of the defects is such that the x and z directions in the surface are equivalent, then $f_x = f_z$ and the term in $m_x^2(\vec{x}_j)$ vanishes. Thus if the defect is a hemisphere with circular footprint, or a square with sides or diagonals parallel to the x and z axes, we have no term in $m_x^2(\vec{x}_j)$. Thus a smooth bump should be elliptical, or an island should be a rectangle rather than a square for this term to be nonvanishing. This term will play a key role in controlling the field dependence of the two magnon contribution to the linewidth.

We now sum over the array of bumps and pits, and use the Fourier representation for the magnetization components to find

$$\begin{aligned} \Delta H_A = & \frac{\bar{S}_d K_s}{M_s^2 L d} \{ [f_x - f_z] m_x^+(\vec{k}'_\parallel) m_x(\vec{k}_\parallel) + [f_y - f_z - 1] \\ & \times m_y^+(\vec{k}'_\parallel) m_y(\vec{k}_\parallel) \} [S_b(\vec{k}'_\parallel - \vec{k}_\parallel) + S_p(\vec{k}'_\parallel - \vec{k}_\parallel)]. \end{aligned} \quad (80)$$

D. Summary of results

In this section we summarize the results obtained above. First, note that within the picture offered, the matrix element $V_{xy}(\vec{k}'_\parallel, \vec{k}_\parallel)$ vanishes. We are then left with $V_{xx}(\vec{k}'_\parallel, \vec{k}_\parallel)$ and $V_{yy}(\vec{k}'_\parallel, \vec{k}_\parallel)$ which we write, recalling that $H_s = 2K_s / (M_s d)$,

$$\begin{aligned} V_{xx}(\vec{k}'_\parallel, \vec{k}_\parallel) = & \frac{V_d}{M_s L d} \left\{ \left[N_x M_s + H_o + \frac{d}{h} (f_x - f_z) H_s \right] \right. \\ & \times S^b(\vec{k}'_\parallel - \vec{k}_\parallel) + \left[N_y M_s - H_o + \frac{d}{h} (f_x - f_z) H_s \right] \\ & \left. \times S^p(\vec{k}'_\parallel - \vec{k}_\parallel) \right\}, \end{aligned} \quad (81)$$

$$\begin{aligned} V_{yy}(\vec{k}'_\parallel, \vec{k}_\parallel) = & \frac{V_d}{M_s L d} \left\{ \left[N_x M_s + H_o + \frac{d}{h} (f_y - f_z - 1) H_s \right] \right. \\ & \times S^b(\vec{k}'_\parallel - \vec{k}_\parallel) - \left[(8\pi - N_y) M_s + H_o \right. \\ & \left. \left. - \frac{d}{h} (f_y - f_z - 1) H_s \right] S^p(\vec{k}'_\parallel - \vec{k}_\parallel) \right\}, \end{aligned} \quad (82)$$

where we have considered that bumps and pits have the same volume, i.e., $V_b = V_p = V_d$, and we have defined $h \equiv V_d / \bar{S}_d$, i.e., h is a measure of the height of the defects.

Consider the order of magnitude of the various contributions to Eqs. (81) and (82). Roughly speaking, M_s , H_o , and H_s are comparable in magnitude. For Fe, for instance, $4\pi M_s \simeq 21$ kG, and $|H_s|$ will be in the range of 10 kG in typical cases. Indeed, the fact that in zero applied field, one realizes ultrathin films with magnetization perpendicular to the surface shows $|H_s|$ can exceed $4\pi M_s$.

If all the fields are indeed comparable in magnitude, the factor of d/h which multiplies H_s in Eqs. (81) and (82) suggests that surface anisotropy provides the dominant contribution to the matrix element. If, for example, we have a film 20-Å thick, we may expect $d/h \sim 5$ to 10 for a typical defect.

