Hamiltonian formalism for two magnon scattering microwave relaxation: Theory and applications

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A two magnon scattering theory for microwave relaxation in magnetic systems is formulated in the framework of the Hamiltonian formalism. The paper provides general expressions for inhomogeneity coupling coefficients in the case of localized inhomogeneities. An approximate solution for the relaxation rate of the ferromagnetic resonance uniform mode relaxation rate is presented. Two examples of the application of the theory are presented, one for bulk polycrystalline ferrites and one for polycrystalline metallic thin films. © 2007 American Institute of Physics.

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I. INTRODUCTION

Processes that involve decay in the amplitude of magnetization oscillations, generally termed relaxation processes, are important for fundamental and practical reasons. These processes, for example, set a natural limit for switching times and magnetic recording data transfer rates. There are two main pathways for magnetization relaxation: (i) energy transfer out of magnetic system to a thermal bath that consists of phonons or conduction electrons, for example, and (ii) energy redistribution within a magnetic system. The former is usually termed as intrinsic damping. The latter involves coupling between the various magnetization oscillation modes, often called spin waves in wave theory or magnons in quantum particle theory. These two terms are often used interchangeably.

In the lowest order magnon coupling process, where only two excitations are involved, one magnon is destroyed and another is created. Such a process is called two magnon scattering (TMS). This process can be described in terms of a scattering of the spin waves due to bulk and/or surface inhomogeneities. In the case of static inhomogeneities, the scattering may be taken as elastic, so that the initial and final magnon states will have the same frequency \( \omega \). The state wave vectors, denoted by \( \mathbf{k} \), however, will differ by amounts that correlate with the spatial variation of the inhomogeneities.

Two magnon scattering processes are often invoked to account for additions to the intrinsic ferromagnetic resonance (FMR) relaxation rate or resonance linewidth for bulk and thin film ferrites as well as metallic magnetic films and multilayers. The driven FMR mode is usually spatially uniform or near uniform and is taken to correspond to a \( \mathbf{k} = 0 \) spin wave excitation. A valid two magnon scattering relaxation channel for the uniform mode requires two things: (1) available nonzero \( \mathbf{k} \) modes that are at the same frequency as the driven FMR mode and (2) inhomogeneities with appropriate sizes or size distributions to couple the FMR mode to these degenerate spin wave modes. The total FMR relaxation rate then reflects a decay of uniform mode magnons due to TMS as well as intrinsic processes.

In the case of weak inhomogeneities, the coupling between uniform and nonuniform modes can be treated as a small perturbation and the TMS contribution to the relaxation rate can be evaluated by Fermi’s golden rule. This quantum mechanical approach has been adopted by several authors. Reviews may be found in Refs. 2 and 12, for example. Alternative methods involve the evaluation of the inhomogeneity contribution to the microwave susceptibility, or the magnetization time response, as well as the direct numerical diagonalization of the Hamiltonian with inhomogeneity coupling terms included.

Higher order approaches include considerations of secondary scattering between nonuniform magnons and modifications in the density of degenerate states in the presence of inhomogeneities.

Two magnon scattering contributions to the uniform mode ferromagnetic resonance relaxation rate have been investigated theoretically and experimentally since the late 1950s. The very early works focused on TMS processes in bulk ferrites. The intense technological interest in ferrite based radar devices drives continuing work in this area to the present day. The main contributions to the TMS relaxation rate in ferrite systems come from (i) coupling due to the dipole fields associated with pores and surface voids and (ii) effective field fluctuations associated with the magneto-crystalline anisotropy of the randomly oriented grains in polycrystalline samples. The particular case of the thin ferrite disk geometry was considered, for example, by Sparks and most recently by Hurben and Patton, among others.

There has also been a strong interest in TMS relaxation in metallic thin films. Wigen was the first to incorporate the TMS theory into the analysis of linewidth data for obliquely magnetized Permalloy films. In this analysis, however, the density of degenerate states was evaluated from bulk spin wave dispersion relations rather than those applicable to thin films. It is known, however, that the dynamic dipole fields associated with film surfaces can modify the applicable dis-
persion relations and density of states considerations in a significant way. Such effects can lead, for example, to profound dependences of the FMR linewidth on the film thickness.

A semiquantitative discussion of two magnon scattering contributions to the FMR linewidth in thin metallic films was first provided by Patton et al. for in-plane magnetized Permalloy films. The full theoretical analysis of two magnon scattering processes in thin metallic films started some decades later with the work of Arias and Mills. Their model involved scattering from regularly shaped surface defects due to both dipole fields and surface anisotropy, and it specifically addressed the situation for ultrathin films. The model predictions have been tested experimentally by Lindner et al. and McMichael and Krivosik, for example. Recently, McMichael and Krivosik presented a detailed classical model of TMS relaxation for localized randomly distributed inhomogeneities. Nonlocal dipole scattering due to a random surface roughness has been treated by Dobin and Victor, and by Maranville et al.

This article follows closely the TMS analysis of McMichael and Krivosik for localized scattering. The theory presented here, however, is formulated within the framework of a Hamiltonian formalism. This approach is widely used in the theory of weakly dissipative systems, and it is particularly suitable for the study of nonlinear magnetic excitations. Recently, for example, there has been a large amount of Hamiltonian based theory on a wave front reversal in ferrite films. This phenomenon involves both two magnon scattering and nonlinear parametric processes, and this combination holds significant potential for new microwave signal processing devices. Hamiltonian approaches to TMS processes have also been used in the analysis of nonlinear magnetization dynamics in the vicinity of the ferromagnetic resonance and large angle switching.

In spite of the above activity, there exists no general and comprehensive statement of the formal Hamiltonian TMS theory and the corresponding Hamiltonian coefficients for basic magnetic systems. This work addresses this problem.

Section II presents the basic theoretical approach. It gives a review of the working equations for the dynamic magnetization response in a homogeneous sample, provides general expressions for the inhomogeneity coupling coefficients for localized inhomogeneities, and presents an approximate solution for a uniform mode free decay response. Section III provides an example calculation for anisotropy scattering in a polycrystalline sphere. In Sec. IV, a simplified theoretical approach is used for the analysis of the original data on Permalloy thin films from Ref. 6. Section V provides a summary and conclusions. Gaussian units are used throughout.

II. THE HAMILTONIAN FORMALISM

A. Dynamic magnetization response and equation of motion

This section provides an overview of the Hamiltonian method in the spin-wave theory. There are several methods for the theoretical analysis of spin wave excitations in ferromagnets. In the classical approach, spin waves are regarded as propagating deviations of the macroscopic magnetization \( \mathbf{M} \) from equilibrium. The dynamics of these deviations derives from the classical torque equation of motion,

\[
\frac{d\mathbf{M}(r,t)}{dt} = -\gamma \mathbf{M}(r,t) \times \mathbf{H}_{\text{eff}}(r,t),
\]

where \( \gamma \) is the electron gyromagnetic ratio and \( \mathbf{H}_{\text{eff}}(r,t) \) is a net effective field that includes both external and internal field components. In order to facilitate the analysis of the spin wave dynamics and interactions in a compact way, Eq. (1) is commonly transformed into a set of equations of motion for scalar complex Fourier component spin-wave amplitudes. The Hamiltonian formalism provides a way to achieve such a transformation in four conceptually simple steps.

In the first step, one needs to write down the total magnetic energy \( \mathcal{H} \) in specific form. In general terms, \( \mathcal{H} \) may be written according to

\[
\mathcal{H} = \int_V \mathcal{W} d^3r,
\]

where \( \mathcal{W} \) is an energy density and the integral is over the sample volume \( V \). Both \( \mathcal{H} \) and \( \mathcal{W} \) are real. The connection between this energy and the effective field in Eq. (1) is given by

\[
\mathbf{H}_{\text{eff}}(r,t) = -\frac{1}{V} \frac{\delta \mathcal{H}}{\delta \mathbf{M}(r,t)},
\]

with the variational derivative defined in the standard way

\[
\frac{\delta \mathcal{H}}{\delta \mathbf{M}} = V \left[ \frac{\partial \mathcal{W}}{\partial \mathbf{M}} - \sum_{a=x,y,z} \frac{d}{d\alpha} \frac{\partial \mathcal{W}}{\partial (d\mathbf{M}/d\alpha)} \right].
\]

In the second step, the magnetization components \( M_{x,y,z} \) are expressed in terms of a pair of conjugate variables \( a(r,t) \) and \( a^*(r,t) \) according to

\[
iM_x(r,t) + M_y(r,t) = M_x a(r,t) \sqrt{2 - a(r,t)a^*(r,t)}
\]

and

\[
M_z(r,t) = M_s [1 - a(r,t)a^*(r,t)].
\]

Here, \( M_s \) denotes the saturation magnetization of the material of interest. This transformation was first developed by Schlömann. It has been used by many authors for the study of spin wave dynamics and related problems. One can see that in the small signal limit \( a \) has a simple physical interpretation, namely, \( a \sim i\alpha_x + \alpha_y \), where \( \alpha_{x,y} \) are the \( x- \) and \( y \)-direction cosines of the magnetization vector \( \mathbf{M} \).

