Two magnon scattering in ultrathin ferromagnets: The case where the magnetization is out of plane

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(Received 17 December 2007; revised manuscript received 26 February 2008; published 3 June 2008)

We extend our earlier treatment of two magnon scattering in ultrathin ferromagnets to the case where the magnetization is tipped out of plane by virtue of the application of an out of plane magnetic field. The general formalism developed earlier is generalized in this regard, so when the magnetization is canted out of plane, one may extract information on the two magnon contribution to the linewidth or on the frequency shift of the ferromagnetic resonance line with origin in two magnon scattering. We provide the full set of response functions for such a film, which describe its response to driving fields of finite wave vector, so one may also explore two magnon effects on Brillouin light scattering if desired. We present explicit calculations that illustrate the behavior of the ferromagnetic resonance linewidth, for the picture employed earlier, where surface or interface defects activate two magnon scattering.

DOI: 10.1103/PhysRevB.77.214405 PACS number(s): 75.70.Ak, 75.30.Ds, 75.40.Gb

I. INTRODUCTION

In nanoscale magnetic structures, it is important to elucidate the mechanisms which control the damping of spin motions. In the current era, structures fabricated from the 3*d* transition metals and their alloys are of primary interest, since their ferromagnetism persists well above room temperature, even for very small samples such as ultrathin (few atomic layer) films. Thus, these materials are suitable for incorporation into devices such as the giant magnetoresistance read heads, whose impact on magnetic data storage has been stunning. The nature of the damping of spin motions realized in such structures controls their response time; it is of interest to know whether the mechanisms well known in the bulk forms of these materials are operative, and if there are new damping processes present that are unique to the nanoenvironment. The answer to both questions is in the affirmative.

The discussion in the present paper will be focused on the motion of the magnetization of ultrathin ferromagnetic films, under circumstances where the wavelengths involved are very long compared to the lattice constant. This is the domain probed by ferromagnetic resonance (FMR) spectroscopy and Brillouin light scattering (BLS). An introduction to both these techniques, and a summary of early data, is found in discussions presented by Heinrich[1] and by Cochran.[2] A more recent review of FMR studies of ultrathin structures is provided by Heinrich.[3] In bulk ferromagnets, the motion of the magnetization and its damping are well described by the Landau Lifschitz equation.[4] The strength of the damping term in this phenomenological description of magnetization motions is controlled by the Gilbert damping parameter G, whose origin lies in spin orbit coupling.[5]

In ultrathin films, it is now clear that processes not present in bulk materials can play a central role in the damping of spin motions. One of these is intrinsic in character for ultrathin ferromagnetic films adsorbed on metallic substrates and is often referred to in literature as the “spin pumping” contribution to the linewidth. After excitation of spin motions by a microwave field in FMR or laser excitation in BLS, the magnetic moments in the film precess coherently. These transfer spin angular momentum to the band electrons in the film, and the band electrons then transport the spin angular momentum to the substrate as they pass through the film/substrate interface. There is thus a net loss of spin angular momentum in the film per unit time, and this constitutes a damping mechanism. This mechanism was first discussed in classic papers by Berger[6] and Slonczewski.[7] The first experimental study of this mechanism was reported by Urban *et al.*[8] The work of Berger and Slonczewski was based on a simple, idealized local moment picture of the ferromagnetic film. Theoretical studies directed toward real materials have appeared subsequently.[9–11] It is of interest to note that it has been demonstrated[11] that the spin pumping mechanism studied in FMR is, in fact, the zero wave vector limit of the very strong damping observed in spin polarized electron loss studies[12] of large wave vector spin waves in ultrathin ferromagnets.

It is also the case that an extrinsic damping mechanism, referred to as two magnon damping, can play an important role in ultrathin ferromagnets. In a FMR experiment, the microwave field excites zero wave vector spin waves. The nature of the dispersion relation of spin waves in ultrathin ferromagnets with the magnetization in plane is such that there are spin waves of finite wave vector degenerate in frequency with the FMR mode.[13] In an ideal, perfect film, all spin wave modes are independent, decoupled normal modes of the system, so the FMR mode never “sees” or communicates with the finite wave vector modes of the same frequency. However, if defects with random spatial character are present, the defects scatter the zero wave vector FMR spin waves into the manifold of degenerate modes. This may be viewed as a dephasing contribution to the linewidth, in the language of spin resonance physics.

Two of us have developed a general formalism within which two magnon scattering and its influence may be studied,[14] for an ultrathin ferromagnet magnetized in plane. In the theory, we provide response functions that describe the response of the film to a finite wave vector applied field, including the influence of two magnon scattering, and from this we extracted expressions for the two magnon induced linewidth and frequency shift of the zero wave vector FMR mode. The general formalism applies to a variety of pictures of the defect structures. Shortly after the appearance of the theory, Azevedo *et al.*[15] reported data on the two magnon contribution to both the linewidth and frequency shift of the FMR mode in Permalloy films. The very interesting analysis in this paper shows that these two effects are linked very nicely, in a fully quantitative manner, by the theory in Ref. 14. The two magnon contribution to the linewidth is also strongly dependent on the wave vector of the spin wave of interest. That this is so has been demonstrated by comparing the linewidth of the FMR mode with that of spin waves excited in BLS, on the same sample.[16] Slebarski *et al.*[16] developed a theory of the wave vector dependence of the linewidth, along with a description of two magnon damping in ultrathin ferromagnets deposited on exchange biasing substrates. It is also the case that the picture set forth in Ref. 14 provides an excellent account of the frequency dependence of the FMR linewidth, on measurements which cover the very broad frequency range from 2 to 80 GHz.[17]. The defect picture introduced in Ref. 14 also leads to an excellent description of the in-plane anisotropy found for the two magnon linewidth in the data reported in Ref. 17.

Woltersdorf and Heinrich[18] discussed the data on samples where dislocation lines parallel to the film surfaces provide the activation mechanism for two magnon scattering in Fe films grown on Pd(100) surfaces. The phenomenological theory set forth in this paper provides a very good account of the data. The dislocation lines form a regular array to first approximation, with fourfold symmetry. Of course, disorder is necessary to initiate two magnon scattering. It would be of interest to construct a microscopic description of the matrix element that couples the FMR spin wave to its short wavelength degenerate partners for this interesting physical situation.

All of the papers above explore the case where the magnetization of the film is in plane. An interesting question is the effect on two magnon scattering of tipping the magnetization out of plane. In a brief note, McMichael *et al.*[19] presented data that demonstrated that as the magnetization is tipped out of plane, the two magnon scattering shuts off. These authors presented numerical calculations appropriate to their sample, which agree nicely with the data. In a recent review,[13] it was noted that if one examines the dispersion relation of dipole exchange spin waves for the case where the magnetization is tipped out of plane, spin waves degenerate with the FMR mode and disappear when the magnetization makes the angle of 45° with the film plane. This is in good agreement with the data presented in Ref. 19. Urban *et al.*[20] and Heinrich *et al.* also noted that the two magnon linewidth is not operative when the magnetization is perpendicular to the film surfaces. Similar observations on a very different system, a Heusler alloy grown on the InP(100) surface, are reported in Ref. 21. These authors provide data that show that the linewidth is less when the film is magnetized normal to its surface than when it is parallel.

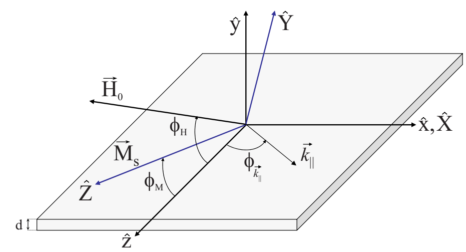

FIG. 1. (Color online) An illustration of the geometry used in the present paper. The externally applied magnetic field $\vec{H}_0$ and the magnetization $\vec{M}_s$ lie in the yz plane and make the angles indicated with respect to the z axis. The xz plane coincides with the surface of the film. The film is taken to lie between $y=-d/2$ and $y=d/2$ in the discussions in the text. See also the text for the definition of other quantities.

There has been no systematic discussion of the influence of tipping the magnetization out of plane on two magnon scattering in ultrathin ferromagnets, though as we have seen from the discussion above, various authors have appreciated that the mechanism is not operative when the magnetization is tipped out of plane. In our view, it is important to have a complete description of the phenomenon, including the out of plane geometry. In this paper, we extend the formalism developed in Ref. 13 to this case. While our numerical work focuses on the FMR mode of the film, our general theoretical development provides the reader with complete response functions for the film at finite wave vectors, including the influence of two magnon scatterings in their structure. Thus, if desired, with the response functions developed here, one can explore two magnon effects in BLS where finite wave vector spin waves are excited. In Sec. II, we present the theoretical development, and in Sec. III, we present numerical calculations of the frequency and angle dependence of the two magnon damping rate for the FMR mode. While it is, indeed, the case that the effect is cut off when the magnetization makes an angle greater than 45° with the plane, the results are striking in our view. Section IV is devoted to concluding remarks.

II. THEORETICAL DISCUSSION

The geometry we consider is displayed in Fig. 1. We have a thin ferromagnetic film of thickness d. The film surfaces are parallel to the xz plane, and the y axis is normal to the film surfaces. This is the coordinate system used in Ref. 13. An external magnetic field of strength H_0 is applied in the yz plane and, in our case, it makes the angle ϕ_H with respect to the z axis. The magnetization is canted out of plane to make an angle ϕ_M with respect to the z axis. We shall also make use of the XYZ coordinate system indicated, where the Z axis is aligned with the canted magnetization, and the X axis coincides with the x axis. We shall consider spin waves whose wave vector parallel to the film surfaces is $\vec{k}_\parallel$, and $\phi_{\vec{k}_\parallel}$ is the angle between $\vec{k}_\parallel$ and the z axis.