Thus, in what follows, we retain only the surface anisotropy terms, to approximate V_{xx} and V_{yy} by the very simple forms

$$V_{xx}(\vec{k}'_\parallel, \vec{k}_\parallel) = \frac{\bar{S}_d H_s}{M_s L} (f_x - f_z) [S^b(\vec{k}'_\parallel - \vec{k}_\parallel) + S^p(\vec{k}'_\parallel - \vec{k}_\parallel)] \quad (83)$$

and

$$V_{yy}(\vec{k}'_\parallel, \vec{k}_\parallel) = \frac{\bar{S}_d H_s}{M_s L} (f_y - f_z - 1) [S^b(\vec{k}'_\parallel - \vec{k}_\parallel) + S^p(\vec{k}'_\parallel - \vec{k}_\parallel)]. \quad (84)$$

V. TWO MAGNON CONTRIBUTION TO LINEWIDTH AND FREQUENCY SHIFTS

A. Linewidth

Our first task is to evaluate the matrix element $N(0, \vec{k}''_\parallel)$ in Eq. (53). We do this utilizing the limiting forms given in Eqs. (83) and (84). We shall average the expression for $N(0, \vec{k}''_\parallel)$ over an array of randomly arranged defects. We assume no correlation between the location of pits and bumps, for simplicity. Thus if averages over configurations are denoted by angular brackets,

$$\langle |S^{b,p}(\vec{q}_{\parallel})|^2 \rangle = \frac{N_{b,p}}{L^2}, \quad (85)$$

and

$$\langle S^b(\vec{q}_{\parallel}) S^p(\vec{q}_{\parallel})^* \rangle = 0, \quad (86)$$

where N_b and N_p are the number of pits and bumps, on the surface area L^2 . In our approximation where the size of the surface defect is small compared to the magnon wavelength, $N(0, \vec{k}'')$ is independent of \vec{k}_{\parallel}'' . We thus drop reference to this, and we then find the simple result

$$N = \frac{2\gamma^2 \bar{S}_d H_s^2}{M_s^2 L^2} p [(B_o + H_s)(f_x - f_z) + H_o(f_y - f_z - 1)]^2, \quad (87)$$

where $p \equiv \bar{S}_d(N_b + N_p)/L^2$ is the fraction of the surface covered by defects. The factor of 2 has its origin in the fact that the film has an upper and a lower surface, each assumed to have a similar defect distribution.

Notice, as remarked earlier, if the defects are round or square in character, $f_x = f_z$ and V_{xx} vanishes. In this case, N is proportional to H_o^2 . In such a picture, the two magnon contribution to the linewidth will have a very strong dependence on the applied magnetic field H_o . In this case, only the component of magnetization associated with the spin waves which is normal to the surface provides coupling for the two magnon process. The ratio $(m_y/m_x)^2$ is equal to $H_o/(H_s + B_o)$ for these modes in the long-wavelength limit. The strong field dependence present when $V_{xx} = 0$ has its origin in this feature of the spin-wave modes in a thin film. So far as we know, there is little evidence for such a strong field dependence of the extrinsic linewidth. We argue, then, that on average, the footprint of the defects on the surface is either elongated or shortened, so $f_x \neq f_z$. Then $V_{xx} \neq 0$, and we realize the much more modest field dependence displayed in Eq. (87).

We now turn to Eq. (55), and we find an explicit expression for Γ . The frequency Ω in the Dirac delta function is replaced by the ferromagnetic resonance frequency Ω_{FM} . Then we have

$$\begin{aligned} \Gamma &= \frac{\gamma^2 H_s^2 p \bar{S}_d}{2\pi D(B_o + H_s + H_o)} [(B_o + H_s)(f_x - f_z) \\ &+ H_o(f_y - f_z - 1)]^2 \int_0^\infty dk_{\parallel} \int_0^{2\pi} d\phi_{k_{\parallel}} \delta \\ &\times [k_{\parallel} - k_{\parallel}^{(c)}(\phi_{k_{\parallel}})], \end{aligned} \quad (88)$$

where

$$k_{\parallel}^{(c)}(\phi_{k_{\parallel}}) \equiv \frac{2\pi M_s d [H_o - (B_o + H_s) \sin^2 \phi_{k_{\parallel}}]}{(B_o + H_s + H_o) D}. \quad (89)$$

For spin waves with propagation angle $\phi_{k_{\parallel}}$, $k_{\parallel}^{(c)}(\phi_{k_{\parallel}})$ is the wave vector of the finite wave-vector spin-wave modes degenerate with the ferromagnetic resonance mode.