With the use of Eqs. (5) and (6), Eq. (1) may be transformed into a pair of coupled equations of motion for \( a(r,t) \) and \( a^*(r,t) \) in Hamiltonian form, according to

\[
i \frac{d a(r,t)}{dt} = \frac{\delta U}{\delta a^*(r,t)}
\]

and the corresponding complex conjugate expression, 

\[-i a^*(r,t)/dt = \frac{\delta U}{\delta a(r,t)}\].

In the above, \( U \) is the magnetic energy from Eq. (2), but now expressed in frequency units according to
\[ U = \frac{|\gamma|}{M_r V} \mathcal{H}. \]  
(8)

As with \( \mathcal{H} \), \( U \) is always real.

In the third step, the spatial dependences of the conjugate variables \( a(\mathbf{r}, t) \) and \( a^*(\mathbf{r}, t) \) are expanded in a suitable basis system. In the usual case, linear normal modes for a given sample geometry are chosen as the basis functions. The appropriate choice in many situations is a plane wave Fourier series expansion according to

\[ a(\mathbf{r}, t) = \sum_k a_k(t) e^{i\mathbf{k} \cdot \mathbf{r}} \]  
(9)

along with the corresponding complex conjugate expression for \( a^*(\mathbf{r}, t) \). Each \( a_k(t) \) and \( a_k^*(t) \) expansion coefficient represents the complex amplitude of a planar spin wave that propagates in a direction of the wave vector \( \mathbf{k} \). The \( \mathbf{k}=0 \) mode corresponds to the uniform magnetization mode. Such a plane wave expansion has been established as a permissible approximation for bulk samples as long as the mode wavelength \((2\pi/|\mathbf{k}|)\) is much smaller than the sample dimensions. Plane wave expansion has been also employed for laterally unbounded ultrathin films for which the condition \( |\mathbf{k}|d \ll 1 \) is satisfied, where \( d \) is the film thickness.\(^{22}\) In this ultrathin film limit, the dynamic magnetization amplitude can be taken to be uniform across the film thickness, and the normal modes are, in essence, propagating plane waves confined in the film cross section. The Fourier expansion is then over \( \mathbf{k} \) vectors constrained to the film plane.

The \( a_k \) variables are often rescaled as \( \hat{a}_k = \sqrt{M_r V/g}\mu_B k \), where \( g \) is the Landé \( g \) factor and \( \mu_B \) is the Bohr magneton.\(^{13,19}\) A given pair of these rescaled \( \hat{a}_k \) and \( \hat{a}_k^* \) then represent the classical analog to the usual magnon annihilation and creation operator pair in the second quantization formalism of quantum mechanics.\(^{43}\) This analog originates from the fact that each excited spin wave reduces the \( z \) component of the total magnetic moment of the sample by a given amount, either by \( M_r V\hat{a}_k \hat{a}_k^* \) or by \( g\mu_B \hat{a}_k \hat{a}_k^* \).

The Fourier transform operation is canonical and the equations of motion for the \( a_k \) and \( a_k^* \) will therefore retain the Hamiltonian form of Eq. (7), but with the variational derivatives replaced by partial derivatives. This replacement relates to the fact that the \( a_k \) and \( a_k^* \) are amplitudes in discrete \( \mathbf{k} \) space.

There is one more crucial modification. Intrinsic dissipation due to interactions between the spin waves and the thermal bath is accommodated through the \textit{ad hoc} addition of a relaxation rate \( \eta_k \) term to the coupled equations of motion for \( a_k \) and \( a_k^* \).\(^{31,32}\) This relaxation rate \( \eta_k \) parameter is real. In principle, such a term could also be obtained directly from additional Hamiltonian terms that represent spin wave coupling to the thermal bath.\(^{44}\) The development will show that in the first order approximation, the two magnon relaxation rate term simply adds to the \( \eta_k \) just introduced. By implication, other two particle interaction terms will have the same effect, at least to lowest order. The \( \eta_k \), therefore, simply accounts for intrinsic thermal bath decay terms in a heuristic way.

Based on the above, the coupled equations of motion for the \( a_k \) and \( a_k^* \) may be written according to

\[ i \left( \frac{d}{dt} + \eta_k \right) a_k(t) = \frac{\partial U}{\partial a_k(t)} \]  
(10)

and

\[ -i \left( \frac{d}{dt} + \eta_k \right) a_k^*(t) = \frac{\partial U}{\partial a_k^*(t)}. \]  
(11)

In the fourth and final step, the energy \( U \) is expanded in a power series in the \( a_k \) and \( a_k^* \), written as \( U = U(0) + U(1) + U(2) + \cdots \). The superscripts indicate the degree in powers of the different \( a_k \), \( a_k^* \), \( a_k^* a_k^* \), \( a_k a_k^* \), etc., products. Although the initial Hamiltonian is usually easy to write down, this expansion in the \( a_k \) and \( a_k^* \) is often tedious to obtain. When completed, however, it allows for the evaluation of all of the different orders of spin wave interaction processes in an extremely simple manner. This operational simplicity will become clear in the sections to follow.

**B. Normal modes and spin wave dispersion for a homogeneous sample**

From the derivative form on the right sides of Eqs. (10) and (11), one can see that a truncation of the \( U \) expansion beyond the quadratic terms in the \( a_k \) and \( a_k^* \) corresponds to a linearization of the equation of motion. The zeroth order term is immaterial for dynamics. The first order term in the \( U \) expansion defines static equilibrium and may also include a pumping term from the external fields. The linearized normal modes of the magnetic system are fully defined, therefore, through the quadratic \( U(2) \) part of the energy. For a spatially homogeneous sample, this part may be expressed as

\[ U_{\text{hom}}^{(2)} = \sum_k \left\{ A_k \dot{a}_k^*(t)a_k(t) + \frac{1}{2} [B_k \dot{a}_k^*(t)a_k^*(t) + \text{c.c.}] \right\}. \]  
(12)

The expansion coefficients \( A_k \) and \( B_k \) take the form

\[ A_k = \frac{|\gamma|}{2} (H_{xx,k} + H_{yy,k}) \]  
(13)

and

\[ B_k = \frac{|\gamma|}{2} (-H_{xx,k} + H_{yy,k} + 2iH_{xy,k}), \]  
(14)

where \( H_{xx,k} \), \( H_{yy,k} \), and \( H_{xy,k} \) are components of what will be termed the \textit{homogeneous stiffness field tensor}. The specific form of these components will depend on the system of interest.\(^{45,46}\) An outline of the development of these terms is given in the Appendix. Two sets of specific expressions, one for bulk samples of ellipsoidal shape and one for in-plane magnetized thin films, are given in Secs. III and IV. Note that \( H_{xx,k}, H_{yy,k}, \) and \( H_{xy,k} \) are real. This means, in turn, that \( A_k \) is also real and the conditions \( A_k = A_{-k} \) and \( B_k = B_{-k} \) are valid.

With the form of \( U^{(2)}_{\text{hom}} \) as given in Eq. (12), Eqs. (10) and (11) will comprise coupled equations of motion for a given pair of \( a_k \) and \( a_k^* \) amplitudes or, equivalently, pairs of \( a_k \) and \( a_k^* \) amplitudes. For some aspects of the analysis, the
\( (a_k, a^*_k) \) form proves convenient. The decoupling can be accomplished through a canonical transformation first developed by Holstein and Primakoff (HP).\(^{57}\) For the present purposes, this transformation may be written as
\[
a_k(t) = u_k c_k(t) + v_k c^*_k(t),
\]
where the transformation coefficients are specified through
\[
u_k = \frac{\sqrt{A_k + \omega_k}}{2\omega_k},
\]
\[
\omega_k = \frac{\sqrt{A_k - \omega_k}}{2\omega_k},
\]
and
\[
\omega_k = \left| \frac{A_k + B_k}{|B_k|^2} \right| \gamma \sqrt{H_1 H_{2x} H_{2y} K + H_4^2/4}.
\]
Equation (18) also gives the \( \omega_k(k) \) dispersion relation for the normal mode excitations associated with these amplitudes. From the properties of \( A_k \) and \( B_k \) it follows that (1) the \( u_k \) are real and (2) the conditions \( u_k = u_{-k}, \ v_k = v_{-k}, \) and \( \omega_k = \omega_{-k} \) are satisfied.