Our analysis applies to ultrathin ferromagnetic films, and in this limit, for all spin waves of concern to us, the limit $|\vec{k}_{\parallel}|d \ll 1$ is appropriate. We remark that the finite wave vector spin waves that enter the two magnon scattering theory have wave vectors in the regime $|\vec{k}_{\parallel}| \sim 10^5\ \mathrm{cm}^{-1}$,[13,14] so for films with thickness in the nanometer range, the inequality just stated is adequately fulfilled. In the thin film limit we employ, the spin wave eigenvectors are uniform across the film. So, following Ref. 14, our discussion will be phrased in terms of the transverse magnetization components

$$m_{X,Y}(x,z;t) = m_{X,Y}(\vec{r}_{\parallel},t) = \int_{-d/2}^{d/2} m_{X,Y}(\vec{r};t)\frac{dy}{d}, \tag{1}$$

where throughout the paper the subscript $\parallel$ refers to two-dimensional vectors in the xz plane.

We shall Fourier transform these amplitudes as follows:

$$m_{X,Y}(\vec{r}_{\parallel},t) = \frac{1}{\sqrt{L^2 d}}\sum_{\vec{k}_{\parallel}} m_{X,Y}(\vec{k}_{\parallel},t)\exp(i\vec{k}_{\parallel}\cdot\vec{r}_{\parallel}). \tag{2}$$

In Eq. (2), L^2 is the basic quantization area to which periodic boundary conditions are applied, in the xz plane.

We shall assume that we have a surface anisotropy of strength H_s present. The sign convention is then such that when $H_s > 0$, the normal to the film surfaces is a hard axis. Given the angle ϕ_H, it is straightforward to derive the equation from which ϕ_M is found by applying the zero torque condition. One then arrives at the implicit equation

$$\sin(\phi_H - \phi_M) = \frac{H_s + 4\pi M_s}{2H_0}\sin(2\phi_M). \tag{3}$$

The nature of the spin wave dispersion relation for the geometry illustrated in Fig. 1 plays a central role in understanding the results that we shall describe below. For the case where the magnetization is canted, as illustrated in Fig. 1, the dispersion relation in the ultrathin film limit was quoted without a derivation in Eqs. (22) and (23) of Ref. 14. To establish our notation, and to generate the structure of the spin wave Hamiltonian for the perfectly uniform film, we begin with the derivation of the spin wave dispersion relation for the canted state, in the ultrathin film limit. Then we turn our attention to the structure of the generalized susceptibilities introduced and discussed in Ref. 14, for the case where the magnetization is canted out of plane. We might remark that while our final goal is to arrive at an expression for the two magnon contribution to the linewidth, the response functions in their general form are useful for the analysis of diverse aspects of the response of imperfect films to applied microwave or laser fields, as noted above.

In what follows, in the interest of brevity, we shall refer the reader to discussions given previously in Ref. 14 at various points in the discussion below.

A. Spin wave dispersion relation for the case where the magnetization is canted out of plane

Our task is to expand the Hamiltonian of the system to second order in the deviation of the magnetization from equilibrium, when the system is disturbed. If we consider small amplitude motions of the magnetization, then we write the operators, in the Schrodinger representation, that describe the magnetization as

$$\vec{M}(\vec{r}_{\parallel}) = M_Z(\vec{r}_{\parallel})\hat{Z} + \vec{m}(\vec{r}_{\parallel}), \tag{4}$$

where $\vec{m}(r_{\parallel}) = \hat{X}m_X(\vec{r}_{\parallel}) + \hat{Y}m_Y(\vec{r}_{\parallel})$ and, if our attention is confined only to second order terms, $M_Z(\vec{r}_{\parallel}) = [M_s - m(\vec{r}_{\parallel})^2]/2M_s$. We may then write $\vec{M}(\vec{r}_{\parallel}) = \vec{M}^{(0)} + \vec{M}^{(1)}(\vec{r}_{\parallel}) + \vec{M}^{(2)}(\vec{r}_{\parallel}) + \cdots$, where

$$\vec{M}^{(0)} = M_s\hat{Z} = M_s(\hat{y}\sin\phi_M + \hat{z}\cos\phi_M), \tag{5a}$$

$$\vec{M}^{(1)}(\vec{r}_{\parallel}) = \vec{m}(\vec{r}_{\parallel}) = \hat{x}m_X(\vec{r}_{\parallel}) + \hat{y}m_Y(\vec{r}_{\parallel})\cos\phi_M - \hat{z}m_Y(\vec{r}_{\parallel})\sin\phi_M, \tag{5b}$$

and

$$\vec{M}^{(2)}(\vec{r}_{\parallel}) = -\hat{z}\frac{m(\vec{r}_{\parallel})^2}{2M_s} = -\hat{z}\frac{m_X(\vec{r}_{\parallel})^2 + m_Y(\vec{r}_{\parallel})^2}{2M_s} \times\{\hat{y}\sin\phi_M + \hat{z}\cos\phi_M\}. \tag{5c}$$

The contributions to the Hamiltonian are the Zeeman interaction with the external field, the interactions of the spins with the static dipolar field along with the dynamic dipolar field generated by the spin motion, the exchange, and, finally, the interaction with the surface anisotropy field. We discuss each in turn. In what follows, we ignore the terms in the Hamiltonian zero order in the deviation from equilibrium.

The Zeeman energy, as one sees from Fig. 1, is given by

$$H_Z = -H_0\sin(\phi_H - \phi_M)\int m_Y(\vec{r}_{\parallel})d^3r + \frac{H_0\cos(\phi_H - \phi_M)}{2M_s}\int m(\vec{r}_{\parallel})^2 d^3r. \tag{6}$$

The linear term in the spin deviation will be cancelled by a linear term in the dipolar interaction and a corresponding term in the surface anisotropy, both displayed below, once the equilibrium condition in Eq. (3) is noted. We thus discard the linear term in what follows. Then upon utilizing the Fourier representation in Eq. (2), we have

$$H_Z = \frac{H_0\cos(\phi_H - \phi_M)}{2M_s}\sum_{\vec{k}_{\parallel}}\{m_X^{\dagger}(\vec{k}_{\parallel})m_X(\vec{k}_{\parallel}) + m_Y^{\dagger}(\vec{k}_{\parallel})m_Y(\vec{k}_{\parallel})\}. \tag{7}$$

We used the relation $m_{X,Y}(-\vec{k}_{\parallel}) = m^{+}_{X,Y}(\vec{k}_{\parallel})$.

The dipolar field $\vec{h}_d(\vec{r})$ consists of a static component (zero order in the spin deviation) when the magnetization is canted, and a dynamic component generated by the motion of the spins. The dynamic component can be expanded in powers of the deviation of the magnetization from equilibrium in a manner similar to the magnetization. We require only the static zero order term $\vec{h}_d^{(0)}$ and the dynamic first order term $\vec{h}_d^{(1)}(\vec{r})$ to proceed. Then the dipolar contribution to the energy of the system, through second order in the spin deviation, has the form

$$H_d = -\frac{1}{2}\int \vec{M}(\vec{r}_\parallel)\cdot\vec{h}_d(\vec{r})d^3r = -\frac{1}{2}\int [\vec{M}^{(0)} + 2\vec{M}^{(1)}(\vec{r}_\parallel) + 2\vec{M}^{(2)}(\vec{r}_\parallel)]\cdot\vec{h}_d^{(0)}d^3r - \frac{1}{2}\int \vec{M}^{(1)}(\vec{r}_\parallel)\cdot\vec{h}_d^{(1)}(\vec{r})d^3r. \tag{8}$$

To obtain this form, we have invoked the reciprocity theorem, which leads to the identity $\int\vec{M}^{(1,2)}(\vec{r}_\parallel)\cdot\vec{h}_d^{(0)}d^3r = \int\vec{M}^{(0)}\cdot\vec{h}_d^{(1,2)}(\vec{r})d^3r$. Upon noting that $\vec{h}_d^{(0)} = -\hat{y}4\pi M_s\sin\phi_M$, by keeping only terms involving deviations of the magnetization from equilibrium, we have

$$H_d = 2\pi M_s\sin 2\phi_M\int m_Y(\vec{r}_\parallel)d^3r - 2\pi\sin^2\phi_M\int [m_X(\vec{r}_\parallel)^2 + m_Y(\vec{r}_\parallel)^2]d^3r - \frac{1}{2}\int \vec{M}^{(1)}(\vec{r}_\parallel)\cdot\vec{h}_d^{(1)}(\vec{r})d^3r. \tag{9}$$

The linear term in Eq. (9) will be cancelled in the end for the reasons discussed in the development of the expression for the Zeeman energy. The development of an expression for $\vec{h}_d^{(1)}(\vec{r})$ is a bit lengthy, so the discussion of this quantity is found in the Appendix, along with the evaluation of the last term in Eq. (9). When Eq. (A11) is combined with Eq. (9) and the linear term set aside, the dipolar contribution to the Hamiltonian may be written as

$$H_d = \frac{1}{2M_s}\sum_{\vec{k}_\parallel}\{2\pi M_s[k_\parallel d\sin^2\phi_{\vec{k}_\parallel} - 2\sin^2\phi_M]m_X^\dagger(\vec{k}_\parallel)m_X(\vec{k}_\parallel) + 2\pi M_s[2\cos 2\phi_M - k_\parallel d\zeta_{\vec{k}_\parallel}]m_Y^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel) + H_{XY}(\vec{k}_\parallel)\times[m_X^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel) + \text{H.c.}]\}. \tag{10}$$

We have defined

$$\zeta_{\vec{k}_\parallel} = \cos 2\phi_M + \sin^2\phi_M\sin^2\phi_{\vec{k}_\parallel} \tag{11a}$$

and

$$H_{XY}(\vec{k}_\parallel) = H_{YX}(\vec{k}_\parallel) = -\pi M_s k_\parallel d\sin\phi_M\sin 2\phi_{\vec{k}_\parallel}. \tag{11b}$$

The exchange energy can be written in the form

$$H_x = \frac{A}{M_s^2}\int [|\vec{\nabla}m_X(\vec{r})|^2 + |\vec{\nabla}m_Y(\vec{r})|^2]d^3r, \tag{12}$$

which, when expressed in terms of our operators, may be written as

$$H_x = \frac{1}{2M_s}\sum_{\vec{k}_\parallel}Dk_\parallel^2[m_X^\dagger(\vec{k}_\parallel)m_X(\vec{k}_\parallel) + m_Y^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel)], \tag{13}$$