The integrals in Eq. (88) are evaluated easily, to give

$$\begin{aligned} \Gamma &= \frac{2\gamma^2 H_s^2 p \bar{S}_d}{\pi D(B_o + H_s + H_o)} [(B_o + H_s)(f_x - f_z) \\ &+ H_o(f_y - f_z - 1)]^2 \sin^{-1} \left[\left(\frac{H_o}{B_o + H_s} \right)^{1/2} \right]. \end{aligned} \quad (90)$$

When we evaluated the integral over $\phi_{k_{\parallel}}$ in Eq. (88), we used that $H_o < B_o + H_s$: in our case this is assured since the main magnetization is assumed to lie in plane, i.e., $0 < 4\pi M_s + H_s$ or equivalently $H_o < B_o + H_s$.

Our subsequent discussion will be based on this form. Notice that as $H_o \rightarrow 0$, in fact $\Gamma \rightarrow 0$. That is, the two magnon scattering contribution to the linewidth vanishes at zero field. The reason is that as $H_o \rightarrow 0$, the phase space available to final state spin waves becomes vanishingly small, since one realizes the negative slope only for propagation directions for which $|\sin(\phi_{k_{\parallel}})| < [H_o/(B_o + H_s)]^{1/2}$. Thus if the mechanism explored here is operational in the sample studied, the term ‘‘zero-field linewidth’’^{1,2} used often is quite inappropriate.

We now need to link our quantity Γ with the actual ferromagnetic resonance linewidth, as measured experimentally. Consider the response function in Eq. (54). In the experiment, the frequency Ω is held fixed, and the dc field H_o is varied, and swept through resonance. In essence, the factor Ω_{FM}^2 in the denominator is swept through Ω by varying H_o . If $H_o = H_o^{(r)} + \Delta H$ where $H_o^{(r)}$ is the resonance field where $\Omega_{FM} = \Omega$, then when ΔH is small, $\Omega_{FM}^2 - \Omega^2 = \gamma^2(H_o + B_o + H_s)\Delta H$. Thus the linewidth ΔH in Gauss is given by

$$\Delta H = \frac{G\Omega_{FM}}{\gamma^2 M_s} + \Delta H^{(2)}, \quad (91)$$

where $\Delta H^{(2)} = \Gamma / [\gamma^2(H_o + B_o + H_s)]$, i.e., from Eq. (90),

$$\begin{aligned} \Delta H^{(2)} &= \frac{2H_s^2 p \bar{S}_d}{\pi D(B_o + H_s + H_o)^2} [(B_o + H_s)(f_x - f_z) \\ &+ H_o(f_y - f_z - 1)]^2 \sin^{-1} \left(\frac{H_o^{1/2}}{(B_o + H_s)^{1/2}} \right). \end{aligned} \quad (92)$$

We now need to resort to a specific model of the shape of the islands and pits, to evaluate f_x , f_y , and f_z . If we have a film made from material whose crystal structure is cubic, we may imagine the defects to be rectangles, with edges perpendicular to and parallel to the magnetization. We adopt this picture, and the dimensions of the model defect are illustrated in Fig. 3. The rectangle has height (or depth) b , and sides with length a and c , respectively. Then for such an island, we have $f_y = 1$, $f_x = 2b/a$, $f_z = 2b/c$, and $\bar{S}_d = ac$, and Eq. (92) becomes

$$\begin{aligned} \Delta H^{(2)} &= \frac{8H_s^2 b^2 pac}{\pi D(B_o + H_s + H_o)^2} \left[(B_o + H_s + H_o) \frac{1}{c} \right. \\ &\left. - (B_o + H_s) \frac{1}{a} \right]^2 \sin^{-1} \left(\frac{H_o^{1/2}}{(B_o + H_s)^{1/2}} \right). \end{aligned} \quad (93)$$

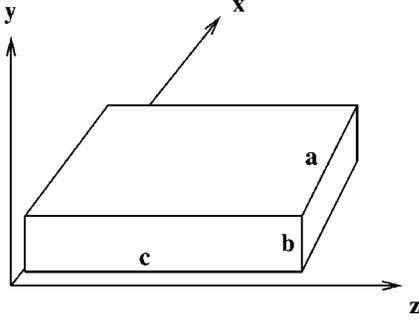


FIG. 3. Geometry of a rectangular defect, with sides a , b , and c in the x , y , and z directions, respectively.

