Microscopic relaxation mechanisms and linear magnetization dynamics

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Linear magnetization dynamics in the presence of a thermal bath is analyzed for two general classes of microscopic damping mechanisms. The resulting stochastic differential equations are always in the form of a damped harmonic oscillator driven by a thermal field. The damping term contains both the interaction mechanisms and the symmetry of the magnetic system. Back transformation from the oscillator coordinates to the magnetization variables results in a macroscopic tensor form of damping that reflects the system anisotropy. Scalar Landau-Lifshitz-Gilbert damping term is valid only for systems with axial symmetry. Analysis of FMR linewidth measurements versus frequency, temperature, and film thickness in NiFe films shows good agreement with a combination of slow-relaxing impurity and magnon-electron confluence processes.

76.50.+g, 75.70.Ak, 75.30.Hx

I. INTRODUCTION

The study of classical magnetization dynamics of fine ferromagnetic particles and thin films is of great interest in connection with the rapid progress of nanotechnologies and high-density magnetic recording. The most important research problems in this field include: (a) the problem of magnetization reversal from the former equilibrium direction to a new one under an applied reversal magnetic field, (b) magnetization escape over a barrier from one equilibrium direction to another one under thermal agitation, and (c) the effect of magnetization thermal fluctuations on the noise characteristics of magnetoresistive devices. All these problems depend on relaxation properties of the system, i.e., detailed mechanisms of magnetization interaction with other degrees of freedom (spin waves, elastic vibrations, conduction electrons, impurities, etc.) of material. Previously substantial effort has been focused on the study of mostly phenomenological magnetization relaxation (e.g., [1], [2], [3], [4], [5]).

The aim of this paper is to analyze linear magnetization dynamics in the presence of a thermal bath for two general classes of microscopic damping mechanisms. This analysis will be restricted to low-level excitations where the magnetization remains close to equilibrium. Recently, magnetization relaxation by direct coupling processes have been analyzed for the case of a general anisotropic magnetic system [6]. The resulting stochastic differential equation obtained was in the form of a simple damped harmonic oscillator driven by a random thermal fluctuating field. The coordinate system of this oscillator (or, normal mode) reflected the magnetic anisotropy of the system. Although specific damping mechanisms were not analyzed, the damping parameter was equivalent to the relaxation time. Transformation back to magnetization variables yielded macroscopic dynamics where the damping term reflected the system symmetry in a tensor form [8], [9], [10].

Here this analysis is expanded to examine generally both direct and indirect mechanisms for coupling to a thermal bath. In all cases a stochastic differential equation in the form of a damped harmonic oscillator with random forces is obtained (and the corresponding tensor form of damping, as well). In addition, two specific relaxation mechanisms are examined in detail: the direct process of magnon-electron confluence and the indirect process of ‘slow-relaxing’ impurity. It is shown that these two processes in combination can explain experimental measurements of ferromagnetic resonance (FMR) linewidth in permalloy thin films. Using reasonable estimates for the parameters associated with these mechanisms, very good fitting is obtained to the linewidth dependencies on temperature, frequency and film thickness.

A phenomenological tool to study magnetization relaxation, which has had widespread use, is the Landau-Lifshitz equation [11] or, its modification with Gilbert damping [12]. These (LLG) equations conserve the absolute value of the magnetization (|M| = const) in a single domain. They are relatively simple and therefore have been utilized for various calculations and micromagnetic simulations. In particular, these equations have been used to analyze the frequency dependence of FMR linewidth measurements in permalloy films (e.g., [1], [2]). Although, this data can be fit using the LLG equations (with a constant, phenomenological damping α), this fact can not be considered as an experimental verification for validity of the general nature of the LLG approach. As we show in detail, the general tensor form of the macroscopic equation reduces to the LLG form only for axially symmetric systems.