The HP transformation casts Eq. (12) into a diagonal form as \( U^{(2)}_{\text{hom}} = \Sigma_k \omega_k c_k^*(t) c_k(t) \). The new pair of conjugate variables \( c_k(t) \) and \( c^*_k(t) \) correspond to the linear normal spin wave modes. Since the transformation from the \( a_k \) and \( a^*_k \) to the \( c_k \) and \( c^*_k \) is canonical, the equations of motion for the \( c_k \) and \( c^*_k \) follow the same form as for the \( a_k \) and \( a^*_k \). However, insofar as \( U^{(2)}_{\text{hom}} \) is diagonal in \( c_k \) and \( c^*_k \), the equations of motion are now uncoupled. One then has
\[
i \left( \frac{d}{dt} + \eta_k \right) c_k(t) = \frac{\partial U^{(2)}_{\text{hom}}}{\partial c^*_k(t)} = \omega_k c_k(t),
\]
along with the counterpart equation for \( c^*_k(t) \). For a homogeneous system, the dispersion \( \omega_k(k) \), together with the relaxation rate parameter \( \eta_k \), now provides all the information needed to describe the linear spin wave dynamics. The \( c_k(t) \) and \( c^*_k(t) \) correspond to plane waves with a well defined dispersion. One can also work backwards, as needed, to extract the physical dynamic magnetization response as a Fourier superposition of these normal modes, suitably transformed.

The working equations developed up to now constitute the lead into the main problem at hand, namely, the spin wave dynamics and the additional relaxation processes that are present in the presence of inhomogeneities. The next section considers the extension of the formalism established so far to this problem. The new ingredient is a spatial variation of the stiffness fields due to the inhomogeneities. This results in a new coupling between spin wave modes at the same frequency but with different wave vectors, or, in other words, two magnon scattering.

**C. Inhomogeneity coupling and mode mixing**

Spatial inhomogeneities introduce an additional contribution to the energy density that will be denoted as \( \tilde{\mathcal{W}} \). In what follows, a tilde (\( \tilde{\cdot} \)) will be used to denote any inhomogeneity related quantity. Spatial inhomogeneities may be classified according to the sources of the spatial fluctuating effective field and separated into two categories, local and nonlocal. A polycrystalline ferrite or thin film with densely packed randomly oriented crystalline grains, for example, constitutes a system with local inhomogeneities. Samples with pores, pits, or surface irregularities that produce long range dipole fields, on the other hand, provide examples of nonlocal inhomogeneities. The formulation here will focus on local inhomogeneities. In the case of nonlocal scattering processes, one can cast the working equations in a similar form, albeit with a somewhat more complex analysis than that given below.\(^{29}\)

For the dense polycrystalline sample with local inhomogeneities, the important contributions to \( \tilde{\mathcal{W}} \) generally derive from the magnetocrystalline anisotropy in randomly oriented grains and the \( r \)-dependent variation in \( M \) associated with the plane and spin waves. This second effect is critical. It is present even for a completely saturated sample with a uniform and \( r \)-independent static magnetization, the situation considered below. In the simple case of uniaxial grains, one can write \( \tilde{\mathcal{W}}(r, M) = -K_r(r)/M^2(\{e(r) \cdot M(r)\})^2 \), where \( K_r(r) \) represents the spatially dependent uniaxial anisotropy energy density parameter and \( e(r) \) is a unit vector that defines the grain-to-grain \( r \)-dependent uniaxial easy axis. Later in the analysis, the practical parameters of importance will be developed in terms of the components of an \( r \)-dependent \( 2 \times 2 \) effective field tensor \( \tilde{h}(r) \) and the corresponding Fourier components of these fields.

Turn now to the dynamics. For purposes of analysis, it is assumed that the sample remains magnetically saturated with a well defined static and \( r \)-independent magnetization vector specified as \( M_0 = M \cdot \hat{z} \). One also takes the dynamic magnetization, given by \( m(r,t) = [M(r,t) - M_0] \), to be small. In this limit, \( \tilde{\mathcal{W}}(r, M) \) may be expanded in a Taylor series about the magnetization equilibrium position. Taken out to second order terms only, \( \tilde{\mathcal{W}}(r, M) \) takes the form
\[
\tilde{\mathcal{W}}(r, M) = \tilde{\mathcal{W}}(r, M_0) + \nabla_M \tilde{\mathcal{W}}(r, M)|_{M_0} \cdot m(r,t) \\
+ \frac{1}{2} m^T(r,t) \cdot \nabla_M \tilde{\mathcal{W}}(r, M)|_{M_0} : m(r,t).
\]
One then proceeds to apply the same transformations developed in the previous sections to the dynamic magnetization, first in terms of the conjugate \( a(r,t) \) and \( a^*(r,t) \) variables, then in terms of the \( a_k \) and \( a^*_k \), and finally in terms of the previously diagonal \( c_k \) and \( c^*_k \).

Recall that the second order homogeneous sample Hamiltonian \( U^{(2)}_{\text{hom}} \) after transformation to the \( c_k \) and \( c^*_k \) is diagonal and leads to the spin wave dispersion and normal modes. The corresponding second order terms in the \( c_k \) and \( c^*_k \) for the transformed \( \tilde{\mathcal{W}}(r, M) \) will be nondiagonal. The additional coupling between the \( c_k \) and \( c^*_k \) modes in this case can be viewed as the basis of two magnon scattering.

The algebra is somewhat tedious but straightforward. Only a brief roadmap is provided here. The first transformation leads to a second order energy density component, written in frequency units, that takes the form
\[ \frac{1}{M_s} \tilde{\mathcal{V}}^{(2)} = A(r) a(r,t) a^*(r,t) + \frac{1}{2} [B(r) a^*(r,t) a^*(r,t) + c.c.], \]

with coefficients \( A(r) = |\gamma h_{xx} + h_{yy}(r)| / 2 \) and \( B(r) = |h_{xx}(r) + h_{yy}(r) + 2i h_{xy}(r)/2. \) The \( h_{xx}(r), h_{yy}(r), \) and \( h_{xy}(r) \) denote the spatially dependent components of the inhomogeneous stiffness field tensor \( \tilde{h}(r). \) Formally, these components are given as

\[ \tilde{h}_{xx}(r) = M_s \frac{\partial^2 \tilde{V}(r, M)}{\partial M_x^2} \bigg|_{M_0}, \]

and

\[ \tilde{h}_{xy}(r) = M_s \frac{\partial^2 \tilde{V}(r, M)}{\partial M_x \partial M_y} \bigg|_{M_0}. \]

Note that the functional forms for the new spatially dependent \( A(r) \) and \( B(r) \) functions in terms of the \( h_{xx}(r), h_{yy}(r), \) and \( h_{xy}(r) \) parallel the original homogeneous sample \( A_k \) and \( B_k \) expressions in terms of the \( H_{xx,k}, H_{yy,k}, \) and \( H_{xy,k}. \) This parallel form is not a coincidence. It is related directly to the quadratic form of the energy in both cases. This parallel structure also carries over to the equations that connect the quadratic form of the energy in both cases. This parallel form is not a coincidence. It is related directly to the internal effective fields taken as linear functionals of the vector magnetization. In contrast, the \( h_{xx}, h_{yy}, \) and \( h_{xy} \) Fourier transformed inhomogeneous stiffness field tensor components with the previously developed \( H_{xx,k}, H_{yy,k}, \) and \( H_{xy,k} \) homogeneous sample stiffness field tensor components. The sum of the development for \( H_{xx,k}, H_{yy,k}, \) and \( H_{xy,k} \) in the Appendix illustrates how these fields derive from internal effective fields taken as linear functionals of the vector coil magnetization. The \( k \) dependences for these Fourier component fields therefore reflect the nature of the interaction between the magnetic moments of the interaction with external field. In contrast, the \( h_{xx}, h_{yy}, \) and \( h_{xy} \) stiffness fields derive from the inhomogeneous energy density \( \tilde{V}(r, M), \) and their wave vector dependences reflect primarily the spatial variation in the material inhomogeneities.