The expression in Eq. (93) assumes all defects to be identical in size and topology. Our final step is to perform an average appropriate to an ensemble of defects, of diverse topology. That is, on a typical surface, in our picture, we may have some defects with $c > a$, and others with $c < a$. We may expect the ratio c/a to fluctuate about unity in value. Upon expanding the angular bracket in Eq. (93), and averaging over this aspect of the defect topology, we find our final form:

$$\begin{aligned} \Delta H_R^{(2)} = & \frac{8H_s^2 b^2 p}{\pi D (B_o + H_s + H_o)^2} \left[H_o^2 + (B_o + H_s + H_o)^2 \right. \\ & \times \left(\left\langle \frac{a}{c} \right\rangle - 1 \right) + (B_o + H_s)^2 \left(\left\langle \frac{c}{a} \right\rangle - 1 \right) \left. \right] \\ & \times \sin^{-1} \left(\frac{H_o^{1/2}}{(B_o + H_s)^{1/2}} \right). \end{aligned} \quad (94)$$

This expression can be further simplified by assuming that there is no anisotropy between both directions, i.e., $\langle a/c \rangle = \langle c/a \rangle$.

Clearly, we have made a number of approximations and model assumptions to reach our final form for the matrix element $N(0, \vec{k}_{\parallel}^{\prime\prime})$. These assumptions affect numerical prefactors which enter Eq. (94), but do not influence the dependence of $\Delta H^{(2)}$ on the film parameters, nor do these affect the magnitude of $\Delta H^{(2)}$ to any great extent. It is clear, for example, that if the surface defects are small in the sense described above, surface anisotropy is the dominant contribution to the matrix element.

B. Defect induced shift in resonance field

As noted in Sec. III, the two magnon scatterings lead not only an extrinsic contribution to the linewidth, but also lead to a shift in the resonance field. We see this in Eq. (49), where the shift in resonance field follows if the real part of $\Sigma(\vec{k}_{\parallel}, \Omega)$ is retained. When this is done, a discussion very similar to that given above provides the following expression for the shift $\Delta H_R^{(2)}$ for the resonance field:

$$\Delta H_R^{(2)} = \frac{1}{\gamma^2} \frac{\Sigma_R(0, \Omega_{FM})}{(B_o + H_s + H_o)}. \quad (95)$$

When the matrix element $N(0, \vec{k}_{\parallel}^{\prime\prime})$ is treated as in our discussion of the linewidth, with pits and islands of rectangular shape, we find

$$\begin{aligned} \Delta H_R^{(2)} = & \frac{8H_s^2 b^2 p a c}{\pi^2 (B_o + H_s + H_o)^2 D} \left[(B_o + H_s + H_o) \frac{1}{c} \right. \\ & \left. - (B_o + H_s) \frac{1}{a} \right]^2 \int_0^{\pi/2} d\phi_{\vec{k}_{\parallel}} \int_0^{k_{\parallel}^{(M)}} \frac{dk_{\parallel}}{k_{\parallel} - k_{\parallel}^{(c)}(\phi_{\vec{k}_{\parallel}})}, \end{aligned} \quad (96)$$

where $k_{\parallel}^{(c)}(\phi_{\vec{k}_{\parallel}})$ is defined in Eq. (89).

The integral in the magnitude of k_{\parallel} diverges logarithmically. We therefore introduce a maximum wave vector $k_{\parallel}^{(M)}$, whose nature is discussed below. The integrals in Eq. (96) may be evaluated analytically by first integrating on angle, and then on k_{\parallel} . We find

$$\begin{aligned} \Delta H_R^{(2)} = & \frac{8H_s^2 b^2 p a c}{\pi (B_o + H_s + H_o)^2 D} \left[(B_o + H_s + H_o) \frac{1}{c} \right. \\ & \left. - (B_o + H_s) \frac{1}{a} \right]^2 \left\{ \ln \left[\left(\frac{k_{\parallel}^{(M)}}{k_o} - \sin^2 \phi^{(c)} \right)^{1/2} \right. \right. \\ & \left. \left. + \left(\frac{k_{\parallel}^{(M)}}{k_o} + \cos^2 \phi^{(c)} \right)^{1/2} \right] \right\}. \end{aligned} \quad (97)$$

Here we have $\sin^2 \phi^{(c)} \equiv H_o / (B_o + H_s)$, and also we have defined $k_o \equiv 2\pi M_s d (B_o + H_s) / (B_o + H_s + H_o) D$.