The paper is organized as follows. In the Sec. II we formulate the basic gyromagnetic motion in a rotating frame and demonstrate that gyromagnetic magnetization dynamics is equivalent to the motion of a harmonic oscillator. Physics of a damped oscillator by direct coupling
to a thermal bath is reviewed and expanded in Sec. III. In Sec. IV the general mechanism of indirect coupling is analyzed. An overview of magnetization dynamics with a brief comparison to the phenomenological LLG form is given in Sec. V. Experimental FMR data for NiFe films are analyzed in Sec. VI. It is shown that relaxation processes due to magnetization interactions with slow-relaxing impurities and conduction electrons fit the data well. A discussion is given at the end of the paper.

II. FORMULATION OF BASIC GYROMAGNETIC MOTION IN ROTATING FRAME

A. Small magnetization motion

Let us consider small-amplitude magnetization motions of a single-domain ferromagnetic particle in the vicinity of equilibrium state \( \mathbf{M} = \mathbf{M}_0 \). Here \( \mathbf{M}_0 \) is the unit vector in the equilibrium direction and \( M_s \) is the saturation magnetization. The magnetization rotation around effective field in this case, in general, is elliptical and the magnetic energy can be represented as a quadratic form (e.g., \([13], [14]\)):

\[
\mathcal{E}/V = \frac{H_{x_0} M_s^2}{2M_s} + \frac{H_{y_0} M_s^2}{2M_s}.
\]

(1)

Here \( \mathbf{x}_0 \) and \( \mathbf{y}_0 \) are the unit orthogonal vectors in the plane perpendicular to the equilibrium direction. \( H_{x_0} \) and \( H_{y_0} \) are positive fields, which include both microscopic and shape anisotropies and the external magnetic field and \( V \) is the particle volume (see Appendix I for an example).

B. Normal mode approach

We begin with a common normal mode approach (e.g., \([13], [14]\)), where the small oscillations of the transverse magnetization coordinates are first transformed to “rotating” complex variables by a linearized Holstein-Primakoff transformation \([13]\):

\[
M_{x_0} \simeq -\sqrt{\hbar \gamma M_s/2V}(a^* + a),
\]

\[
M_{y_0} \simeq i\sqrt{\hbar \gamma M_s/2V}(a^* - a).
\]

(2)

The magnetic energy \([1]\) now can be rewritten in the quadratic form:

\[
\mathcal{E}/\hbar = Aa^* a + (B/2)(aa + a^* a^*),
\]

(3)

where \( A = \gamma (H_{x_0} + H_{y_0})/2 \) and \( B = \gamma (H_{x_0} - H_{y_0})/2 \). This classical transformation yields a form, common to quantum mechanics, where \( a \) and \( a^* \) correspond to creation and annihilation operators so that in \([8]\) the term \( a^* a = 1 - M_{x_0}^2/M_s \). Note that for an isotropic system where \( H_{x_0} = H_{y_0} \), the term \( B \) vanishes.

The dynamic precession equations are given by

\[
da/dt = -iAa - iB a^*, \quad da^*/dt = iA a^* + iB a.
\]

(4)

The mixed terms in \([8]\) can be eliminated by the linear canonical transformation:

\[
a = uc + vc^*, \quad a^* = uc^* + vc,
\]

\[
\begin{align*}
 u &= \sqrt{A + \omega_0/2\omega_0}, \quad v = -B/|B| \sqrt{A - \omega_0/2\omega_0}.
\end{align*}
\]

The magnetization components in terms of \( c \) and \( c^* \) can be expressed as

\[
M_{x_0} \simeq - (\hbar \gamma M_s/2V)^{1/2} (H_{y_0}/H_{x_0})^{1/4} (c^* + c),
\]

\[
M_{y_0} \simeq i (\hbar \gamma M_s/2V)^{1/2} (H_{x_0}/H_{y_0})^{1/4} (c^* - c).
\]

With this transformation the energy in terms of the normal mode coordinates is simply:

\[
\mathcal{E}/\hbar = \omega_0 c^* c,
\]

(7)

where

\[
\omega_0 = \sqrt{A^2 - B^2} = \gamma \sqrt{H_{x_0} H_{y_0}}.
\]

(8)

is the ferromagnetic resonance (FMR) frequency. The dynamic equations in terms of \( c \) and \( c^* \) are now independent:

\[
dc/dt = -i\omega_0 c, \quad dc^*/dt = i\omega_0 c^*.
\]

(9)

In order to derive a damped motion for this oscillator it is necessary to consider the interaction with a thermal bath.