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

We finally have the surface anisotropy term, which we write as

$$H_A = (K_s/M_s^2)\int M_y^2(\vec{r}_\parallel)dxdz. \tag{14}$$

If the term independent of the spin deviation from equilibrium is ignored, with $H_s=2K_s/M_sd$, this can be written as

$$H_A = \frac{H_s}{2}\sin 2\phi_M\int m_Y(\vec{r}_\parallel)d^3r + \frac{K_s}{M_s^2}\left[-\sin^2\phi_M\int m_X^2(\vec{r}_\parallel)dxdz + \cos(2\phi_M)\int m_Y^2(\vec{r}_\parallel)dxdz\right]. \tag{15}$$

Once again, the linear term cancels out when all linear terms are combined and the equilibrium condition in Eq. (1) is invoked, and if once again we introduce the surface anisotropy field $H_s=2K_s/M_sd$, then the quadratic terms in Eq. (15) become

$$H_A = \frac{-1}{2M_s}\sum_{\vec{k}_\parallel}[H_s\sin^2\phi_M m_X^\dagger(\vec{k}_\parallel)m_X(\vec{k}_\parallel) + H_s\cos 2\phi_M m_Y^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel)]. \tag{16}$$

When the four contributions to the spin Hamiltonian are combined, then we can collect them and to obtain the Hermitian quadratic form as

$$H = \frac{1}{2M_s}\sum_{\vec{k}_\parallel}\{H_X(\vec{k}_\parallel)m_X^\dagger(\vec{k}_\parallel)m_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)m_Y^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel) + H_{XY}(\vec{k}_\parallel)m_X^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel) + H_{YX}(\vec{k}_\parallel)m_Y^\dagger(\vec{k}_\parallel)m_X(\vec{k}_\parallel)\}, \tag{17}$$

where $H_{XY}(\vec{k}_\parallel)=H_{YX}(\vec{k}_\parallel)$ is defined in Eq. (11b) above, and

$$H_X(\vec{k}_\parallel) = H_X(0) + 2\pi M_s k_\parallel d\sin^2\phi_{\vec{k}_\parallel} + Dk_\parallel^2, \tag{18a}$$

$$H_Y(\vec{k}_\parallel) = H_Y(0) - 2\pi M_s k_\parallel d\zeta_{\vec{k}_\parallel} + Dk_\parallel^2, \tag{18b}$$

where

$$H_X(0) = H_0\cos(\phi_H - \phi_M) - (H_s + 4\pi M_s)\sin^2\phi_M \tag{18c}$$

and

$$H_Y(0) = H_0 \cos(\phi_H - \phi_M) + (H_s + 4\pi M_s)\cos(2\phi_M). \tag{18d}$$

For the case where the magnetization lies in plane, where $\phi_H=\phi_M=0$, the Hamiltonian in Eq. (17) reduces to Eq. (14) of Ref. 14.

One can proceed with a discussion of the spin dynamics through the use of the commutation relation stated in Eq. (26) of Ref. 14, which we state again here for completeness:

$$[m_X(\vec{k}_\parallel), m_Y^\dagger(\vec{k}_\parallel')] = i\mu_0 M_s \delta_{\vec{k}_\parallel,\vec{k}_\parallel'}, \tag{19}$$

where μ_0 is the magnetic moment in each unit cell of the material. All other operators commute. For instance, $[m_{X,Y}(\vec{k}_\parallel), m_{X,Y}(\vec{k}_\parallel')]=0$.

Through the use of the commutation relation in Eq. (19), one may readily derive equations of motion for the operators $m_{X,Y}(\vec{k}_\parallel)$ and, from these, the dispersion relation of spin waves in the uniform film is easily obtained. The result is

$$\Omega(\vec{k}_\parallel) = \gamma[H_X(\vec{k}_\parallel)H_Y(\vec{k}_\parallel) - H_{XY}^2(\vec{k}_\parallel)]^{1/2}. \tag{20a}$$

The gyromagnetic ratio is $\gamma=-\mu_0/\hbar$ (its absolute value). The ferromagnetic resonance frequency of the film is then

$$\Omega_{FMR} = \Omega(0) = \gamma[H_X(0)H_Y(0)]^{1/2}. \tag{20b}$$

The expression in Eq. (20b) is identical to Eq. (23) of Ref. 14, which was quoted without derivation. For long wavelengths, we can expand Eq. (20a) in powers of the wave vector $\vec{k}_\parallel$. If we retain only the two leading terms, for the square of the spin wave frequency, we have

$$\Omega^2(\vec{k}_\parallel) = \Omega_{FMR}^2 - 2\pi\gamma^2 M_s k_\parallel d[H_X(0)\zeta_{\vec{k}_\parallel} - H_Y(0)\sin^2\phi_{\vec{k}_\parallel}\} + \gamma^2 D k_\parallel^2[H_X(0)+H_Y(0)]. \tag{21}$$

It may be shown that Eq. (21) is identical to Eq. (22) of Ref. 14, which once again was quoted without derivation.

Of interest from the perspective of two magnon scattering are conditions wherein we have finite wave vector spin waves degenerate with the ferromagnetic resonance mode. It is under these conditions that two magnon scattering contributes to the ferromagnetic resonance linewidth. We have degenerate modes for conditions that we now state. The first requirement for this to be so, as we shall see in a moment, is that we have $\phi_M < \phi_M^{(c)} = \pi/4$, a condition that was stated and discussed earlier.[13] When this condition is satisfied, then for a selected range of propagation directions $\phi_{\vec{k}_\parallel}$, one realizes degenerate modes. There are two sets of such degenerate modes. The first lie in the angular region $-\phi^{(c)} < \phi_{\vec{k}_\parallel} < \phi^{(c)}$, and the second in the region $\pi-\phi^{(c)} < \phi_{\vec{k}_\parallel} < \pi+\phi^{(c)}$, where $\phi^{(c)}$ will be defined shortly. For each direction of propagation within these regimes, there is one such degenerate mode, as one sees from the structure of Eq. (21). The magnitude of the wave vector $k(\phi_{\vec{k}_\parallel})$ of these degenerate modes may be written in the form

$$k(\phi_{\vec{k}_\parallel}) = \tilde{k}[\sin^2(\phi^{(c)}) - \sin^2(\phi_{\vec{k}_\parallel})], \tag{22}$$

where

$$\sin^2(\phi^{(c)}) = \frac{H_X(0)}{H_X(0)+H_s+4\pi M_s}\frac{\cos(2\phi_M)}{\cos(\phi_M)} \tag{23a}$$

and

$$\tilde{k} = \frac{2\pi M_s d}{D}\left(\frac{H_X(0)+H_s+4\pi M_s}{H_X(0)+H_Y(0)}\right)\cos^2\phi_M. \tag{23b}$$

In order to have degenerate modes, the condition $k(\phi_{\vec{k}_\parallel})>0$ must be satisfied. Hence, when $\phi_M<\pi/4$, we have finite wave vector modes degenerate with the ferromagnetic resonance mode in the two angular regimes discussed earlier.

We now turn our attention to the response functions that describe the ultrathin ferromagnet when the magnetization is tipped out of plane, and also the effect of defects of various sorts on the structure of these response functions. In the end, after a suitable analysis of these response functions, we can obtain expressions for the two magnon contribution to the linewidth and the two magnon induced frequency shift.

B. Response functions of the film with canted magnetization

As in Ref. 14, we shall study the structure of the frequency and wave vector dependent susceptibilities of our film, and the influence of random defects on these functions. Then, in the next section, we extract expressions for the two magnon contribution to the FMR linewidth and also for the two magnon induced frequency shift.

We begin with the ideal, defect-free film as described by the Hamiltonian in Eq. (17). The functions we study are the set

$$S_{\alpha\beta}^{(0)}(\vec{k}_\parallel,t) = i\frac{\theta(t)}{\hbar}\langle[m_\alpha(\vec{k}_\parallel,t), m_\beta^\dagger(\vec{k}_\parallel,0\ ]\rangle. \tag{24}$$

Here, $m_\alpha(\vec{k}_\parallel,t)$ is the operator for the αth component of the dynamic magnetization in the Heisenberg representation of quantum mechanics. When the time argument is omitted, we refer to the operator in the Schrödinger representation.

It is actually the Fourier transform with respect to time that will be of interest:

$$S_{\alpha\beta}^{(0)}(\vec{k}_\parallel,\Omega) = \int_{-\infty}^{+\infty} e^{i\Omega t} S_{\alpha\beta}^{(0)}(\vec{k}_\parallel,t)dt. \tag{25}$$

The physical significance of $S_{\alpha\beta}^{(0)}(\vec{k}_\parallel,\Omega)$ is as follows. Suppose we expose the film to a microwave field with the space and time dependence $\vec{h}(\vec{r}_\parallel,t)=\hat{n}_\beta h^{(ext)}\exp[i\vec{k}_\parallel\cdot\vec{r}_\parallel - i\Omega t]$, with $\hat{n}_\beta$ a unit vector parallel to the coordinate axis β. This field will induce a time dependent component to the magnetization, which we may write $\vec{m}(\vec{r}_\parallel,t)=\vec{m}(\vec{k}_\parallel,\Omega)\exp[i\vec{k}_\parallel\cdot\vec{r}_\parallel - i\Omega t]$, assuming the external field is sufficiently weak that linear response theory is applicable. Then one may show that $m_\alpha(\vec{k}_\parallel,\Omega)=S_{\alpha\beta}^{(0)}(\vec{k}_\parallel,\Omega)h_\beta$. Thus, as stated above, $S_{\alpha\beta}^{(0)}(\vec{k}_\parallel,\Omega)$ is the wave vector and frequency dependent susceptibility of the film. When $\vec{k}_\parallel=0$, we have a description of the FMR response, and one may show[22] that the finite wave vector response can be used to describe the BLS spectrum of the film.