Finally, if we average over the topology of the defects, as we did when we were led from Eq. (93) to Eq. (94), we have

$$\begin{aligned} \Delta H_R^{(2)} = & \frac{8H_s^2 b^2 p}{\pi (B_o + H_s + H_o)^2 D} \left[H_o^2 + (B_o + H_s + H_o)^2 \right. \\ & \times \left(\left\langle \frac{a}{c} \right\rangle - 1 \right) + (B_o + H_s)^2 \left(\left\langle \frac{c}{a} \right\rangle - 1 \right) \left. \right] \\ & \times \left\{ \ln \left[\left(\frac{k_{\parallel}^{(M)}}{k_o} - \sin^2 \phi^{(c)} \right)^{1/2} \right. \right. \\ & \left. \left. + \left(\frac{k_{\parallel}^{(M)}}{k_o} + \cos^2 \phi^{(c)} \right)^{1/2} \right] \right\}. \end{aligned} \quad (98)$$

The results displayed in Eqs. (94) and (98) are the final results of the paper. Our aim has been to obtain simple, analytic expressions for the two magnon contribution to the linewidth, and the frequency shift, to provide insight into the key physical features of the surface defects which control these extrinsic contributions.

What remains is to discuss the value of the cutoff wave vector $k_{\parallel}^{(M)}$ which appears in Eq. (98). We argue this is set by the average width, or transverse length scale of the surface defects. Upon averaging over an array of defects, we shall have $\langle a \rangle = \langle c \rangle$. Then $k_{\parallel}^{(M)} \approx 1/\langle a \rangle$. The reason for this choice is as follows. In our evaluation of $N(0, \vec{k}_{\parallel})$, we assume, in essence, $k_{\parallel} \langle a \rangle \ll 1$ for the spin waves involved in the final state of the two magnon process. From the numbers characteristic of the materials of interest, this is a reasonable

assumption, when the linewidth is analyzed. However, as we have seen, much shorter wavelengths enter the analysis of the frequency shift. A more complete account of the matrix element will show $N(0, \vec{k}_{\parallel})$ will fall off when $k_{\parallel} \langle a \rangle \gg 1$. This will ensure convergence of the integral. Since $k_{\parallel}^{(M)}$ appears only in the argument of a logarithm, a crude estimate of $k_{\parallel}^{(M)}$ will suffice.

VI. GENERAL COMMENTS AND DISCUSSION

We begin with a review of the manner in which ferromagnetic resonance linewidth data on ultrathin films have been analyzed in the literature.^{1,6,16}

In the experiments just cited, FMR experiments are performed with four frequencies at most, ranging from 9.5–73 GHz. The linewidth is found to increase with frequency, in a linear manner. Extrapolation of the data back to zero frequency yields a finite intercept referred to as the zero-field linewidth, $\Delta H(0)$. We note from Eq. (91) that the Gilbert damping term in the Landau-Lifshitz equation provides a contribution linear in frequency. In the data analysis, the constant G is determined from a fit to the slope of the linewidth ΔH as a function of the FMR resonance frequency. It is assumed that G determined by this means then provides a measure of damping present throughout the body of the film. It is found that G is very frequently larger in the ultrathin films than in bulk Fe.¹⁷ This is reasonable, of course, since the electronic structure of such films surely differs from bulk Fe. Finally $\Delta H(0)$ is argued to have its origin in surface defects.

The view just summarized is problematical, given our result in Eq. (94). Quite clearly, $\Delta H^{(2)}$ depends on the external magnetic field H_o , and hence on Ω_{FMR} . Thus, by fitting only a few data points, it is difficult to separate the bulk damping, surely linear in the frequency, from the extrinsic contribution with origin in surface defects.