III. DIRECT COUPLING TO A THERMAL BATH

Linear relaxation can be most simply described in the thermal bath approximation which represents coupling to a reservoir in thermodynamic equilibrium. The thermal bath model has the following restriction: the interaction with a thermal bath is assumed to be weak. Thus, dynamics of a single-domain ferromagnetic particle in the zeroth (no thermal bath) approximation represents the gyromagnetic rotation of magnetization around an effective field. This means that the symmetry of the gyromagnetic motion is dominant in the system and imposes some restrictions for damping and fluctuations. Mathematical methods for a dynamic system interacting with a thermal bath have been developed in detail in quantum optics (see, e.g., \([13], [17], [18], [19]\)). Here we apply these methods for magnetic particle dynamics.
A. Direct linear coupling with a thermal bath

We review and consider classically a general linear interaction of a harmonic oscillator with a set of harmonic oscillators that represent the thermal bath. The Hamiltonian for small oscillations \( \{ a^*, a \} \) (see, ) interacting with harmonic oscillators \( \{ b_k^*, b_k \} \) has the form:

\[
\mathcal{H}/\hbar = \mathcal{E}/\hbar + \sum_k \omega_k b_k^* b_k + \sum_k [G_k(ab_k^* + c.c.) + F_k(ab_k + c.c)].
\]  

(10)

The first term is the energy of the magnetic system, the second term gives the oscillations of the thermal bath with coordinates (modes) \( b_k \) and \( b_k^* \), and the last term gives a direct interaction coupling. The last term, if transformed back to the magnetization variables, can be represented as a scalar product \( \mathbf{M} \cdot \mathbf{H}_b \), where \( \mathbf{H}_b \) is an effective field from the thermal bath.

Applying the transformation , we can write the dynamic equations for \( c \) and \( b_k \) as

\[
dc/dt = -i\omega_0 c - i \sum_k (\bar{G}_k b_k + \bar{F}_k b_k^*),
\]  

\[
db_k/dt = -i\omega_k b_k - i\bar{G}_k c - i\bar{F}_k c^*.
\]  

(11)

(12)

Here \( \bar{G}_k = G_k u + F_k v \) and \( \bar{F}_k = G_k v + F_k u \). The terms \( F_k b_k^* \) in (11) and \( i\bar{F}_k c^* \) in (12) vanish as fast oscillating terms (they describe rotations in the complex plane in the opposite direction to \( c \) and \( b_k \), respectively) and therefore can be omitted (see, for details). A solution of (12) is:

\[
b_k(t) = b_k(0)e^{-i\omega_k t} - i \int_0^t \bar{G}_k c(t')e^{-i\omega_k (t-t')}dt'.
\]  

(13)

Substituting (13) into (11), yields the integro-differential equation for \( c(t) \):

\[
dc/dt = -i\omega_0 c - \sum_k |\bar{G}_k|^2 \int_0^t c(t')e^{-i\omega_k (t-t')}dt' + f(t).
\]  

(14)

Here

\[
f(t) = -i \sum_k \bar{G}_k b_k(0)e^{-i\omega_k t}
\]  