Apart from the details, however, these Fourier component field terms will be large when \( |q| \) is on the order of an inverse grain size. The sizes of these terms will be on the order of the anisotropy field parameter \( 2K/\mu_s. \) At the same time, the \( \Delta_{kq-kk'}c_k \) factors in the terms in \( \tilde{U}^{(2)} \) will tend to couple specific \( k \) and \( k' \) modes that satisfy the condition \( |k-k'| \sim |q| \). This coupling is the source of the two magnon scattering.

In terms of the \( c_k \) and \( c_k^* \), the total second order Hamiltonian \( \tilde{U}^{(2)} \) takes the form

\[ \tilde{U}^{(2)} = \tilde{U}_{hom}^{(2)} + \tilde{U}^{(2)} \]

\[ = \sum_k \omega_k c_k^*(t)c_k(t) + \frac{1}{2} \sum_{kk'} [\tilde{G}_{kk'}c_k^*(t)c_{k'}(t) + c.c.], \]

The coefficients of the new \( c_k^*c_k' \) and \( c_k^*c_{k'}^* \) pair terms are given by

\[ \tilde{G}_{kk'} = \tilde{A}_{kk'} (u_k u_{k'} + v_k v_{k'}) + \tilde{B}_{kk'} (v_k u_{k'} - u_k v_{k'}), \]

and

\[ \tilde{F}_{kk'} = \tilde{A}_{kk'} (u_k v_{k'} + u_{k'} v_k) + \tilde{B}_{kk'} (v_k v_{k'} + u_k u_{k'}). \]

From the properties of the HP transformation, one can show that these coefficients satisfy the conditions \( \tilde{G}_{kk'} = \tilde{G}_{k',k}^* \) and \( \tilde{F}_{kk'} = \tilde{F}_{k',k}^*. \)

The full \( \tilde{G}_{kk'} \) and \( \tilde{F}_{kk'} \) expressions given above appear to be fairly formidable. Section II D below, however, will show that the relevant parameter for uniform mode two magnon relaxation is \( |\tilde{G}_{0,k}|^2 \). Section II E will then consider a further reduction in the form for \( |\tilde{G}_{0,k}|^2 \) based on general considerations for the spatial correlation of the Fourier components of the stiffness field tensor \( \tilde{h}(r). \) Keep in mind that the \( u_k \) and \( v_k \) are defined through the \( A_k \) and the \( B_k \) coefficients in \( \tilde{U}^{(2)} \). At the same time, the \( \tilde{A}_q \) and \( \tilde{B}_q \) track back to the Fourier components of \( \tilde{h}(r). \) For all terms, the surviving \( q \) values are specified through the Kronecker delta functions in the terms of Eq. (24), with either \( k \) or \( k' \) set to zero. This means that all of the related parameters in \( |\tilde{G}_{0,k}|^2 \) will ultimately track back to correlations between the surviving Fourier components of \( \tilde{h}(r) \), with multipliers that involve products of the homogeneous stiffness field tensor components \( H_{xx,k}, H_{xy,k}, \) and \( H_{xy,k} \) for \( k=0 \) with those for \( k \neq 0 \). A practical and fairly general working expression for \( |\tilde{G}_{0,k}|^2 \) will be given in Sec. II E. Part of the power of the Hamiltonian approach is that the needed \( |\tilde{G}_{0,k}|^2 \) for real systems can often be extracted in a relatively simple form. This will become clear from the examples considered in Secs. III and IV.
Apart from the apparent complexity of the $\tilde{G}_{k,k'}$ and $\tilde{F}_{k,k'}$, it is clear that the previously diagonal equation of motion for $c_k(t)$ is no longer diagonal. This equation of motion now takes the form
\[
i \left( \frac{d}{dt} + \eta_k \right) c_k(t) = \omega_k c_k(t) + \sum_{k'} \left[ \tilde{G}_{k,k'} c_{k'}(t) + \tilde{F}_{k,k'} c_{k'}^*(t) \right].
\] (28)

One can see that the inclusion of inhomogeneities in the analysis, in combination with a proper Hamiltonian treatment of the extra terms in the energy, leads directly to new coupling terms in the equation of motion. The main contribution to the two magnon scattering relaxation for a given $c_k$ mode is due to $\tilde{G}_{k,k'}$ terms for $|k'|$ values that are within an inverse grain size of the given $k$. If the $k$ mode of interest is the uniform mode, for example, the coupling will be to all $k'$ modes within a sphere of radius $\sim \pi/\xi$, where $\xi$ is the size of the grain or other inhomogeneity. As this analysis is applied to specific situations, the parameter $\xi$ will carry over to a correlation length for the inhomogeneity of interest.

The above relations constitute a general formulation of the linearized spin wave equations of motion in the case of local inhomogeneity scattering. Nonlocal inhomogeneity scattering will lead to the same form of the Hamiltonian but with different coupling coefficients. From the development of the equations of motion, one can also see that the analysis can be easily extended beyond the small signal limit. One simply has to take higher order terms in the inhomogeneity energy Taylor expansion into account, along with the appropriate transformations. This relatively simple and conceptually straightforward inclusion of nonlinearity is a particular advantage of the Hamiltonian method. However, in the problems associated with nonlinear magnetization dynamics in the presence of inhomogeneities, it is usually sufficient to take the inhomogeneity contribution in the linearized form along with the higher order terms from the homogeneous Hamiltonian.\textsuperscript{31,33,34}

It is possible, in principle, to diagonalize the new total Hamiltonian (25) and find the new normal modes and the corresponding eigenfrequencies. Such a procedure has been used in Refs. 17 and 18, for example, for the numerical analysis of FMR spectra in thin films with simple types of inhomogeneities characterized by randomly distributed local fields. It is found, however, that the absorption profile for weak scattering is reasonably well described by the approximate solution for the uniform mode relaxation rate that can be found from the above working equations. Uniform mode decay is considered below.

**D. Uniform mode free decay rate**

A series of simplifications allows one to solve the augmented uniform mode ($k=0$) free decay problem embodied in the above equations. The first such simplification is to ignore the secondary scattering between the nonuniform or $k \neq 0$ modes. The basic argument here is that the multiple scattering events among the $k \neq 0$ will always occur with a very short characteristic time that can be neglected in the overall sequence of scattering events.

The set of coupled equations of motion for the single $c_0(t)$ and ensemble of $c_k(t)$ amplitudes implicit in Eq. (28) may then be written in a simple form as
\[
i \left( \frac{d}{dt} + \eta_0 \right) c_0(t) = \omega_0 c_0(t) + \sum_{k \neq 0} \left[ \tilde{G}_{0,k} c_k(t) + \tilde{F}_{0,k} c_k^*(t) \right]
\] (29)

and
\[
i \left( \frac{d}{dt} + \eta_k \right) c_k(t) = \omega_k c_k(t) + \tilde{G}_{0,k} c_0(t) + \tilde{F}_{0,k} c_0^*(t), \quad k \neq 0.
\] (30)

It is to be emphasized that the $k$ parameters in the above are explicitly for $k \neq 0$ modes. The surviving coupling coefficients now involve only the $k=0$ mode and individual uncoupled $k \neq 0$ modes.

The second simplification is to seek $c_0(t)$ solutions in the adiabatic approximation. The uniform mode and $k \neq 0$ modes are taken to have the forms $c_0(t)=\bar{c}_0(t)e^{-i\omega_0 t}$ and $c_k(t)=\bar{c}_k(t)e^{-i\omega_k t}$, respectively, where the $\bar{c}_0(t)$ and $\bar{c}_k(t)$ specify slowly time varying envelope functions. The above equations may then be combined to yield a $\bar{c}_0(t)$ equation of motion of the form
\[
\frac{d\bar{c}_0(t)}{dt} = -\eta_0 \bar{c}_0(t) - \int_0^t \tilde{W}(s)\bar{c}_0(t-s)ds,
\] (31)

with a driving function $\tilde{W}(s)$ given by
\[
\tilde{W}(s) = \sum_{k \neq 0} e^{-\eta_k s} \left| \tilde{G}_{0,k} \right|^2 e^{i(\omega_k s - \omega_0 s)} - \left| \tilde{F}_{0,k} \right|^2 e^{i(\omega_k s + \omega_0 s)}. \] (32)