The equation of motion of the wave vector and frequency dependent susceptibility tensor is

$$i\hbar\frac{\partial}{\partial t}S^{(0)}_{\alpha\beta}(\vec{k}_{\parallel},t) = \delta(t)\langle[m^{\dagger}_{\beta}(\vec{k}_{\parallel}),m_{\alpha}(\vec{k}_{\parallel})]\rangle + i\frac{\theta(t)}{\hbar}\langle[[m_{\alpha}(\vec{k}_{\parallel},t),H],m^{\dagger}_{\beta}(\vec{k}_{\parallel},0)]\rangle. \quad (26)$$

It is a straightforward matter to use the commutators in Eq. (19) and the discussion just afterward to construct the response functions of the ideal film. As we proceed, we add damping as described by the Landau Lifschitz equation. Before we display the results, we introduce the quantities

$$\gamma\tilde{H}_{\alpha}(\vec{k}_{\parallel}) = \gamma H_{\alpha}(\vec{k}_{\parallel}) - ig\Omega, \quad (27a)$$

where $g=G/\gamma M_s$, with G the Gilbert damping parameter,

$$\Omega^{\pm}(\vec{k}_{\parallel}) = \Omega \pm i\gamma H_{XY}(\vec{k}_{\parallel}), \quad (27b)$$

and, finally,

$$\tilde{\Omega}(\vec{k}_{\parallel}) = \gamma[\tilde{H}_X(\vec{k}_{\parallel})\tilde{H}_Y(\vec{k}_{\parallel}) - H^2_{XY}(\vec{k}_{\parallel})]^{1/2}. \quad (27c)$$

We then have

$$S^{(0)}_{XX}(\vec{k}_{\parallel},\Omega) = \frac{\gamma^2 M_s\tilde{H}_Y(\vec{k}_{\parallel})}{\tilde{\Omega}^2(\vec{k}_{\parallel}) - \Omega^2}, \quad (28a)$$

$$S^{(0)}_{YX}(\vec{k}_{\parallel},\Omega) = i\frac{\gamma M_s\Omega^{+}(\vec{k}_{\parallel})}{\tilde{\Omega}^2(\vec{k}_{\parallel}) - \Omega^2}, \quad (28b)$$

$$S^{(0)}_{XY}(\vec{k}_{\parallel},\Omega) = -i\frac{\gamma M_s\Omega^{-}(\vec{k}_{\parallel})}{\tilde{\Omega}^2(\vec{k}_{\parallel}) - \Omega^2}, \quad (28c)$$

and

$$S^{(0)}_{YY}(\vec{k}_{\parallel},\Omega) = \frac{\gamma^2 M_s\tilde{H}_X(\vec{k}_{\parallel})}{\tilde{\Omega}^2(\vec{k}_{\parallel}) - \Omega^2}. \quad (28d)$$

We now introduce two magnon scattering, and obtain a description of its influence on the response functions displayed in Eqs. (28a)–(28d). The most general form for the two magnon term in the Hamiltonian that we can write down is

$$V_2 = \frac{1}{2}\sum_{\alpha,\beta=X,Y}\sum_{\vec{k}_{\parallel},\vec{k}'_{\parallel}} V_{\alpha\beta}(\vec{k}'_{\parallel},\vec{k}_{\parallel})m^{\dagger}_{\alpha}(\vec{k}'_{\parallel})m_{\beta}(\vec{k}_{\parallel}). \quad (29)$$

One can work out the form of the matrix elements $V_{\alpha\beta}(\vec{k}'_{\parallel},\vec{k}_{\parallel})$ for any desired model of the defects, which initiate two magnon scattering. As remarked in Sec. I, it is essential for randomness to be present, i.e., one must have matrix elements with nonzero terms when $\vec{k}'_{\parallel}\neq\vec{k}_{\parallel}$. We shall provide an explicit example for which the matrix elements are worked out in the discussion below. From the point of view of the general development of the theory, at this point, we keep the form of V_2 to be as general as possible.

For the form in Eq. (29) to be Hermitian, the following constraints on the matrix elements must hold:

$$V_{\alpha\alpha}(\vec{k}'_{\parallel},\vec{k}_{\parallel}) = V_{\alpha\alpha}(\vec{k}_{\parallel},\vec{k}'_{\parallel})^{*} = V_{\alpha\alpha}(-\vec{k}'_{\parallel},-\vec{k}_{\parallel})^{*} = V_{\alpha\alpha}(-\vec{k}_{\parallel},-\vec{k}'_{\parallel}) \quad (30a)$$

and

$$V_{\alpha\beta}(\vec{k}'_{\parallel},\vec{k}_{\parallel}) = V_{\alpha\beta}(-\vec{k}'_{\parallel},-\vec{k}_{\parallel})^{*} \quad \text{for } \alpha\neq\beta. \quad (30b)$$

In the presence of the defects, the film is no longer translationally invariant. We then must consider the equations of motion for the generalized response function

$$S_{\alpha\beta}(\vec{k}_{\parallel},\vec{k}'_{\parallel},t) = i\frac{\theta(t)}{\hbar}\langle[m_{\alpha}(\vec{k}_{\parallel},t),m^{\dagger}_{\beta}(\vec{k}'_{\parallel},0)]\rangle. \quad (31)$$

In the presence of static disorder, if an external field of wave vector $\vec{k}_{\parallel}$ and frequency Ω is applied to the system, the transverse magnetization excited by the field has components described by wave vectors $\vec{k}'_{\parallel}\neq\vec{k}_{\parallel}$. For this reason, we must consider the generalized set of response functions.

It is a straightforward matter to generate equations of motion for the response functions defined in Eq. (31), which include the terms introduced by the perturbation in Eq. (29). We then average over the random defect array to generate an equation of motion for the appropriately averaged response function. If we denote the ensemble averaged response functions (after Fourier transformation with respect to time) by adding angular brackets, we have $\langle S_{\alpha\beta}(\vec{k}_{\parallel},\vec{k}'_{\parallel})\rangle = \delta_{\vec{k}_{\parallel},\vec{k}'_{\parallel}}\bar{S}_{\alpha\beta}(\vec{k}_{\parallel},\Omega)$. In the equations of motion for the averaged functions $\bar{S}_{\alpha\beta}(\vec{k}_{\parallel},\Omega)$, we retain the first corrections from the presence of disorder, which are quadratic in the matrix elements $V_{\alpha\beta}(\vec{k}'_{\parallel},\vec{k}_{\parallel})$ introduced in Eq. (29). We proceed to generate the equations of motion through a procedure employed in a different context by Huang and Maradudin.[23] The application of this procedure to the present situation is discussed in detail in Ref. 14. We shall thus just quote the final equations of motion. In terms of quantities defined in Eqs. (27a)–(27c), we find these to be written as follows:

$$[i\Omega^{+}(\vec{k}_{\parallel}) + \Sigma_{YX}(\vec{k}_{\parallel},\Omega)]\bar{S}_{XX}(\vec{k}_{\parallel},\Omega) - [\gamma\tilde{H}_Y(\vec{k}_{\parallel}) - \Sigma_{YY}(\vec{k}_{\parallel},\Omega)]\bar{S}_{YX}(\vec{k}_{\parallel},\Omega) = 0, \quad (32a)$$

$$[\gamma\tilde{H}_X(\vec{k}_{\parallel}) - \Sigma_{XX}(\vec{k}_{\parallel},\Omega)]\bar{S}_{XX}(\vec{k}_{\parallel},\Omega) + [i\Omega^{-}(\vec{k}_{\parallel}) - \Sigma_{XY}(\vec{k}_{\parallel},\Omega)]\bar{S}_{YX}(\vec{k}_{\parallel},\Omega) = \gamma M_s, \quad (32b)$$

$$[i\Omega^{+}(\vec{k}_{\parallel}) + \Sigma_{YX}(\vec{k}_{\parallel},\Omega)]\bar{S}_{XY}(\vec{k}_{\parallel},\Omega) - [\gamma\tilde{H}_Y(\vec{k}_{\parallel}) - \Sigma_{YY}(\vec{k},\Omega)]S_{YY}(\vec{k}_{\parallel},\Omega) = -\gamma M_s \quad (32c)$$

$$[\gamma\tilde{H}_X(\vec{k}_{\parallel}) - \Sigma_{XX}(\vec{k}_{\parallel},\Omega)]\bar{S}_{XY}(\vec{k}_{\parallel},\Omega) + [i\Omega^{-}(\vec{k}_{\parallel}) - \Sigma_{XY}(\vec{k}_{\parallel},\Omega)]\bar{S}_{YY}(\vec{k}_{\parallel},\Omega) = 0. \quad (32d)$$

We have introduced self-energies in matrix form as given by

$$\Sigma_{\alpha\beta}(\vec{k}_{\parallel},\Omega) = \sum_{\vec{k}'_{\parallel}}\frac{\gamma^2 M_s^2}{\tilde{\Omega}(\vec{k}'_{\parallel})^2 - \Omega^2}N_{\alpha\beta}(\vec{k}_{\parallel},\vec{k}'_{\parallel}), \quad (33)$$

where

$$N_{XX}(\vec{k}_\parallel,\vec{k}'_\parallel) = \gamma\tilde{H}_Y(\vec{k}'_\parallel)|V_{XX}(\vec{k}_\parallel,\vec{k}'_\parallel)|^2 + \gamma\tilde{H}_X(\vec{k}'_\parallel)|V_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel)|^2 - 2\,\mathrm{Im}[\Omega^+(\vec{k}'_\parallel)V_{XX}(\vec{k}'_\parallel,\vec{k}_\parallel)\cdot V_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel)], \quad (34a)$$

$$N_{YY}(\vec{k}_\parallel,\vec{k}'_\parallel) = \gamma\tilde{H}_Y(\vec{k}'_\parallel)|V_{XY}(\vec{k}'_\parallel,\vec{k}_\parallel)|^2 + \gamma\tilde{H}_X(\vec{k}'_\parallel)|V_{YY}(\vec{k}'_\parallel,\vec{k}_\parallel)|^2 - 2\,\mathrm{Im}[\Omega^\dagger(\vec{k}'_\parallel)V_{XY}(\vec{k}'_\parallel,\vec{k}_\parallel)V_{YY}(\vec{k}'_\parallel,\vec{k}_\parallel)^*], \quad (34b)$$

$$\begin{aligned} N_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel) = {}& \gamma\tilde{H}_Y(\vec{k}'_\parallel)V_{XX}(\vec{k}_\parallel,\vec{k}'_\parallel)V_{XY}(\vec{k}'_\parallel,\vec{k}_\parallel) \\ &+ \gamma\tilde{H}_X(\vec{k}'_\parallel)V_{YY}(\vec{k}'_\parallel,\vec{k}_\parallel)V_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel) \\ &- i\Omega^-(\vec{k}'_\parallel)V_{XX}(\vec{k}_\parallel,\vec{k}'_\parallel)V_{YY}(\vec{k}'_\parallel,\vec{k}_\parallel) \\ &+ i\Omega^+(\vec{k}'_\parallel)V_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel)V_{XY}(\vec{k}'_\parallel,\vec{k}_\parallel), \end{aligned} \quad (34c)$$

and

$$N_{YX}(\vec{k}_\parallel,\vec{k}'_\parallel) = N_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel)^*. \quad (34d)$$