(15)

describes a noise: \( b_k(0) \) are random (and in thermal equilibrium). A solution of (14) can be represented as \( c(t) = \tilde{c}(t) \exp(-i\omega_0 t) \), where \( \tilde{c}(t) \) is a “slow” variable. Neglecting the memory for the slow variable \( \tilde{c}(t') \to \tilde{c}(t') \), we can rewrite Eq. (14) as:

\[
dc/dt = -i\omega_0 c - c \sum_k |\bar{G}_k|^2 \int_0^t e^{i(\omega_0 - \omega_k)(t-t')}dt' + f(t).
\]  

(16)

For a long time \( t \gg 1/(\omega_0 - \omega_k) \) one can put the upper integral limit at \( \infty \) and use the formula:

\[
\int_0^\infty du \exp(i\Omega u) = \pi \delta(\Omega) + i\text{P.v.} \left( \frac{1}{\Omega} \right),
\]  

(17)

where \( \delta(\Omega) \) is the Dirac delta function and P.v. is the principal value of the integral. Thus, we obtain the dynamic equation of a damped harmonic oscillator:

\[
dc/dt + \eta c = -i(\omega_0 + \Delta \omega)c + f(t),
\]  

(18)

where

\[
\eta = \pi \sum_k |\bar{G}_k|^2 \delta(\omega_0 - \omega_k),
\]  

(19)

\[
\Delta \omega = -\text{P.v.} \sum_k \frac{|\bar{G}_k|^2}{\omega_0 - \omega_k}.
\]

The damping parameter \( \eta \) represents a particular form of “Fermi’s Golden Rule” (e.g., ). For example, has been used for calculating two-magnon scattering and magnon-phonon processes.

B. Arbitrary coupling to a thermal bath

Lax has developed the most general approach for a dynamic description of a harmonic oscillator interacting directly with a thermal bath (reservoir). He used the following Hamiltonian:

\[
\mathcal{H}_{\text{total}} = \hbar \omega_0 \hat{c}^\dagger \hat{c} + (\hat{c}^\dagger \hat{F} + \hat{c} \hat{F}^\dagger) + \mathcal{H}_R,
\]  

(20)

where \( \hat{c}^\dagger \) and \( \hat{c} \) are creation and annihilation Bose operators, \( \hat{F} \) and its Hermitian conjugate \( \hat{F}^\dagger \) describe thermal bath variables, \( \mathcal{H}_R \) is the reservoir Hamiltonian. Taking the density matrix equation (i.e., one of the most fundamental equations in physics), Lax derived the master equation in the second approximation on interaction and finally showed that the damped harmonic oscillator should obey the following classical Langevin equation:

\[
dc/dt + \eta c = -i(\omega_0 + \Delta \omega)c + f(t).
\]  

(21)

Here \( c = \langle \hat{c} \rangle \) is the complex amplitude (classical analog of annihilation operator) and \( f(t) \) is a noise. The damping \( \eta \) and frequency shift \( \Delta \omega \) are defined by

\[
\eta - i\Delta \omega \equiv -\frac{1}{\hbar^2} \int_0^\infty du e^{-i\omega_0 u} \langle (\hat{F}(0), \hat{F}^\dagger(0)) \rangle_R,
\]  

(22)

where \( \langle \ldots \rangle_R \) denotes averaging over reservoir. As before the formula (22) for elementary processes can be reduced to the “Fermi’s Golden Rule” (second order perturbation theory, e.g., ).
1. Magnon-electron scattering

As an example of magnetization relaxation due to direct coupling to the thermal bath we shall consider the interaction of magnons with conduction electrons in a ferromagnetic metal. From a microscopic point of view the most probable process is the confluence of a magnon with wave vector \( \mathbf{k} = 0 \) and energy \( \hbar \omega_0 \) and a conduction electron with wave vector \( \mathbf{k}_1 \neq 0 \) and energy \( \epsilon(\mathbf{k}_1) \) into a conduction electron with wave vector \( \mathbf{k}_2 \) and energy \( \epsilon(\mathbf{k}_2) = \epsilon(\mathbf{k}_1) + \hbar \omega_0 \). It is obvious that this process is forbidden for an ideal crystal, where the momentum conservation is valid \( \mathbf{k}_2 = \mathbf{k}_1 + 0 \). Such confluence processes exist only for non-uniform magnetic modes and demonstrate a strong \( k \)-dependence for the relaxation rate \( \langle \chi k^2 \rangle ^{[23, 24]} \).