Under the slowly varying envelope assumption, one can neglect the retardation implicit in the $\tilde{W}(s)\bar{c}_0(t-s)$ integral term by setting $\bar{c}_0(t-s)=\bar{c}_0(t)$. The differential equation in Eq. (31) can then be solved by separation of variables. For times much longer than the inverse relaxation rate, or for $t \gg 1/\eta_0$, however, the upper integration limit may be set to $\infty$. One then obtains a relatively simple equation of motion for the envelope function $\bar{c}_0(t)$ given by
\[
\frac{d\bar{c}_0(t)}{dt} = -\left( \eta_0 + \bar{\Gamma}_0 \right) \bar{c}_0(t),
\] (33)

where $\bar{\Gamma}_0$ is given as a sum over the available nonzero $k$ states according to
\[
\bar{\Gamma}_0 = i \sum_{k \neq 0} \left\{ \frac{|\tilde{G}_{0,k}|^2}{\omega_0 - \omega_k + i\eta_k} - \frac{|\tilde{F}_{0,k}|^2}{\omega_0 + \omega_k + i\eta_k} \right\} = i\omega_0 + \bar{\eta}_0.
\] (34)

Keep in mind that the original homogeneous sample harmonic frequency response at $\omega_0$ has already been taken out of the problem through the substitution $c_0(t)=\bar{c}_0(t)e^{-i\omega_0 t}$. It is clear, therefore, that the inhomogeneities bring in two effects. First, the inhomogeneity coupling produces a shift of...
the uniform mode eigenfrequency $\omega_0$ by an amount that corresponds to the imaginary part of $\Gamma_0$, namely, $\tilde{\eta}_0$. Second, the two magnon scattering also brings in an additional contribution to the relaxation rate given by the real part of $\Gamma_0$, namely, $\tilde{\eta}_0$. Both results are tied directly to the coupling coefficients $\tilde{G}_{0,k}$ and $\tilde{F}_{0,k}$. These, in turn, connect back to the fluctuating field Fourier components $\tilde{h}_{x,x',0}$, $\tilde{h}_{y,y',0}$ and $\tilde{h}_{y,y',0}$.

Notice two further characteristics of the $\Gamma_0$ source function for the two magnon decay and frequency shift. First, the denominators for the $\tilde{G}_{0,k}$ terms are resonant. These terms are large only when $|\omega_0 - \omega_k|$ is on the order of or smaller than the intrinsic spin wave relaxation rate $\eta_k$. Loosely speaking, this means that only those modes within $\eta_k$ of the uniform mode frequency $\omega_0$ will contribute to the scattering. In quantum terms, this corresponds to energy conservation for the individual $\omega_0 \rightarrow \omega_k$ mode scattering events, apart from the caveat of broadened spin wave modes. In contrast with the $\tilde{G}_{0,k}$ term responses, the $\tilde{F}_{0,k}$ term denominators are nonresonant. Under the reasonable assumption that $|\omega_0 + \omega_k| \gg |\omega_0 - \omega_k|$ is true, one can safely neglect the contributions of the $\tilde{F}_{0,k}$ term to either the two magnon frequency shift $\tilde{\eta}_0$ or the two magnon relaxation rate $\tilde{\eta}_0$.

In the $\eta_k \rightarrow 0$ limit the inhomogeneity contribution to the relaxation rate is reduced to the form

$$\tilde{\eta}_0 = \pi \sum_k |\hat{G}_{0,k}|^2 \delta(\omega_0 - \omega_k). \quad (35)$$

This Fermi golden rule form closely matches the result one would obtain directly from the quantum formulation of spin wave scattering. The usual factor of 2 in front of the sum from the quantum analysis is absent. This is due to the fact that relaxation rate $\tilde{\eta}_0$, as defined, reflects the decay of the uniform mode amplitude $c_0$ rather than the magnon occupation number which scales with $|c_0|^2$. The delta function in Eq. (35) conveys the stipulation that the usable k modes must be degenerate with the uniform mode frequency. This degeneracy is one of the key constraints in practical calculations.

E. Randomly distributed inhomogeneities

It was noted in Sec. II C that the sought after $|\hat{G}_{0,k}|^2$ factors will all involve correlations between pairs of the surviving Fourier components of $\hat{h}(r)$ multiplied by products of the homogeneous stiffness field tensor components for $k=0$ and those with $k \neq 0$. The latter are a matter of algebra. The former require some discussion. These correlations may be expressed through a working relation,

$$\frac{1}{V} \int V \tilde{h}_{\alpha\beta}(r) \tilde{h}_{\alpha',\beta'}(r + R) d^3r$$

$$= \langle \tilde{h}_{\alpha\beta}(r) \tilde{h}_{\alpha',\beta'}(r) \rangle C(R) \quad (\alpha, \beta, \alpha', \beta' = x, y), \quad (36)$$

where $\langle \cdots \rangle$ denotes a spatial average and $C(R)$ is an appropriate correlation function. For practical purposes, the spatial average may be replaced by a configuration average.

Following Schömann, plausible choices for $C(R)$ should generally include functions that are nonzero for $|R|$ values that are on the order of or smaller than $\xi$ and zero for $|R| > \xi$, where $\xi$ denotes the size of the inhomogeneity. The condition $C(0)=1$ is true, by definition. As a rule, the particular choice for $C(R)$ does not affect the final results in any significant way. For applications to $|\hat{G}_{0,k}|^2$, one needs the Fourier transform of Eq. (36). This is given as

$$\tilde{G}_{\alpha\beta,k} \tilde{G}_{\alpha',\beta',k} = \langle \tilde{h}_{\alpha\beta}(r) \tilde{h}_{\alpha',\beta'}(r) \rangle C_k,$$

where $C_k$ is the Fourier transform of $C(R)$. Two points should be noticed here. (1) The upper cutoff wave number in $C_k$ is of the order of $|k| = \pi/\xi$. (2) The limiting value of $C_k$ in the low wave number limit will be on the order of $C_0 = V_\xi/V$, where $V_\xi$ is the mean inhomogeneity volume. Both properties may be readily obtained from the properties of the correlation function $C(R)$ and the Fourier transform.

Based on the outline given in Sec. II C and the above correlation function nomenclature, it is straightforward to obtain $|\hat{G}_{0,k}|^2$ in terms of the various parameters developed or defined above. The result is

$$|\hat{G}_{0,k}|^2 = \frac{\pi}{4|\omega_0|} \left\{ |\langle \tilde{h}_{x,x'}^2 \rangle| H_{y,y,k} H_{y,y,0} + \langle \tilde{h}_{x,x'}^2 \rangle H_{y,y,k} H_{x,x,0} \right. \right.$$

$$\left. + 2|\langle \tilde{h}_{x,y} \rangle| (\omega_0 \omega_0) \gamma^2 + H_{y,y,k} H_{x,x,0} \right. \right.$$

$$\left. + \langle \tilde{h}_{x,y}^2 \rangle H_{x,x,k} H_{y,y,0} + H_{y,y,k} H_{x,x,0} \right. \right.$$

$$\left. - 2(\omega_0 \omega_0) \gamma^2 - H_{y,y,k} H_{y,y,0} \right) \right). \quad (38)$$

Note that the $H_{x,x,k}$, etc., denote the homogeneous sample stiffness field tensor components, and these occur as products of terms for $k=0$ and $k \neq 0$, while the $\langle \tilde{h}_{x,x'}^2 \rangle$, etc., denote the spatially averaged values of the indicated inhomogeneous stiffness field tensor component products, as indicated.

The evaluation of the specific spatial averages involves only knowledge of the specific energy form of interest. By way of example, Sec. III will present expressions for the spatially averaged product terms for cubic magnetocrystalline anisotropy obtained from previous work, along with general comments on the procedure.