We illustrate this with a numerical calculation appropriate to Fe. Here $4\pi M_s = 21$ kG, and $D = 2.5 \times 10^{-9}$ G cm². We have chosen H_s to be -15 kG, which means the normal to the surface is an easy axis. Note, by the way, that in general the H_s^2 in the prefactor of Eq. (94) need not coincide in value with the H_s appearing in the remaining factors in the expression. The former has its origin in low-symmetry sites on the sides of the model defects in our picture, while the latter in the anisotropy experienced by spins on the flat portions of the film surface. To calculate $\langle a/c \rangle$ and $\langle c/a \rangle$ we assume both a and c are randomly distributed from 0 to the value $N \times b$. Then

$$\left\langle \frac{c}{a} \right\rangle = \left\langle \frac{a}{c} \right\rangle = \frac{\int_0^{Nb} da \int_0^{Nb} dc \left(\frac{a}{c} \right)}{\int_0^{Nb} da \int_0^{Nb} dc} = \frac{(N+1)}{2(N-1)} \ln(N). \quad (99)$$

We let $N = 10$, suppose $b = 3 \text{ \AA}$, and $p = 0.3$. These choices provide the result for $\Delta H^{(2)}$ presented in Fig. 4. In the frequency range from 10–40 GHz, we see $\Delta H^{(2)}$ is roughly linear with $\nu_{FMR} = \Omega_{FMR}/(2\pi)$, though curvature is present clearly. If we approximate $\Delta H^{(2)}$ by a straight line in this

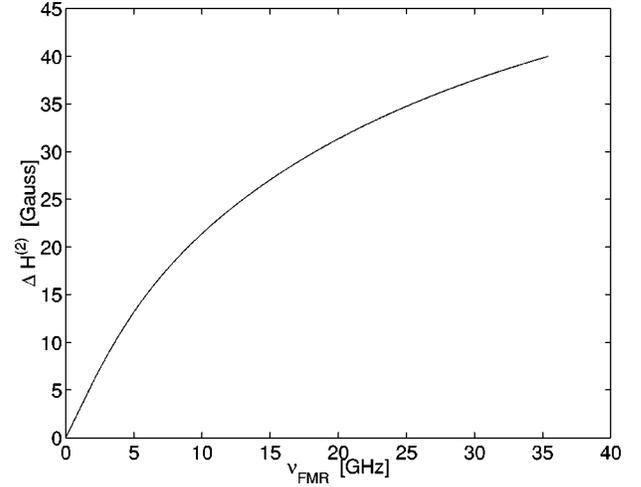


FIG. 4. Extrinsic resonance field linewidth $\Delta H^{(2)}$ as a function of resonant frequency $\nu_{FMR} = \Omega_{FMR}/(2\pi)$, for the following choice of parameters: $H_s = -15$ kG, $4\pi M_s = 21$ kG, $N = 10$ or $\langle a/c \rangle - 1 = 0.4$, $b = 3 \text{ \AA}$, and $p = 0.3$.

frequency window, and extrapolate to zero frequency, we obtain a “zero-field linewidth” of approximately 15 G. Of course, the actual extrinsic linewidth vanishes as the frequency goes to zero, for the physical reasons discussed earlier.

The slope of $\Delta H^{(2)}$ vs ν_{FMR} in Fig. 4 is very close to 1 G/GHz, if one approximates the curve by a straight line in the frequency region from 10–40 GHz. The slope provided by the bulk term in Eq. (91) is 1.13 G/GHz, if we assume $G = 0.8 \times 10^8 \text{ sec}^{-1}$ as in bulk Fe. If we combine the two mechanisms, we will find a slope of 2.13 G/GHz within this scheme. The two curves in Fig. 1 of Ref. 6 with largest slope have a slope of 2.5 G/GHz.

We could fit the actual experimental slope in the data just discussed by a slightly different choice of parameters, of course. In view of our schematic model of surface defects, this seems of little value. The point we wish to emphasize is the extrinsic, surface defect induced contribution to the linewidth depends on FMR frequency in a manner which obscures one’s ability to separate intrinsic and defect induced contributions to the linewidth.

As a consequence, it is difficult to make a clear separation between bulk damping of Gilbert form, and the surface defect induced linewidth. The dominant source of the frequency dependence in the extrinsic linewidth is the $\sin^{-1}[(H_o/(B_o + H_s))^{1/2}]$ factor in Eq. (92). This has its origin in the nature of the spin-wave dispersion curve as we have discussed, and is not influenced by details of the matrix element. In this sense, our predicted field dependence is robust.