For our purposes, it will prove sufficient to display explicitly only the function $\bar{S}_{XX}(\vec{k}_\parallel,\Omega)$, which has the form, suppressing reference to the wave vector and frequency dependence of the quantities which enter,

$$\bar{S}_{XX} = \frac{\gamma M_s[\gamma\tilde{H}_Y - \Sigma_{YY}]}{[\gamma\tilde{H}_X - \Sigma_{XX}][\gamma\tilde{H}_Y - \Sigma_{YY}] + [i\Omega^+ + \Sigma_{YX}][i\Omega^- - \Sigma_{XY}]}. \quad (35)$$

The remaining response functions are readily obtained from Eqs. (32a)–(32d) if desired.

We will apply the theory to ultrathin films in which the two magnon corrections may be viewed as modest in magnitude. The linewidths in the samples of interest are small compared to the resonance frequency. Thus, the constant $g = G/\gamma M_s$, which enters Eq. (27a), is small compared to unity ($\sim 10^{-2}$ typically), and the corrections provided by the self-energy terms $\Sigma_{\alpha\beta}$ in Eq. (35) are roughly of the same order of magnitude in typical films of interest to us. Thus, we may simplify Eq. (35) by ignoring the small term Σ_{yy} in the numerator, and for most purposes, we may set aside the factor of $ig\Omega$ in the numerator as well. In the denominator, we may also keep only the leading terms in the self-energy and the Landau Lifschitz damping. When these approximations suffice, Eq. (35) simplifies to the form

$$\bar{S}_{XX}(\vec{k}_\parallel,\Omega) = \frac{\gamma^2 M_s H_Y(\vec{k}_\parallel)}{\Omega(\vec{k}_\parallel)^2 - \Omega^2 - ig\Omega\gamma[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)] - \Sigma(\vec{k}_\parallel,\Omega)}, \quad (36)$$

where

$$\Sigma(\vec{k}_\parallel,\Omega) = \sum_{\vec{k}'_\parallel} \frac{\gamma^2 M_s^2}{\tilde{\Omega}(\vec{k}'_\parallel)^2 - \Omega^2} N(\vec{k}_\parallel,\vec{k}'_\parallel) \quad (37a)$$

and

$$N(\vec{k}_\parallel,\vec{k}'_\parallel) = \gamma\tilde{H}_X(\vec{k}_\parallel)N_{YY}(\vec{k}_\parallel,\vec{k}'_\parallel) + \gamma\tilde{H}_Y(\vec{k}_\parallel)N_{XX}(\vec{k}_\parallel,\vec{k}'_\parallel) - 2\,\mathrm{Im}[\Omega^+(\vec{k}_\parallel)N_{XY}(\vec{k}_\parallel,\vec{k}'_\parallel)]. \quad (37b)$$

The term in $ig\Omega$ describes the influence of intrinsic damping, as described by the Landau Lifschitz equation. With the self-energy $\Sigma(\vec{k}_\parallel,\Omega)$ set aside, the expression in Eq. (36) extends the often quoted description of the ferromagnetic resonance ($\vec{k}_\parallel=0$) response of the film to finite wave vectors. Of course, when the definitions above are utilized, our results apply not only to the in-plane magnetized film, but to the case where the magnetization is canted out of plane as well. The remaining response functions are readily obtained from Eqs. (32a)–(32d) above, if desired. The present authors are unaware of any publication that presents the response functions for the perfect, defect-free ultrathin ferromagnet at finite wave vector, for the case where the magnetization is out of plane, as remarked earlier. It is our view that these functions should prove useful for a variety of applications.

The influence of the two magnon scattering is contained in the self-energy term $\Sigma(\vec{k}_\parallel,\Omega)$ in Eq. (36). To obtain a description of these effects to lowest order in the strength of the two magnon scattering, one replaces $\Sigma(\vec{k}_\parallel,\Omega)$ by $\Sigma(\vec{k}_\parallel,\Omega(\vec{k}_\parallel))$. The imaginary part of the self-energy then describes the effect of two magnon scattering on the damping of the mode of wave vector $\vec{k}_\parallel$, and the real part contains information on the two magnon contribution to the frequency shift of the mode.

This completes the extension of the formalism developed in Ref. 14 to the case where the magnetization of the film does not lie in the plane but has an out of plane component. In the next section, we turn to the development of a description of two magnon scattering effects for a particular picture of a possible defect structure in the film.

C. Two magnon induced linewidth and frequency shift of the ferromagnetic resonance mode

The formalism developed above gives us the complete response functions of the film at finite wave vector, including the influence of two magnon scattering in these objects. Since the FMR mode is the uniform mode of the film with $\vec{k}_\parallel=0$, we begin with

$$\bar{S}_{XX}(0,\Omega) = \frac{\gamma^2 M_s H_Y(0)}{\Omega_{FMR}^2 - \Omega^2 - ig\Omega\gamma[H_X(0) + H_Y(0)] - \Sigma(0,\Omega)}. \quad (38)$$

Here, $\Omega_{FMR} = \gamma[H_X(0)H_Y(0)]^{1/2}$ is the ferromagnetic resonance frequency. After considerable algebra, one may show that the quantity $N(0,\vec{k}_\parallel)$ in the numerator of $\Sigma(0,\Omega)$ is positive definite and can be written as

$$N(0,\vec{k}_\parallel) = \gamma^2|H_Y(0)V_{XX}(0,\vec{k}_\parallel) + H_X(0)V_{YY}(0,\vec{k}_\parallel) + i\sqrt{H_X(0)H_Y(0)}\{V_{XY}(0,\vec{k}_\parallel) - V_{XY}(\vec{k}_\parallel,0)^*\}|^2. \quad (39)$$

The self-energy at zero wave vector may then be cast into the form

$$\Sigma(0,\Omega) = \gamma^2 M_s^2 \sum_{\vec{k}_\parallel} \frac{N(0,\vec{k}_\parallel)\{\Omega^2(\vec{k}_\parallel) - \Omega^2 + ig\Omega\gamma[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)]\}}{\{\Omega^2(\vec{k}_\parallel) - \Omega^2\}^2 + (g\Omega\gamma)^2[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)]^2}. \quad (40)$$

We identify the two magnon contribution to the damping of the FMR mode, $\Gamma_{FMR}^{(2)}$, with $\mathrm{Im}\{\Sigma(0,\Omega_{FMR})\}$. Thus, we have

$$\Gamma_{FMR}^{(2)} = \gamma^2 M_s^2 \sum_{\vec{k}_\parallel} \frac{N(0,\vec{k}_\parallel) g\Omega_{FMR}\gamma[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)]}{[\Omega^2(\vec{k}_\parallel) - \Omega_{FMR}^2]^2 + (g\Omega\gamma)^2[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)]^2}. \quad (41)$$

In the limit that the damping of the finite wave vector modes is small, we make the replacement

$$\frac{g\Omega_{FMR}\gamma[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)]}{[\Omega^2(\vec{k}_\parallel) - \Omega_{FMR}^2]^2 + (g\Omega\gamma)^2[H_X(\vec{k}_\parallel) + H_Y(\vec{k}_\parallel)]^2} \to \pi\delta(\Omega^2(\vec{k}_\parallel) - \Omega_{FMR}^2) \quad (42)$$

so we have the rather simple expression

$$\Gamma_{FMR}^{(2)} = \pi(\gamma M_s)^2 \sum_{\vec{k}_\parallel} N(0,\vec{k}_\parallel)\delta(\Omega^2(\vec{k}_\parallel) - \Omega_{FMR}^2). \quad (43)$$

For the purposes of the calculations here, the dispersion relation given in Eq. (21) may be used to evaluate the integral in Eq. (43). Notice then that we have the relation

$$\Omega^2(\vec{k}_\parallel) - \Omega_{FMR}^2 = \gamma^2 D[H_X(0) + H_Y(0)]k_\parallel\{k_\parallel - k(\phi_{\vec{k}_\parallel})\} \quad (44)$$

so the Eq. (43) becomes

$$\Gamma_{FMR}^{(2)} = \frac{\pi M_s^2}{D[H_X(0) + H_Y(0)]} \sum_{\vec{k}_\parallel} \frac{N(0,\vec{k}_\parallel)}{k(\phi_{\vec{k}_\parallel})} \delta(k_\parallel - k(\phi_{\vec{k}_\parallel})). \quad (45)$$

The two magnon induced frequency shift of the FMR mode is given by $\delta\Omega_{FMR}^{(2)} = -\mathrm{Re}\{\Sigma(0,\Omega_{FMR})\}/2\Omega_{FMR}$. In the limit that the damping of the finite wave vector modes is small, we have

$$\delta\Omega_{FMR}^{(2)} = \frac{\gamma^2 M_s^2}{2\Omega_{FMR}} \sum_{\vec{k}_\parallel} \frac{N(0,\vec{k}_\parallel)}{\Omega_{FMR}^2 - \Omega(\vec{k}_\parallel)^2}, \quad (46)$$

which, upon using Eq. (44), becomes

$$\delta\Omega_{FMR}^{(2)} = \frac{M_s^2}{2\Omega_{FMR} D[H_X(0) + H_Y(0)]} \sum_{\vec{k}_\parallel} \frac{N(0,\vec{k}_\parallel)}{k_\parallel[k(\phi_{\vec{k}_\parallel}) - k_\parallel]}. \quad (47)$$

The expressions in Eqs. (45) and (47) may be used to assess the two magnon induced damping and two magnon induced frequency shift of the FMR mode, for any assumed defect geometry, provided the assumptions stated in their derivation are satisfied. These assumptions should be satisfied in nearly all ultrathin ferromagnets. To proceed further, we need to resort to a specific model of the defects which initiate the two magnon scattering. We turn to this in the next section.