F. Two magnon scattering as a uniform mode frequency perturbation

Before the consideration of specific calculations based on the above working equations, it is useful to examine a relatively simple intuitive result based on spatial line width broadening considerations. From Sec. II B, the uniform mode frequency for a homogeneous sample may be written as $\omega_0 = \gamma (H_{x,x,0} H_{y,y,0} - H_{y,y,0}^2/2)$. One may now consider the inhomogeneous sample to be comprised of small grains, each with a slightly different local uniform mode frequency, specified as $\omega_0(r)$. The spatial dependence of $\omega_0(r)$ can be then obtained from the spatial variation of the uniform mode stiffness fields $H_{x,x}(r) = H_{x,x,0} + \tilde{h}_{x,x}(r)$, $H_{y,y}(r) = H_{y,y,0} + \tilde{h}_{y,y}(r)$, and $H_{y,y}(r) = H_{y,y,0} + \tilde{h}_{y,y}(r)$. The overall form of $\omega_0(r)$ will be the same as given above, but with the inhomogeneous sample stiffness fields replaced by the total stiffness fields.
\[ \omega_0(r) = |\gamma| \sqrt{\tilde{H}_{x,0}(r)\tilde{H}_{y,0}(r) - \tilde{H}_{y,0}^2(r)}. \]  

To the lowest order in the \( \tilde{h}_{x,0}(r), \tilde{h}_{y,0}(r), \) and \( \tilde{h}_{z,0}(r), \) the spatial frequency deviation from \( \omega_0, \) taken as \( \delta \omega_0(r), \) is obtained as

\[ \delta \omega_0(r) = \left[ \frac{\gamma}{2 \omega_0} \right] \left[ \tilde{H}_{x,0}(r)\tilde{h}_{x,0}(r) + \tilde{H}_{y,0}(r)\tilde{h}_{y,0}(r) - 2 \tilde{H}_{y,0}(r)\tilde{h}_{y,0}(r) \right]. \]  

One can see, therefore, that the spatial variations in the inhomogeneous sample stiffness fields yield a corresponding spatial inhomogeneous broadening in the FMR frequency. One can then take the Fourier transform of \( \delta \omega_0(r), \) denoted as \( \tilde{G}(k) \), as a coupling coefficient between \( k=0 \) and \( k \neq 0 \) magnetization modes. Indeed, in the limit of low \( k \) scattering, that is, for scattering to modes with \( k=0, \) one can show that |\( \tilde{G}(k) |^2 \) is reduced to a simple physical form, namely,

\[ |\tilde{G}(k)|^2 \approx \rho(k) \delta \omega_0(k)^2 = \langle \delta \omega_0^2 \rangle Ck. \]  

For weak, long wavelength inhomogeneities, therefore, the two magnon scattering process amounts to a frequency broadening effect ameliorated by the correlation function of the fluctuations that are the source of the scattering in the first place.

In the limit of zero intrinsic damping, one can make use of Eq. (35), apply the frequency line broadening connection, and obtain

\[ \tilde{\eta}_0 = \pi \langle \delta \omega_0^2 \rangle \sum_k C_k \delta(\omega_0 - \omega_k) = \pi \langle \delta \omega_0^2 \rangle C_{00} \delta(\omega_0), \]  

where \( \delta(\omega_0) = \sum_k \delta(\omega_0 - \omega_k) \) is the total density of states for the low \( k \) spin wave modes that are degenerate with the uniform mode. For a well characterized sample, all of the quantities in Eq. (42) are known. One has, therefore, a reasonable expression for the two magnon relaxation rate based on relatively simple physical considerations on the resonance frequency broadening.

III. ANISOTROPY SCATTERING IN A POLYCRYSTALLINE FERRITE SPHERE

This section summarizes a representative calculation of the two magnon scattering relaxation rate for a bulk [three-dimensional (3D)] insulating material. As a specific example, a dense polycrystalline ferrite sphere magnetized to saturation by a uniform static field \( H \) is considered. It is assumed that grain orientations are randomly distributed and that each grain is comprised of a single crystal with cubic magneto

crystalline anisotropy. This is the classic two magnon scattering problem first treated by Schlömann.\textsuperscript{21} As in Ref. 21, intrinsic damping will be neglected. The starting point for the analysis is Eq. (35).

The needed parameters for the isotropic sphere sample are (1) the homogeneous sample stiffness fields, \( H_{x,k}, H_{y,k}, H_{z,k}, \) and \( H_{x,k}, \) both for a general \( k \) value and for \( k=0, \) (2) the corresponding spatial averages for the inhomogeneous sample stiffness fields given by \( \langle \tilde{h}_{x,0}^2 \rangle, \langle \tilde{h}_{y,0}^2 \rangle, \langle \tilde{h}_{z,0}^2 \rangle, \) and \( \langle \tilde{h}_{x,0}\tilde{h}_{y,0} \rangle, \) and (3) the correlation function Fourier transform, \( C_k. \) For computational purposes, one also needs to convert from the \( k \) sum over degenerate spin wave modes to an integral, according to \( \Sigma_k \rightarrow V/(2\pi)^3 \int \int k^2 \sin \theta_h dk d\theta_h d\phi_h. \) This conversion is justified for typical macroscopic sample sizes.

The homogeneous sample stiffness field algebra is based on the method outlined in the Appendix. The internal effective field is comprised of the static field, uniform demagnetizing fields, exchange fields, and dipole-dipole fields with surface effects neglected. The components of the spin wave stiffness fields are obtained as

\[ H_{x,0} = H_i + Dk^2 + 4\pi M_s \sin^2 \theta_h \cos^2 \phi_h; \]  

\[ H_{y,0} = H_i + Dk^2 + 4\pi M_s \sin^2 \theta_h \sin^2 \phi_h; \]  

and

\[ H_{z,0} = 4\pi M_s \sin^2 \theta_h \sin \phi_h \cos \phi_h. \]  

In the above, \( H_i = H - 4\pi M_s / 3 \) is the internal static field and \( D \) is an exchange constant. The \( \phi_h \) and \( \theta_h \) denote the standard azimuthal and polar spin wave propagation angles, respectively, that define the direction of the wave vector \( k \) relative to the direction of the static field. The above fields combine to give a bulk sample spin wave frequency \( \omega_k \) written in the standard form as

\[ \omega_k = \sqrt{(H_i + Dk^2)(H_i + Dk^2 + 4\pi M_s \sin^2 \theta_h)}. \]  

Note that the bulk sample \( \omega_k(k) \) function contains no azimuthal angle dependence. The homogeneous sample stiffness fields for \( k=0 \) are given by

\[ H_{x,0} = H_{y,0} = H = \omega_0 / |\gamma| \]  

and

\[ H_{z,0} = 0. \]  

Note that the FMR frequency \( \omega_0 \) for a spherical sample is just equal to \( |\gamma| H. \) The spatial averages for the inhomogeneous sample stiffness fields \( \langle \tilde{h}_{x,0}^2 \rangle, \langle \tilde{h}_{y,0}^2 \rangle, \langle \tilde{h}_{z,0}^2 \rangle, \) and \( \langle \tilde{h}_{x,0}\tilde{h}_{y,0} \rangle \) involve an analysis based on the cubic magneto

crystalline energy density as developed in general terms in Sec. II C. To the lowest order, cubic symmetry leads to an anisotropy energy density of the form \( \tilde{W} = K_1 (\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2), \) where \( K_1 \) is the usual first order magneto

crystalline anisotropy energy density parameter and the \( \alpha_{X,Y,Z} \) denote the direction cosines between the magnetization vector \( M(r) \) and the local cubic \( X, Y, \) and \( Z \) axes in the random crystalline grains. It is useful to define a cubic anisotropy field parameter \( H_{y} = 2K_1 / M_s. \) The extraction of the spatial average quantities \( \langle \tilde{h}_{x,0}^2 \rangle, \langle \tilde{h}_{y,0}^2 \rangle, \langle \tilde{h}_{z,0}^2 \rangle, \) and \( \langle \tilde{h}_{x,0}\tilde{h}_{y,0} \rangle \) is a matter of (1) a careful analysis of energy derivatives for \( \tilde{W} \) specified in Sec. II C and (2) proper spatial averaging. McMichael and Krivosik gave the details.
of such an analysis for a variety of inhomogeneities in Ref. 14. The needed spatial averages for cubic anisotropy are obtained as
\[ <\tilde{h}_{xx}^2> = <\tilde{h}_{yy}^2> = \frac{29}{105} H_a^2, \tag{49} \]
\[ <\tilde{h}_{xy}^2> = \frac{9}{105} H_a^2, \tag{50} \]
and
\[ <\tilde{h}_{xx}\tilde{h}_{yy}> = \frac{11}{105} H_a^2. \tag{51} \]
Note that all terms involve the square of the anisotropy field multiplied by some numerical factor. Other types of anisotropy generally lead to the same form, but with different numerical factors.

Finally, turn to the correlations. A relatively standard choice for the correlation function \( C(R) \) is
\[ C(R) = e^{-|R|/\xi}, \tag{52} \]
where \( \xi \) is now a characteristic correlation length that may be associated with the grain size in the polycrystalline sample, for example. The Fourier transform of Eq. (52) yields
\[ C_k = C_k = \frac{2\pi^2 f(k)}{V \eta^2}, \tag{53} \]
with
\[ f(k) = \frac{4}{\pi} \frac{\xi^4 k^2}{1 + (k\xi)^2}. \tag{54} \]
The \( f(k) \) nomenclature follows the notation of Ref. 21. Note that \( f(k) \) is a steeply peaked function with its maximum at \( k = 1/\xi \). In wave number space, \( f(k) \) will select out only those Fourier components around this peak position for a strong contribution to the scattering.