The abovementioned confluence process can occur in the presence of defects, impurities or fluctuations which permit violation of momentum conservation in the crystal (Fig.1a). This opportunity was qualitatively discussed by Kambersky and Patton [23]. Because of its importance, we shall consider this process in detail (see Appendix B). The relaxation rate for the process can be written as:

\[
\eta_{m-e} = c_{def}(u^2 + v^2)\omega_0 \alpha_c, \tag{23}
\]

\[
\alpha_c \simeq \frac{\pi}{N^2} \sum_{\mathbf{k}} \left( \frac{\partial \epsilon(\mathbf{k})}{\partial \omega_{\mathbf{k}}} \right) \sum_{\mathbf{k'}} |f_{\mathbf{k}\mathbf{k'}c}|^2 \delta(\omega_{\mathbf{k'}} - \omega_{\mathbf{c}}).
\]

This relaxation rate is linear with the defect concentration \( c_{def} \) and frequency dependent: it contains transformation terms \( (u^2 + v^2)\omega_0 = \gamma(\hbar \omega_0 + H_{xy})/2 \). The temperature dependence can occur through the term: \( \sum_{\mathbf{k}} (-\partial \epsilon(\mathbf{k})/\partial \omega_{\mathbf{k}}) \). An estimate of Eq.(23) for \( f_{\mathbf{k}\mathbf{k'}c} = f = \text{const} \) and \( T \ll \epsilon_F/k_B \sim 3 \cdot 10^4 \text{K} \) gives

\[
\eta_{m-e} \simeq \frac{c_{def}}{16\pi^3} \gamma(\hbar \omega_0 + H_{xy}) m^3 \nu^2 |f|^2 \epsilon_F, \tag{24}
\]

where \( m \) is the conduction electron mass and \( \epsilon_F \) is the Fermi energy.

IV. RELAXATION VIA AN INTERMEDIATE DAMPED DYNAMIC SYSTEM

There is a whole class of relaxation mechanisms that can not be analyzed as elementary processes. The energy loss in this case occurs via intermediate damped dynamic system. Here we shall consider the magnetization damping via so-called, 'slow relaxing' impurities (see, Fig.1b and Refs. [25, 26, 27, 28, 29]). In this mechanism the magnetization motion modulates the impurity splitting (levels, see, Fig.1b). Thus the thermal equilibrium population of the energy levels varies and transitions between the levels occur (arrows in Fig.1b). There is a delay for these transitions due to a finite impurity relaxation time. This delay results in a magnetization oscillation energy loss.

A general Hamiltonian describing the impurity level modulation is given in Appendix C, Eq.(C2). In the case of only the coherent motion of the host spins we have \( S(\mathbf{R}_j + \mathbf{r}_\nu) = (V_0/\hbar \gamma) \mathbf{M} \), where \( V_0 = V/N \) is the volume of elementary cell. Let us consider a two-level paramagnetic impurity with the energy

\[
H_{imp,j} = \hbar [\Omega_{0,j} + \delta \Omega_j(t)]n_j, \tag{25}
\]

where \( \Omega_{0,j} \) is the splitting frequency, \( n_j = s_{zj}(\mathbf{R}_j) + 1/2 \) is the upper lever population and \( j \) is the impurity index. Applying Eqs.(6) to (C2), we can write the impurity level modulation as

\[
\delta \Omega_j(t) = \Phi_j c(t) + \Phi_j^* c^*(t), \tag{26}
\]

where

\[
\Phi_j = \frac{V_0}{\hbar} \left( \frac{M_k}{\hbar \gamma V} \right)^{1/2} \sum_{\nu} \left[ B_{zj,x0}(\mathbf{R}_j, \mathbf{r}_\nu) \left( \frac{H_{xy}}{H_{y0}} \right)^{1/4} + iB_{zj,y0}(\mathbf{R}_j, \mathbf{r}_\nu) \left( \frac{H_{yo}}{H_{y0}} \right)^{1/4} \right]. \tag{27}
\]