We have seen that in addition to providing an extrinsic two magnon scattering to the linewidth, the presence of surface and interface defects lead to a frequency shift of the resonance field as well. In Fig. 5, we show the frequency variation of the defect induced shift in resonance field. The calculations are performed for the same model of the defects used to generate Fig. 4. We see that the shift in resonance field is quite appreciable, and shows little frequency depen-

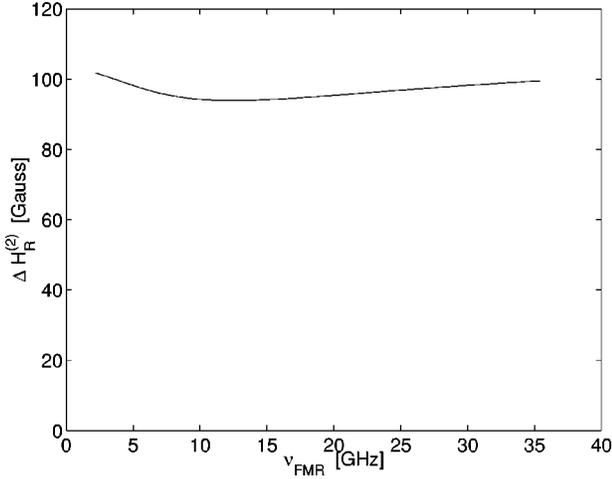


FIG. 5. Resonance field shift $\Delta H_R^{(2)}$ as a function of resonant frequency $\nu_{FMR} = \Omega_{FMR}/(2\pi)$, for the following choice of parameters: $H_s = -15$ kG, $4\pi M_s = 21$ kG, $N = 10$ or $\langle a/c \rangle - 1 = 0.4$, $b = 3$ Å, $p = 0.3$, and $d = 10$ Å.

dence. The presence of this shift will limit one's ability to extract precise values of material parameters from FMR resonance frequencies.

Of course, it will prove difficult to make a realistic model of surface and interface defects, and then follow through to the required matrix elements and a quantitatively reliable prediction of the resonance linewidth and the frequency shift. We propose the following procedure. Since our numerical calculations show the extrinsic contribution to the linewidth has a dependence on ν_{FMR} which is dominated by the $\sin^{-1}[(H_o/(B_o + H_s))^{1/2}]$ factor in Eq. (94), for the purposes of fitting data, one may employ the simple expression

$$\Delta H^{(2)} = \Gamma_{ex}(H_o) \sin^{-1} \left[\sqrt{\frac{H_o}{B_o + H_s}} \right], \quad (100)$$

where, in fact the field dependence [and hence the dependence of $\Gamma_{ex}(H_o)$ on ν_{FMR}] may be assumed weak and set aside. Unless the film is very thin indeed, we suggest that the bulk damping may be accounted for by using the Gilbert constant $G = 0.8 \times 10^8$ sec $^{-1}$, for bulk Fe. Given a value of $\Gamma_{ex}(H_o)$ for a particular film, the shift in resonance field is

$$\Delta H_R^{(2)} = \Gamma_{ex}(H_o) \left\{ \ln \left[\left(\frac{k_{\parallel}^{(M)}}{k_o} - \sin^2 \phi^{(c)} \right)^{1/2} + \left(\frac{k_{\parallel}^{(M)}}{k_o} + \cos^2 \phi^{(c)} \right)^{1/2} \right] \right\}. \quad (101)$$

To employ Eq. (101), one needs a value for $k_{\parallel}^{(M)}$. Since this enters only in the argument of the logarithm, a rough estimate of $\langle a \rangle$ and $\langle c \rangle$ should suffice for this purpose. Thus the parameter $\Gamma_{ex}(H_o)$ may be extracted from linewidth data without the need to resort to a specific model of the defect. With this parameter in hand, the shift in the resonance frequency induced by the defects may be estimated from Eq. (101).

We inquire about one further issue. This is the question of whether the defect induced modifications in the response function have a significant effect in the shape of the FMR

absorption line, or its integrated strength. We note that values for the magnetization M_s are inferred from the integrated strength of the FMR line,¹⁸ so a correction to the standard expression from two magnon scattering processes is potentially significant.