D. Model for surface and interface defects and explicit expressions for the two magnon induced damping and frequency shift

To proceed, we require an explicit form for the matrix element $N(\vec{k}_\parallel,\vec{k}_\parallel')$, which appears in the formulas above. This requires us to model the possible character of defects that activate the two magnon scattering. Of course, the defect structure realized in various samples may differ considerably, so it is difficult to envision a universal structure for the matrix element. In this section, we extend the particular picture set forth in Ref. 14 to the case where the magnetization is canted out of plane. We note that this picture leads to a structure which accounts nicely for the data on both the two magnon induced linewidth and frequency shift reported for an in plane magnetized sample in Ref. 15.

We suppose that the defects consist of well defined and separated bumps or pits on the outer surface and also at the interface between the film and the substrate. In the end, these will be supposed to have spatial length scales small compared to both the initial and final state spin waves in the two magnon process. Such defects lead to perturbations in the various terms in the Hamiltonian discussed above for the perfect film. The relative magnitude of these contributions was explored in detail in Ref. 14, and it was concluded that the dominant contribution to the two magnon matrix element has its origin in the surface anisotropy term. We assume that the anisotropy axis is always normal to the local surface element of the film, so we write the defect induced change in surface anisotropy as

$$\Delta H_A = \frac{K_s}{M_s^2}\left\{\int_S dS[\vec{n}\cdot\vec{M}(\vec{r}_\parallel)]^2 - \int_{\bar{S}} dS M_y^2(\vec{r}_\parallel)\right\}. \quad (48)$$

In the first term, the integral is over the (imperfect) surface of the real film S and, in the second, the integral is over the perfectly flat surface $\bar{S}$. The unit vector $\hat{n}$ is normal to the local surface of the real film. To second order in the deviation of the magnetization from equilibrium, we have

$$[\hat{n}\cdot\vec{M}(\vec{r}_\parallel)]^2 = (\hat{Z}\cdot\hat{n})^2 M_Z^2(\vec{r}_\parallel) + 2M_s(\hat{Z}\cdot\hat{n})[\hat{n}\cdot\vec{m}(\vec{r}_\parallel)] + [\hat{n}\cdot\vec{m}(\vec{r}_\parallel)]^2, \quad (49)$$

where $M_z(\vec{r}_\parallel)$ is defined in Eq. (4). One finds that $\hat{Z}\cdot\hat{n} = \hat{n}_y \sin\phi_M + \hat{n}_z\cos\phi_M$, and $\hat{n}\cdot\vec{m}(\vec{r}_\parallel) = \hat{n}_x m_X + (\hat{n}_y\cos\phi_M - \hat{n}_Z\sin\phi_M)m_Y$.

Our assumption that the spatial dimensions of the defects are small compared to the wavelength of the spin waves involved in two magnon scattering allows us to evaluate the magnetization components at a fiducial point in each defect, and remove these terms from the integral. For simplicity, we shall also assume that the topology of the defects is such that $\int dS\hat{n}_\alpha(\vec{r}_\parallel)n_\beta(\vec{r}_\parallel)=0$, when $\alpha\neq\beta$. Finally in the second term in Eq. (48), we have $M_y^2(\vec{r}_\parallel)=M_Z^2(\vec{r}_\parallel)\sin^2\phi_M+m_Y^2(\vec{r}_\parallel)\cos^2\phi_M+2M_s m_Y(\vec{r}_\parallel)\sin\phi_M\cos\phi_M$. When these statements are combined, for the contribution of defect j to the two magnon coupling, we find

$$\Delta H_A^j=\frac{2K_s\langle S_d\rangle}{M_s}m_Y(\vec{r}_\parallel^j)[f_y^j-f_z^j-1]\sin\phi_M\cos\phi_M+\frac{K_s\langle S_d\rangle}{M_s^2}\{g_X^j m_X^j(\vec{r}_\parallel)^2+g_Y^j m_Y^j(\vec{r}_\parallel)^2\}. \tag{50}$$

We have introduced

$$g_X^j=f_x^j+(1-f_y^j)\sin^2\phi_M-f_z^j\cos^2\phi_M, \tag{51a}$$

$$g_Y^j=(f_y^j-f_z^j-1)\cos(2\phi_M), \tag{51b}$$

$$\text{and}\quad f_\alpha^j=\frac{1}{\langle S_d\rangle}\int_j \hat{n}_a^2(\vec{r}_\parallel)dS. \tag{51c}$$

In these expressions, $\langle S_d\rangle$ is the average of the projected areas of the defects onto the perfect surface. Notice that $\Sigma_\alpha f_\alpha^j=S^j/\langle S_d\rangle$, with S^j the surface area of defect j.

In Eq. (50), we see a term linear in the Y component of the transverse magnetization. As we see, no such term is present for the case considered in Ref. 14, where attention was confined to the case where the magnetization is in plane. In principle, these terms will lead to a change in the orientation of the equilibrium magnetization, assuming they remain nonzero when averaged over the array of random defects. We assume this effect is small and, in what follows, we shall set this term aside and retain only the quadratic terms, which then leads to an explicit form for the two magnon term in the Hamiltonian:

$$\Delta H_A=\frac{\langle S_d\rangle K_s}{M_s^2 Ld}\sum_{\vec{k}_\parallel,\vec{k}_\parallel'}\{G_X(\vec{k}_\parallel-\vec{k}_\parallel')m_X^\dagger(\vec{k}_\parallel)m_X(\vec{k}_\parallel')+G_Y(\vec{k}_\parallel-\vec{k}_\parallel')m_Y^\dagger(\vec{k}_\parallel)m_Y(\vec{k}_\parallel')\}. \tag{52}$$

We have $G_\alpha(\vec{k}_\parallel-\vec{k}_\parallel')=(1/L)\Sigma_j g_\alpha^j\exp[-i(\vec{k}_\parallel-\vec{k}_\parallel')\cdot\vec{r}_\parallel^j]$, where $\alpha=X,Y$. Thus, within this picture, we have $V_{XY}(\vec{k}_\parallel,\vec{k}_\parallel')=0$ and $V_{\alpha\alpha}(\vec{k}_\parallel,\vec{k}_\parallel')=(\langle S_d\rangle H_s/M_sL)G_\alpha(\vec{k}_\parallel,\vec{k}_\parallel')$ again with $\alpha=X,Y$. When we insert these results into Eq. (39) for the matrix element $N(0,\vec{k}_\parallel)$, we then have

$$N(0,\vec{k}_\parallel)=\frac{\gamma^2H_s^2\langle S_d\rangle^2}{M_s^2L^2}|H_Y(0)G_X(-\vec{k}_\parallel)+H_X(0)G_Y(-\vec{k}_\parallel)|^2. \tag{53}$$

When we expand the right hand side of Eq. (53) and average over a random array of defects, we encounter the averages $\langle\Sigma_{j,j'}g_\alpha^j g_\beta^{j'}\exp[i\vec{k}_\parallel\cdot(\vec{r}_\parallel^j-\vec{r}_\parallel^{j'})]\rangle=2N_d\langle g_\alpha^j g_\beta^j\rangle$ with $2N_d$ the total number of defects on the film; N_d is the number on one of the two surfaces. For a random array of uncorrelated defects, $\langle\exp[i\vec{k}_\parallel\cdot(\vec{r}_\parallel^j-\vec{r}_\parallel^{j'})]\rangle=0$ if $\vec{r}_\parallel^j\neq\vec{r}_\parallel^{j'}$. If we let $p=N_d\langle S_d\rangle/L^2$ be the fraction of the surface covered by defects, we then have

$$N(0,\vec{k}_\parallel)=\frac{2\gamma^2p\langle S_d\rangle H_s^2}{M_s^2L^2}\langle\bar{H}(0)^2\rangle, \tag{54a}$$

where

$$\langle\bar{H}(0)^2\rangle=\langle|H_Y(0)g_X^j+H_X(0)g_Y^j|^2\rangle. \tag{54b}$$

For the picture we are using, the matrix element $N(0,\vec{k}_\parallel)$ is independent of wave vector. Thus, in Eqs. (45) and (47), we may remove the matrix element from the integral, convert the sums on $\vec{k}_\parallel$ to integrals, and carry out the elementary integrations. For the damping rate given in Eq. (45), we find for the two magnon damping rate

$$\Gamma_{FMR}^{(2)}=\frac{2\gamma^2pH_s^2\langle S_d\rangle\langle\bar{H}(0)^2\rangle}{\pi D[H_X(0)+H_Y(0)]}\phi^{(c)}, \tag{55}$$

where $\phi^{(c)}$ is the critical angle defined in Eq. (23a). It should be noted that in Ref. 18, it is pointed out that the data presented in this paper suggests the two magnon contribution to the linewidth scales as the critical angle $\phi^{(c)}$.