The kinetics of the impurity population is defined by the following equation:

\[
dn_j/dt = -\Gamma_{\parallel,j} [n_j - n_T(\Omega_j)]. \tag{28}
\]

Here \( \Gamma_{\parallel,j} \) is the impurity relaxation rate and \( n_T(\Omega_j) = \exp(\hbar \Omega_j/k_B T) + 1 \) is the equilibrium population at frequency \( \Omega_j = \Omega_{0,j} + \delta \Omega_j(t) \). Taking into account

\[
n_j(t) = n_T(\Omega_{0,j}) + \delta n_j(t), \tag{29}
\]

\[
n_T(\Omega_j) = n_T(\Omega_{0,j}) + [\partial n_T/\partial \Omega_{0,j}] \delta \Omega_j(t),
\]

we can solve Eq.(28) and obtain

\[
\delta n_j(t) = \Gamma_{\parallel,j} \frac{\partial n_T}{\partial \Omega_{0,j}} \left[ \frac{\Phi_j c(t) + \Phi_j^* c^*(t)}{\Gamma_{\parallel,j} + \omega_0^2} \right]. \tag{30}
\]

The dynamic equation for the normal mode interacting with impurities is obtained by substituting [23] into [24]. This yields an additional term to the basic mode Hamiltonian [7]. Similar to [1], the dynamic equation is now:

\[
dc/dt = -i\omega_0 c - i\partial (H_{imp,j}/\hbar)/dc^* - i\sum_j \Phi_j^* \delta n_j(t).
\]

Substituting Eq.(30) into (28) and (31), we obtain the equation of damped harmonic oscillator [30]:

\[
dc/dt + \eta_{sr} c = -i(\omega_0 + \Delta \omega_{sr}) c + f(t). \tag{32}
\]

Here the frequency shift is equal to

\[
\Delta \omega_{sr} \simeq -\sum_j |\Phi_j|^2 \left( \frac{-\partial n_T(\Omega_{0,j})}{\partial \Omega_{0,j}} \right) \frac{\Gamma_{\parallel,j}^2}{\Gamma_{\parallel,j}^2 + \omega_0^2}. \tag{33}
\]
and the relaxation rate is:
\[ \eta_s \simeq \sum_j |\Phi_j|^2 \left( -\frac{\partial \Omega_{j,0}(\Omega_{0,j})}{\partial \Omega_{0,j}} \right) \frac{\omega_0 \Gamma_{||,j}}{\Gamma_{||,j}^2 + \omega_0^2}, \] (34)

where

\[ |\Phi_j|^2 = \frac{M_z V_0}{h^2} N \left[ \sum_\nu B_{z,j,x_0}(R_j, r_\nu) \right]^2 \left( \frac{H_{y_0}}{H_{x_0}} \right)^{1/2} + \left[ \sum_\nu B_{z,j,y_0}(R_j, r_\nu) \right]^2 \left( \frac{H_{x_0}}{H_{y_0}} \right)^{1/2}. \] (35)