The stage is now set for the evaluation of the two magnon relaxation rate from Eq. (35). With the sum over \( k \) replaced by a volume integral over \( k \) space as given above, use of the prescribed homogeneous stiffness fields and the spatially averaged inhomogeneous stiffness field products just listed, the final result is obtained as
\[ \tilde{\eta}_0 = \frac{\pi^2 H_a^2}{105} \int_0^\infty dk \int_0^1 d \cos \theta f(k) \]
\[ \times \left[ 1 + 19 \frac{|\gamma|}{\omega_0} \left( H_1 + Dk^2 + 2\pi M_s \sin^2 \theta_k \right) \right] \]
\[ \times \delta(\omega_0 - \omega_k). \tag{55} \]
Keep in mind that even though this is a 3D problem, the \( |G_{0,k}| \) is independent of the azimuthal spin wave propagation angle \( \phi_k \). The result in Eq. (55) matches the original result given by Eq. (47b) in Ref. 21. This result has formed much of the basis of the linewidth analysis for dense bulk polycrystalline ferrites from the 1960s. The value of the present rendering of this result is in the step by step and relatively transparent development directly from Hamiltonian principles.

### IV. TWO MAGNON SCATTERING LINEWIDTH IN THIN FILMS

Ferromagnetic resonance in a variety of forms is widely used to determine the uniform mode relaxation or damping rate through linewidth measurements for thin magnetic films in general and metallic films, in particular. It has been well known since the late 1960s, moreover, that two magnon scattering can make significant contributions to these linewidths, as in Ref. 6, for example. In spite of this, the predominant popular approach for most workers has been simply to analyze all data on the basis of a single phenomenological damping parameter based Landau-Lifshitz or Gilbert damping model. While there have been notable exceptions to this phenomenological approach for data analysis, mainly through the working equations from the Arias and Mills theory, as provided in Refs. 8, 27, and 49, for example, there has really been no cogent application of the two magnon theory to a wide range of thin film FMR linewidth data. The purpose of this section is to provide such an application based on the equations developed above and the long standing linewidth data from Ref. 6 for a range of Permalloy film thicknesses and FMR frequencies. These data were obtained for in-plane static fields.

The data from Ref. 6 are analyzed in terms of a simple model that includes three contributions to the relaxation rate: intrinsic magnon-electron scattering, eddy current Ohmic losses, and two magnon scattering. The magnon-electron scattering contribution was analyzed in terms of Gilbert damping.50,51

Within the framework of the Gilbert model, the intrinsic contribution to the uniform mode relaxation rate can be written as
\[ \eta_0, \text{m}= \alpha_G \gamma_0 P_A, \tag{56} \]
where \( \alpha_G \) is the Gilbert damping parameter. The \( \alpha_G \) parameter can, in principle, be obtained in terms of the band structure, as discussed in Ref. 50 and elsewhere. For the present purposes, \( \alpha_G \) will be used simply as an adjustable parameter. The \( P_A \) factor is related to the uniform mode ellipticity and can be expressed as
\[ P_A = \frac{|\gamma|}{2\omega_0} \left( H_{xx,0} + H_{yy,0} \right) = \left. \frac{\partial \omega_0}{\partial \gamma H} \right|_{\text{FMR}}, \tag{57} \]
where \( H \), as in the last section, is the external static field and the derivative is evaluated at the resonance point. Note that the second equality in Eq. (57) is valid only if the change in static field \( H \) does not affect the magnetization static equilibrium. In such a case the \( P_A \) factor serves also as a conversion factor between the relaxation rate and the field linewidth.52,53

If the \( x \) and \( y \) directions are taken to be perpendicular and parallel to the film plane, respectively, one has \( H_{xx,0} = H + 4\pi M_s \) and \( H_{yy,0} = H \). The off-diagonal \( H_{xy,0} \) stiffness field component for this geometry is zero. The films in Ref. 6 had the usual in-plane uniaxial anisotropy that is typical of field deposited Permalloy films. This has not been included in the...
analysis here. Trial calculations that include the anisotropy show no appreciable effect.

Eddy current losses can be particularly pronounced for thicker films and higher frequencies. For a negligible exchange interaction and small dissipation, the eddy current relaxation rate can be written in the same form as Gilbert damping given by Eq. (56) with \( \alpha_G \) replaced by \(^{54,55}\)

\[
\alpha_{ec} = \frac{\pi |\gamma|^2 \pi M_s d^2}{3c^2 \rho}.
\]

(58)

Here, \( \rho \) is the electrical resistivity, \( d \) is the film thickness, and \( c \) is the speed of light.

The final and critical contribution to the relaxation rate and the FMR linewidth for this discussion is the two magnon contribution due to inhomogeneities, based on the formalism in Sec. II. For in-plane magnetized high magnetization thin films and relatively low frequency ferromagnetic resonance, the degenerate nonuniform modes have relatively low \( k \) values. In this case, the approximations discussed in Sec. II F are applicable.\(^{27}\)

If one further assumes that the inhomogeneous stiffness fields do not differ significantly from each other, one can write \( \langle \vec{h}^2 \rangle = \langle \vec{h}_x^2 \rangle = \langle \vec{h}_y^2 \rangle \). In this case Eq. (42) is simplified to

\[
\bar{\alpha}_h = \pi |\gamma|^2 |\langle \vec{h}^2 \rangle| C_0 P_A^2 \delta(\omega_0).
\]

(59)

For a very thin film, moreover, the dynamic magnetization does not vary significantly across the film thickness. In this uniform magnetization mode approximation, the calculation of the density of states function \( g(\omega_0) \) may be reduced to a two dimensional \( k \)-space formulation for in-plane propagating modes and may be rewritten as

\[
g(\omega_0) = \sum_k \delta(\omega_0 - \omega_k) \approx \frac{A}{(2\pi)^2} \int \delta(\omega_0 - \omega_k) d\vec{k},
\]

(60)

where \( A \) is the film area. The integration in Eq. (60) may be evaluated analytically for the ultrathin film limit.\(^{14,16}\) In such a case the result does not depend on the film thickness. Here, however, numerical integration has been used, with the full spin wave dispersion \( \omega_k \) taken in the uniform magnetization mode approximation.\(^{14,42}\) This dispersion can be expressed through \( \omega_k = |\gamma| \sqrt{H_{xx,k} H_{yy,k}} \) with the homogeneous spin wave stiffness fields given as

\[
H_{xx,k} = H + Dk^2 + 4\pi M_s N_k(kd)
\]

(61)

and

\[
H_{yy,k} = H + Dk^2 + 4\pi M_s \sin^2 \theta_k [1 - N_k(kd)].
\]

(62)

Here, \( \theta_k \) is now the angle between the in-plane spin wave \( \vec{k} \) vector and the direction of the external static field \( H \). The ultrathin film dipole field function \( N_k \) is given by \( N_k = (1 - e^{-kd})/kd \). Finally, a two dimensional Fourier transform of the correlation function given by Eq. (52) in the \( k = 0 \) limit yields \( C_0 \approx 2A_f / A \), where \( A_f \) is a mean inhomogeneity area.

Based on the above working equations, the total relaxation rate may be written as

\[
\eta_0 = (\alpha_G + \alpha_{ec}) \omega_0 P_A + K P_A^2 \int \delta(\omega_0 - \omega_k) d\vec{k}.
\]

(63)

where \( K = 0.16 |\gamma|^2 \langle \vec{h}^2 \rangle A_f \). Note that the specific numerical factor in the parameter \( K \) will depend on the choice of correlation function. Other than this, the form above for the total relaxation rate can be taken as fairly general, subject to the limits already noted.

The field swept FMR linewidth \( \Delta H \) is then given as

\[
\Delta H(\omega_0, d) = 2 \eta_0 \frac{1}{|\gamma| P_A} + 2K P_A \int \delta(\omega_0 - \omega_k) d\vec{k}.
\]

(64)

Equation (64) has been written in such a form as to keep all dependences on the FMR frequency \( \omega_0 \) and film thickness \( d \) explicit.