At the level of approximation we are using here, once $N(0,\vec{k}_\parallel)$ is removed from the integral in Eq. (47), we are left with a logarithmically divergent integral:

$$\delta\Omega_{FMR}^{(2)}=\frac{\gamma^2pH_s^2\langle S_d\rangle\langle\bar{H}(0)^2\rangle}{4\pi^2\Omega_{FMR}D[H_X(0)+H_Y(0)]}\int\frac{dk_\parallel d\phi_{\vec{k}_\parallel}}{k(\phi_{\vec{k}_\parallel})-k_\parallel}. \tag{56}$$

The origin of the divergence is our assumption that all spin waves involved in the two magnon scatterings have wavelengths long compared to the length scale, which characterizes the defects. This is quite reasonable in the discussion of two magnon induced damping, but in the calculation of the frequency shift, we reach out into the Brillouin zone sufficiently far that this assumption breaks down. To remedy this properly would require a microscopic picture of the defects; a form factor would then enter the matrix element, which would provide a wave vector cutoff. To introduce this feature fully and completely would involve an extensive analysis. Since the divergence is only logarithmic in character, the precise details of how the wave vector cutoff is handled are not so critical. Thus, as in Ref. 14, we shall just cutoff the integration over the magnitude of the wave vector in Eq. (56) at a value $k^{(M)}$, which is understood to have a value equal to the inverse of the length scale which characterizes the defects. The resulting integral may be taken from Ref. 14, so we have

$$\delta\Omega_{FMR}^{(2)} = -\frac{\gamma^2 p H_s^2 \langle S_d\rangle\langle \bar{H}(0)^2\rangle}{\pi\Omega_{FMR} D[H_X(0)+H_Y(0)]} \ln\left[\left(\frac{k^{(M)}}{\tilde{k}} - \sin^2\phi^{(c)}\right)^{1/2} + \left(\frac{k^{(M)}}{\tilde{k}} + \cos^2\phi^{(c)}\right)^{1/2}\right]. \quad (57)$$

The results in Eqs. (56) and (57) apply to any circumstance where random surface and interface defects with length scale small as compared to the wavelength of the magnons are degenerate in frequency with the FMR mode. The wave vector of the degenerate modes is on the scale of $\tilde{k}$ as defined in Eq. (23b). Thus, if the length scale that characterizes the defects is l, we require $\tilde{k}l \ll 1$ to be satisfied. For typical parameters, one has $\tilde{k} \approx 10^5$ cm^{-1}, so if the defects are on the nanometer length scale, the inequality just stated is satisfied.

It is perhaps useful to have an explicit form for the averages, which appear in the quantity $\langle \bar{H}(0)^2\rangle$. For this we need to introduce a possible picture of the topology of the defects. We follow Ref. 14 and suppose these to be rectangular in nature, with sides of length a parallel to the x axis, sides of length c parallel to the z axis, and height (or depth) b if the defect is a island (or a pit). We regard a and c to be randomly distributed. Then, after averaging over the ensemble of defects, $\langle a\rangle = \langle c\rangle = l$, where l is the length introduced in the previous paragraph, we can take the cutoff wave vector $k^{(M)}$ in Eq. (57) to be $1/\langle a\rangle$. For this picture, $f_y=1$ for each defect, and $f_x=2b/a$, $f_z=2b/c$ for a particular defect with dimensions $a\times c\times b$. We can insert these expressions into $\langle|\bar{H}(0)|^2\rangle$, and average over an ensemble as described in Ref. 14 if desired. Of course, our formalism can readily be adapted to numerous physical pictures of the defect array which activates two magnon scattering, including the dislocation line array described in Ref. 18, though the nature of the disorder present in the dislocation array is not so clear to the present writers.

III. CALCULATIONS OF THE LINEWIDTH AND GENERAL DISCUSSION

In this section, we present calculations of the two magnon contribution to the FMR linewidth which explore the effect of tipping the magnetization out of the plane. For the purposes of illustration, the results presented below employ the parameters utilized in the calculations presented in Ref. 14. We do need to provide a brief discussion of how the quantity $\Gamma_{FMR}^{(2)}$ discussed in the previous section is related to the FMR linewidth.

We examine the response function in Eq. (38) and, for the moment, we set aside the two magnon induced frequency shift, so we retain just the imaginary part of the self-energy Σ. The response function then can be written as

$$\bar{S}_{XX}(0,\Omega) = \frac{\gamma^2 M_s H_Y(0)[(\Omega_{FMR}^2-\Omega^2)+i\Lambda]}{(\Omega_{FMR}^2-\Omega^2)^2+\Lambda^2}, \quad (58)$$

where $\Lambda=\gamma g\Omega[H_X(0)+H_Y(0)]+\Gamma_{FMR}^{(2)}$. In a FMR experiment, the frequency is held fixed, and the external magnetic field H_0 is swept through the resonance field $H_0^{(R)}$ for which $\Omega_{FMR}=\Omega$. We need to examine the structure of the response function in Eq. (58) for applied fields near the resonance field. We write $H_0=H_0^{(R)}+\Delta H$ and, for small ΔH, we have $\Omega_{FMR}^2-\Omega^2=\gamma^2\cos(\phi_H-\phi_M)[H_X(0)+H_Y(0)]\Delta H$. The response function can then be written in the form

$$\bar{S}_{XX}(0,\Omega) = \left(\frac{M_s H_Y(0)[(\Omega_{FMR}^2-\Omega^2)+i\Lambda]}{\gamma^2\cos^2(\phi_H-\phi_M)[H_X(0)+H_Y(0)]^2}\right)\frac{1}{\Delta H^2+(\Delta H_{FMR})^2}, \quad (59)$$

where ΔH_{FMR} is the FMR linewidth in gauss. We have

$$\Delta H_{FMR} = \frac{G\Omega_{FMR}}{\gamma^2 M_s\cos(\phi_H-\phi_M)} + \frac{\Gamma_{FMR}^{(2)}}{\gamma^2\cos(\phi_H-\phi_M)[H_X(0)+H_Y(0)]}. \quad (60)$$

Of course, the first term on the right hand side of Eq. (60) is the Landau Lifschitz damping term, and the second is the two magnon contribution to the linewidth. In Fig. 2, we show the frequency dependence of the linewidth for various cases where the angle between the applied magnetic field and the plane of the film are fixed. For each value of the applied field, we use Eq. (3) to calculate the angle ϕ_M the magnetization makes with the plane of the film. Clearly, $\phi_M < \phi_H$ always. The curve labeled $\phi_H=0°$ agrees with the result

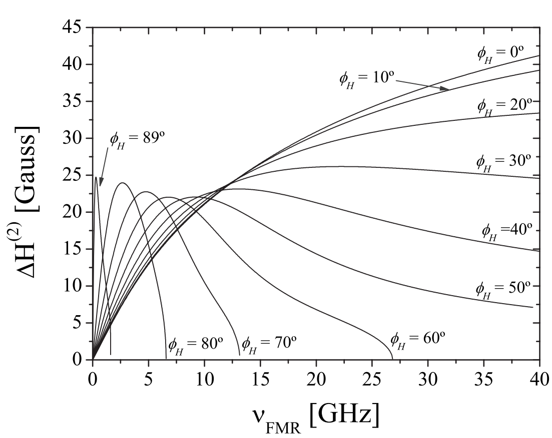

FIG. 2. The frequency dependence of the two magnon contribution to the FMR linewidth, for the situation where the angle between the externally applied field and the plane of the film is held fixed, as the external field is increased. The parameters employed in these calculations are the same as those utilized in Ref. 14.

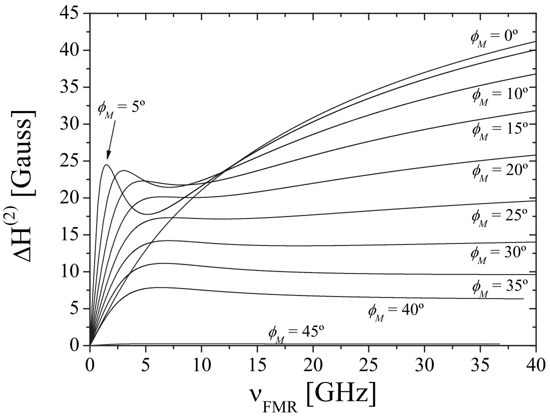

FIG. 3. We illustrate the frequency dependence of the linewidth, as the angle between the magnetization and the film plane is kept fixed, as the external field is varied. As in Fig. 2, the parameters are the same as those employed in Ref. 14

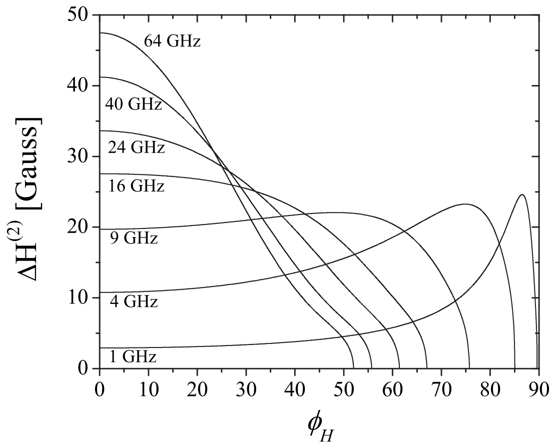

FIG. 4. For various frequencies, we show the variation of the two magnon contribution to the FMR linewidth as a function of the angle between the external magnetic field and the plane of the film.

presented in Ref. 14. When the angle ϕ_H exceeds 45°, the linewidth goes to zero at that value of the frequency where the angle ϕ_M between the magnetization and the film plane equals 45°.

It must be kept in mind, of course, that in actual samples the intrinsic Gilbert damping is always present as well, in addition to the two magnon process under study in this paper. The Gilbert damping leads to a contribution to the FMR linewidth linear in frequency[1,13] and, at high frequencies, this intrinsic source of damping will dominate two magnon scattering. For the case where the film is magnetized in plane, in Fig. 1 of Ref. 17, one sees an illustration of the effect on the linewidth of including intrinsic Gilbert damping in addition to two magnon scattering. In Fig. 3, we see data on an actual sample and a comparison with theory, again for an in-plane magnetized film, for frequencies from 1 to over 70 GHz.

In Fig. 3, we show the frequency variation of the linewidth as the angle ϕ_M between the magnetization and the film plane is held fixed. In actual experiments, of course, as the magnitude of the applied field is varied, one does not vary ϕ_H in such a way that ϕ_M remains constant, as we have done in generating Fig. 3. We do note that in Ref. 19, we find a plot of the linewidth as a function of ϕ_M for a fixed frequency ν_{FMR}. Such plots are readily constructed for any desired frequency from Fig. 3. Notice that when ϕ_M is greater than 45°, the two magnon damping is silent. The nonmonotonic behavior of the linewidth with frequency at small tipping angles is striking in our view.