To address this issue, we need to explore a specific geometry for the resonance measurement. We suppose the sample is exposed to a plane polarized microwave field, applied parallel to the film surface, and to the x direction in Fig. 1. The rate at which energy is absorbed, $\alpha(H_o)$, is then proportional to $\text{Im}[\Omega \bar{S}_{xx}(0, \Omega)]$, where $\bar{S}_{xx}(\vec{k}_{\parallel}, \Omega)$ is given in Eq. (48). While the shift in resonance field and the linewidth are deduced from the structure of the denominator in Eq. (48), the factor of Σ_{yy} in the numerator contributes a two magnon scattering induced asymmetry in the lineshape, and corrections to the integrated strength of the line. In what follows, we write $\Sigma_{yy}(0, \Omega_{FM}) = \Sigma_{yy}^{(R)} + i\Sigma_{yy}^{(I)}$. After a brief calculation based on approximations similar to those used above, we find

$$\alpha(H_o) = \Omega_{FM} M_s \left(\frac{B_o + H_s - \Sigma_{yy}^{(R)}/\gamma}{B_o + H_s + H_o} \right) \times \left(\frac{\Delta H + \lambda(H_r - H_o)}{(\Delta H)^2 + (H_r - H_o)^2} \right). \quad (102)$$

In this expression, H_r is the resonance field, and ΔH the total linewidth given in Eq. (91). To lowest order in the roughness,

$$\lambda \equiv \frac{1}{\gamma(B_o + H_s)} \left(\frac{G\Omega_{FM}}{\gamma M_s} + \Sigma_{yy}^{(I)} \right). \quad (103)$$

Upon integrating $\alpha(H_o)$ over the absorption line, we find

$$\int \alpha(H_o) dH_o = \pi \Omega_{FM} M_s \left(\frac{B_o + H_s - \Sigma_{yy}^{(R)}/\gamma}{B_o + H_s + H_o} \right), \quad (104)$$

independent of the parameter λ .

The integrated strength of the absorption line, and hence one's ability to extract values for M_s from this in our model of the resonance experiment, is influenced by the factor of $\Sigma_{yy}^{(R)}/\gamma$ in Eq. (104). If we have Fe in mind, B_o is larger than 20 kG. The quantity $\Sigma_{yy}^{(R)}/\gamma$ is comparable in value to the shift in resonance field estimated above. Thus unless H_s is such that we are in the near proximity of the spin reorientation transition ($H_s \approx -B_o$), $\Sigma_{yy}^{(R)}/\gamma$ introduces a correction to the integrated strength of the line at the level of one percent, within the framework of the picture of the defects used above.

The parameter λ introduces deviation from Lorentzian shape in the FMR line, through the asymmetric term which reminds one of the Fano line shape. Note that $\lambda \neq 0$ even for a perfect film where $\Sigma_{yy} = 0$. Clearly, the presence of the two magnon scattering influences the value of λ , and hence the degree of asymmetry present. However, we expect λ to lie in the range of 10^{-2} or somewhat smaller. Thus, once again, within the framework of the model offered here, the effect is a very modest one.

We conclude by reminding the reader that the two magnon mechanism described and analyzed here is operative only when the magnetization is in plane. As we have seen in Sec. II, when the magnetization is normal to the film surfaces, there are no short-wavelength spin waves degenerate with the FMR mode. In Ref. 10, where the mechanism explored here is discussed briefly, as noted above, data is presented which shows that the linewidth indeed decreases markedly, as the magnetization is tipped out of plane.

Of interest would be the study of the in plane anisotropy of the FMR linewidth, for a film deposited on a stepped surface. Consider an ideal stepped surface, where the plateaus are perfectly flat, and the step edges straight, with no kinks present. The $k_{\parallel}=0$ FMR mode may then scatter only to short-wavelength final-state spin waves with wave vector perpendicular to the step edges. If the magnetization \vec{M}_s is parallel to the film surfaces, but perpendicular to the step edges, the final-state spin waves will have a negative coefficient for the dipole induced linear term in the dispersion relation. Thus for this geometry one will realize short-

wavelength modes degenerate with the FMR mode. If, however, \vec{M}_s is parallel to the step edges, the final-state spin waves will propagate perpendicular to the step edges. Thus there should be an in-plane anisotropy of the FMR linewidth for a film grown on stepped surfaces, with a minimum realized when \vec{M}_s is parallel to the step edges. Here the linear term is positive, there are no modes degenerate with the FMR mode, and the two magnon process will be quenched. Experimental studies of the in-plane anisotropy of the FMR linewidth on stepped surfaces will thus prove of interest.

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