Figures 1 and 2 summarize the field linewidth data from the published graphs in Ref. 6 for a series of Permalloy films, along with corresponding fit results based on the above working equations. Graph (a) in Fig. 1 shows the half power field swept linewidth \( \Delta H \) as a function of film thickness \( d \) for the indicated frequencies. The various symbols show the data and the solid curves show the fits. Graph (b) shows the data and fit results at 7 GHz, along with a breakdown of the component eddy current (ec), magnon-electron (m-e), and two magnon (m-m) contributions to the linewidth, as indicated.

The fitting was done in a general way, with common material and structure parameters used for all the films. The focus was on the realization of a reasonable overall fit that adequately demonstrates the overall trend of the data rather than a point-by-point matchup. The curves shown were ob-

\[\text{FIG. 1. Field swept linewidth } \Delta H \text{ as a function of film thickness } d \text{ for a series of } Ni_7Fe_{23} \text{ thin films. Graph (a) shows data from Ref. 6 at specified frequencies indicated by the various symbols. The solid lines represent fits to the data with fitting parameters as given in the text. Graph (b) shows data and fit results at 7 GHz, along with a breakdown of the component eddy current (ec), magnon-electron (m-e), and two magnon (m-m) contributions to the linewidth, as indicated.}\]
in the density of states for the two magnon scattering. The calculational two magnon component with more complicated frequency and quadratically with film thickness. Both the eddy current damping and magnon-electron damping of, in particular, the small but distinct $m$-$m$ components in the graphs clearly as one might like, but the overall trend, especially in Fig. 2.

The data and fit results for the 320 nm thick film along with a breakdown of the component eddy current (ec), magnon-electron (m-e), and two magnon (m-m) contributions to the linewidth, as indicated.

One can see that the fits generally replicate the trends of the data in both graphs. Keep in mind that the combination of eddy current damping and magnon-electron damping alone would give only a linewidth that scales linearly with frequency and quadratically with film thickness. Both the data and the fits provide convincing evidence for an additional two magnon component with more complicated $d$ and $f$ dependences. These dependences come from the variation in the density of states for the two magnon scattering. The breakdowns in Figs. 1(b) and 2(b) show these effects. Note, in particular, the small but distinct $m$-$m$ components in the (b) graphs in both figures. The scatter in the data in Fig. 1 is too large to see the two magnon effect on the response as clearly as one might like, but the overall trend, especially in graph (a), provides reasonably convincing evidence of such a response. The two magnon knee can be discerned somewhat more clearly in Fig. 2.

Considered overall, the data, the fits, and the breakdowns demonstrate fairly clearly the role of the two magnon scattering contribution to the Permalloy film in the linewidth. In spite of the fairly basic simplified model used here, mainly for the sake of simplicity of analysis, the quantitative as well as qualitative match of the theory to the entire ensemble of data for a single set of fitting parameters is remarkable.

V. SUMMARY AND CONCLUSIONS

In summary, this work presents a compact but complete synopsis of two magnon scattering theory done within the framework of the Hamiltonian formalism. It provides general expressions for the inhomogeneity coupling coefficients in the case of localized inhomogeneities. The key ideas may be extended to nonlocal scattering without any modification in the formalism. At the present stage, the theory is applicable to problems where the nonuniform magnetization excitations can be described as plane wave modes. In principle, other types of normal mode expansions could be used in place of the plane waves, but such modifications will, by necessity, significantly expand the amount of algebra. For practical purposes, the plane wave approach as presented appears to be adequate for most material systems of technological interest.

By way of example, two specific applications of the theory are presented. The first involves a representative calculation of the two magnon scattering relaxation rate for anisotropy scattering in bulk polycrystalline spheres. It is shown that the result obtained from the Hamiltonian approach matches exactly the classic solution from Ref. 21. In the second example, previously published FMR linewidth data versus film thickness and frequency for thin NiFe films have been analyzed on the basis of the working TMS equations developed here from the Hamiltonian formalism. It is shown that the theory provides results that are in reasonable agreement with the data.

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APPENDIX

This appendix provides a roadmap for the evaluation of homogeneous stiffness field tensor introduced in Sec. II B. The analysis starts from the magnetic energy $\mathcal{H}$ written in the form

$$\mathcal{H} = -\int_V \mathbf{M}(r,t) \cdot \mathbf{H}_{\text{ex}}(r,t) d^3r$$

$$-\frac{1}{2} \int_V \mathbf{M}(r,t) \cdot \mathbf{H}_M(r,t) d^3r.$$  \hspace{1cm} (A1)

Here, the first term represents the Zeeman energy that derives from the interaction of the magnetization with an external magnetic field $\mathbf{H}_{\text{ex}}(r,t)$. The second term represents the interaction energy of the magnetization with the separate and distinct internal magnetic self-field $\mathbf{H}_M(r,t)$. This field is assumed to be a linear functional of the magnetization $\mathbf{M}(r,t)$. Examples of such internal fields include the Maxwellian dipole field, the non-Maxwellian macroscopic exchange field, and the non-Maxwellian effective field associated with a uniaxial anisotropy. Other self-fields that are given by higher order functionals of the magnetization, such...
as an effective field due to cubic anisotropy, for example, may be incorporated into a linearized form through an appropriate Taylor expansion. It is important to keep in mind that the magnetization response in the small signal limit is fully described by a Hamiltonian that contains powers of the magnetization up to the second order alone.

For simplicity, take $H_{\text{ext}}(r,t) = H \hat{z}$ as a spatially homogeneous static field. Within the plane wave approximation, the spatial dependences of $M(r,t)$ and $H_{\text{ext}}(r,t)$ in Eq. (A1) can be expanded in the Fourier series according to

$$M(r,t) = \sum_k M_k(t) e^{i \mathbf{k} \cdot \mathbf{r}}, \quad (A2)$$

and

$$H_{\text{m}}(r,t) = \sum_k H_{\text{mk}}(t) e^{i \mathbf{k} \cdot \mathbf{r}}. \quad (A3)$$

From the linearity of the Fourier transform, it follows that the Fourier components $H_{\text{mk}}$ will depend linearly on $M_k$. For a spatially homogeneous sample one can write this dependence as

$$H_{\text{mk}}(t) = -4\pi \hat{N}_k \cdot M_k(t), \quad (A4)$$

where $\hat{N}_k$ is a $3 \times 3$ spin wave tensor. The factor $-4\pi$ is chosen to give a form for $H_{\text{mk}}$ that resembles expressions for the demagnetization fields of a homogeneously magnetized ellipsoid. The components of the $\hat{N}_k$ tensor, namely, $N_{ijk}(i,j=x,y,z)$, depend on the specific type of the internal magnetic field. If the form of the energy is known, these components can be readily evaluated. For example, for exchange, one has a simple functional for the corresponding effective exchange field, $H_{\text{exch}}(r,t) \propto \nabla^2 M(r,t)$. This corresponds, in turn, to a spin wave tensor $\hat{H}_{\text{exch}} \propto k^2 \mathbf{I}$, where $\mathbf{I}$ is a unitary matrix. Insertion of Eqs. (A2)–(A4) into Eq. (A1) yields a homogeneous sample magnetic energy of the form

$$\mathcal{H}_{\text{hom}} = -VHM_{z,0} + V \frac{4\pi}{2} \sum_k M_k^T \cdot (N_k \cdot M_k). \quad (A5)$$

Since the energy in Eq. (A5) is real, one can show that $N_{ijk}^* = N_{jik}$. This quadratic part of the magnetization can be eliminated from Eq. (A5) by taking a small signal limit approximation, $M^* = M - (M_x^2 + M_y^2)/2M_z$. The quadratic part of the energy in Eq. (A5) then terms that are quadratic in $M_{x,k}$ and $M_{y,k}$. This quadratic part of the energy can be written as

$$\mathcal{H}_{\text{hom}}^{(2)} = \frac{V}{2M_z^2} \sum_k \left( M_{x,k}^* M_{y,k}^* \right) \left( \begin{array}{cc} H_{x,k} & H_{y,k} \\ H_{y,k} & H_{y,k} \end{array} \right) \left( \begin{array}{c} M_{x,k} \\ M_{y,k} \end{array} \right), \quad (A6)$$

where the components of the homogeneous stiffness field tensor are given as

$$H_{jj,k} = H_j + 4\pi M_j N_{j,j,k} (jj = xx, yy) \quad (A7)$$

and $H_{ij,k} = 4\pi M_j N_{ij,k}$ is the internal static field. As a final step, the expression in Eq. (A6) can be transformed into the form in Eq. (12) by means of the connections in Eq. (5) taken in the small signal limit according to $i M_{x,k} + M_{y,k} = \sqrt{2} M_{\alpha,k}$.