Finally, at various fixed frequencies, in Fig. 4 we show the dependence of the two magnon contribution to the FMR linewidth as a function of the angle ϕ_H. The cutoff frequencies once again occur when the angle between the magnetization and the film plane becomes 45°.

In Figs. 2–4, we see that the dependence of the two magnon contribution to the linewidth can show a striking behavior as the magnetization is tipped out of plane. It should be kept in mind that these calculations employ a particular model of the defect structure, and the behavior may change if the defect structure that activates two magnon scattering deviates substantially from the picture utilized in these calculations. As remarked earlier, the notion that the two magnon contribution to the linewidth is cut off when the angle between the magnetization and the film plane is greater than 45° is supported nicely by the data reported in Ref. 19. Of course, it will be of great interest to see detailed studies of the influence of tipping the magnetization out of plane.

IV. FINAL REMARKS

In this paper, we have extended the theory of two magnon scattering and its influence on the response of ultrathin ferromagnetic films developed in Ref. 14 to the case where the magnetization is tipped out of plane. We provide here explicit expressions for the two magnon contribution to the FMR linewidth and the two magnon induced frequency shift of the FMR line, in a form sufficiently general for application to a diverse array of defect structures. We also provide complete expressions for the frequency and wave vector dependent susceptibility tensor denoted above by $\bar{S}_{\alpha\beta}(\vec{k}_{\parallel},\Omega)$. This will allow the reader to analyze the influence of defect induced spin wave scatterings on Brillouin light scattering spectra as well, if desired. It is our view that to have the full form of the response functions in hand will prove useful in a variety of contexts.

It is the case, as illustrated by the calculations presented in Sec. III, that the two magnon contribution to the FMR linewidth exhibits a striking dependence on the angle of both the magnetization and the externally applied field with respect to the film plane. At least in principle, in a given sample, it should be possible to control the damping rate of spin motions by tipping the magnetization with respect to the film plane, if the circumstances are such that two magnon scattering is responsible for an appreciable fraction of the total damping rate. This situation is realized in practice when the ultrathin ferromagnet is grown on an exchange biasing substrate.[16,19]

It is our hope that the results presented here will stimulate new and more detailed studies of the influence of tipping the magnetization out of plane on the response characteristics of ultrathin ferromagnets.

ACKNOWLEDGMENTS

During the course of this research, we have enjoyed interactions with J. Lindner. The research of D.L.M. has been supported by the U.S. Army through Contract No. CS000128. That of P.L. and R.E.A. has been partially supported by this source as well, and they also acknowledge support from the Millenium Science Nucleus "Basic and Applied Magnetism," P06-022-F (Chile). P.L. also acknowledges support from the program Bicentario en Cienca y Tecnologia PBCT under Project PSD-031.

APPENDIX: DIPOLAR FIELD GENERATED BY THE SPIN MOTION

Here, we generate an expression for the dynamic dipole field generated by the spin motion. As noted in the caption of Fig. 1, we take the film to lie in the region $-d/2<y<+d/2$. We shall derive a general expression that is first order in the amplitude of the spin motion without taking the ultrathin limit, so the reader is provided with a general form. Then we take the ultrathin limit when the discussion is completed.

In the magnetostatic limit, we may write $\vec{h}_d^{(1)}(\vec{r})=-\vec{\nabla}\Phi^{(1)}(\vec{r})$, where

$$\nabla^2\Phi^{(1)}(\vec{r})=4\pi\vec{\nabla}\cdot\vec{M}^{(1)}(\vec{r}_\parallel). \tag{A1}$$

We have

$$\vec{\nabla}\cdot\vec{M}^{(1)}(\vec{r}_\parallel)=\frac{\partial m_X(\vec{r}_\parallel)}{\partial x}-\frac{\partial m_Y(\vec{r}_\parallel)}{\partial z}\sin\phi_M. \tag{A2}$$

We write the potential in the form

$$\Phi(\vec{r})=\frac{1}{\sqrt{L^2d}}\sum_{\vec{k}_\parallel}\Phi^{(1)}_{\vec{k}_\parallel}(y)\exp(i\vec{k}_\parallel\cdot\vec{r}_\parallel), \tag{A3}$$

and one sees easily that

$$\left[\frac{\partial^2}{\partial y^2}-k_\parallel^2\right]\Phi^{(1)}_{\vec{k}_\parallel}(y)=f(\vec{k}_\parallel), \tag{A4}$$

where

$$f(\vec{k}_\parallel)=4\pi i[k_{\parallel x}m_X(\vec{k}_\parallel)-k_{\parallel z}\sin\phi_M m_z(\vec{k}_\parallel)]. \tag{A5}$$

Equation (A4) applies only in the film, of course, and outside the film, the potential satisfies the homogeneous form of Eq. (A4). Thus, we seek a solution where

$$\Phi^{(1)}_{\vec{k}_\parallel}(y)=\begin{cases}A_{\vec{k}_\parallel}\exp(-k_\parallel y), & y>d/2\\ a_{\vec{k}_\parallel}\exp(-k_\parallel y)+b_{\vec{k}_\parallel}\exp(k_\parallel y)-[f(\vec{k}_\parallel)/k_\parallel^2], & -d/2<y<+d/2\\ B_{\vec{k}_\parallel}\exp(k_\parallel y), & y<-d/2.\end{cases} \tag{A6}$$

The coefficients in Eq. (A6) may be determined from the boundary conditions on the two surfaces of the film. These are that $\Phi^{(1)}_{\vec{k}_\parallel}(y)$ is continuous at $y=\pm d/2$, along with $b_y^{(1)}(y)=-(\partial\Phi^{(1)}_{\vec{k}_\parallel}/\partial y)+4\pi m_y^{(1)}(\vec{k}_\parallel)$. From the boundary conditions, we find the relations

$$A_{\vec{k}_\parallel}=a_{\vec{k}_\parallel}-b_{\vec{k}_\parallel}\exp(k_\parallel d)+g(\vec{k}_\parallel)\exp(k_\parallel d/2) \tag{A7a}$$

and

$$B_{\vec{k}_\parallel}=-a_{\vec{k}_\parallel}\exp(k_\parallel d)+b_{\vec{k}_\parallel}-g(\vec{k}_\parallel)\exp(k_\parallel d/2), \tag{A7b}$$

where we have defined

$$g(\vec{k}_\parallel)=4\pi m_Y(\vec{k}_\parallel)\cos\phi_M/k_\parallel. \tag{A7c}$$

We also have

$$a_{\vec{k}_\parallel}=\frac{\exp(-k_\parallel d/2)}{2}\left\{\frac{f(\vec{k}_\parallel)}{k_\parallel^2}-g(\vec{k}_\parallel)\right\} \tag{A8a}$$

and

$$b_{\vec{k}_\parallel}=\frac{\exp(-k_\parallel d/2)}{2}\left\{\frac{f(\vec{k}_\parallel)}{k_\parallel^2}+g(\vec{k}_\parallel)\right\} \tag{A8b}$$

so that

$$\Phi^{(1)}_{\vec{k}_\parallel}(y)=[e^{-k_\parallel d/2}\cosh(k_\parallel y)-1]\frac{f(\vec{k}_\parallel)}{k_\parallel^2}+e^{-k_\parallel d/2}\sinh(k_\parallel y)g(\vec{k}_\parallel). \tag{A9}$$

It is now a straightforward matter to form an expression for $\vec{h}_d^{(1)}(\vec{r})$ and carry out the integral that appears in Eq. (9). One finds

$$-\frac{1}{2}\int\vec{M}^{(1)}(\vec{r}_\parallel)\cdot\vec{h}_d^{(1)}(\vec{r})d^3r=\frac{\cos\phi_M}{2d}\sum_{\vec{k}_\parallel}m_Y^\dagger(\vec{k}_\parallel)(1-e^{-k_\parallel d})g(\vec{k}_\parallel)$$

$$+\frac{i}{2}\sum_{\vec{k}_{\parallel}}[k_{\parallel x}m_X^{\dagger}(\vec{k}_{\parallel})-k_{\parallel z}m_Y^{\dagger}(\vec{k}_{\parallel})\sin\phi_M]\left[\frac{(1-e^{-k_{\parallel}d})}{k_{\parallel}d}-1\right]\frac{f(\vec{k}_{\parallel})}{k_{\parallel}^2}. \tag{A10}$$

Our interest is in the ultrathin film limit, so we take the limit $k_{\parallel}d \ll 1$ in Eq. (10), retaining the zero order and first order terms in $k_{\parallel}d$. We then have

$$-\frac{1}{2}\int \vec{M}^{(1)}(\vec{r}_{\parallel})\cdot\vec{h}_d^{(1)}(\vec{r})d^3r = 2\pi\sum_{\vec{k}_{\parallel}}\left(1-\frac{k_{\parallel}d}{2}\right)\cos^2\phi_M m_Y^{\dagger}(\vec{k}_{\parallel})m_Y(\vec{k}_{\parallel}) + \pi\sum_{\vec{k}_{\parallel}}k_{\parallel}d[\sin^2\phi_{\vec{k}_{\parallel}}m_X^{\dagger}(\vec{k}_{\parallel})m_X(\vec{k}_{\parallel}) + \sin^2\phi_M\cos^2\phi_{\vec{k}_{\parallel}}m_Y^{\dagger}(\vec{k}_{\parallel})m_Y(\vec{k}_{\parallel}) - \sin\phi_M\cos\phi_{\vec{k}_{\parallel}}\sin\phi_{\vec{k}_{\parallel}}\{m_Y^{\dagger}(\vec{k}_{\parallel})m_X(\vec{k}_{\parallel}) + m_X^{\dagger}(\vec{k}_{\parallel})m_Y(\vec{k}_{\parallel})\}]. \tag{A11}$$

When Eq. (A11) is inserted into Eq. (9), the result is that displayed in Eq. (10).

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