To simplify (34), we assume that \( \Omega_{0,j} = \Omega_0 \) and \( \Gamma_{||,j} = \Gamma_{||} \) for all impurities. Summing over \( j \) gives an average anisotropic exchange \( B \):

\[ \sum_j \left[ \sum_\nu B_{z,j,x_0}(R_j, r_\nu) \right]^2 \overset{\text{imp}}{=} N \text{imp}(z_{\text{imp}} B)^2, \] (36)

where \( N \text{imp} \) is the total number of impurities in the sample and \( z_{\text{imp}} \) is the average number of magnetic neighbors for one impurity. Finally the relaxation rate (34) is:

\[ \eta_s \simeq c_{\text{imp}} \left( z_{\text{imp}} B \right)^2 \frac{\exp(h\Omega_0/k_B T)}{[\exp(h\Omega_0/k_B T) + 1]^2} \frac{\gamma(H_{x_0} + H_{y_0})\Gamma_{||}}{\Gamma_{||}^2 + \omega_0^2}, \] (37)

where \( c_{\text{imp}} = N \text{imp}/N \) is the impurity concentration and \( S = M_z V_0/h\gamma \) is the value of the host spin. This relaxation rate exhibits a definite temperature dependence. As discussed in the previous section for magnon-electron scattering processes, the impurity relaxation rate also is frequency dependent, via the term \( \gamma(H_{x_0} + H_{y_0}) \) (with identical dependence as in magnon-electron scattering).

V. MACROSCOPIC DYNAMIC EQUATIONS

A. Tensor form of magnetization damping

We have seen that both direct and indirect magnetization interactions with a thermal bath yield damped harmonic oscillator dynamics in the form:

\[ \frac{d}{dt} \left( \begin{array}{c} M_{x_0} \\ M_{y_0} \end{array} \right) = \left( \begin{array}{cc} -\eta & -\gamma H_{y_0} \\ \gamma H_{x_0} & -\eta \end{array} \right) \left( \begin{array}{c} M_{x_0} \\ M_{y_0} \end{array} \right) + \gamma M_s \left( \begin{array}{c} h_{y_0}(t) \\ -h_{x_0}(t) \end{array} \right). \] (39)

Here

\[ h_{y_0}(t) = -\left( \frac{h}{2\gamma M_z V} \right)^{1/2} \left( \frac{H_{y_0}}{H_{x_0}} \right)^{1/4} [f^*(t) + f(t)], \] (40)

\[ h_{x_0}(t) = -i \left( \frac{h}{2\gamma M_z V} \right)^{1/2} \left( \frac{H_{x_0}}{H_{y_0}} \right)^{1/4} [f^*(t) - f(t)] \]

are the independent random fields.

The nonlinear macroscopic dynamic equation for an anisotropic magnetic system which conserves the length of magnetization \( |M| = M_s \) has the form \[ 2, 8, 9, 10 \]:

\[ \frac{dM}{dt} = -\gamma M \times [H_{\text{eff}} + h(t)] - \frac{\gamma M}{M_s} \times \left[ \hat{\alpha} \cdot (M \times H_{\text{eff}}) \right]. \] (41)

Here \( \hat{\alpha} \) is a dimensionless damping tensor, which contains all necessary information about the system symmetry. In the vicinity of equilibrium one has

\[ \hat{\alpha} = \frac{\eta}{\gamma} \left( \begin{array}{ccc} 1/H_{y_0} & 0 & 0 \\ 0 & 1/H_{x_0} & 0 \\ 0 & 0 & 0 \end{array} \right), \] (42)

and Eq. (41) is equivalent to (39).

Note that the tensor form of damping in (41) is a natural description of an anisotropic system. It reflects an elliptical motion of magnetization and contains just one relaxation parameter \( \eta \). The \( \hat{\alpha} \) should not be confused with a damping tensor which contains two or more independent relaxation parameters.

B. Linearized Landau-Lifshitz equation

The Landau-Lifshitz equation with random fields \( h(t) \) has the form:

\[ \frac{dM}{dt} = -\gamma M \times [H_{\text{eff}} + h(t)] - \frac{\alpha M}{M_s} \times (M \times H_{\text{eff}}), \] (43)

where \( \alpha \) is a dimensionless damping parameter. Using (43), we can calculate the effective field \( H_{\text{eff}} = -\partial(E/V)/\partial M \) and write down the linearized equations for the transverse magnetization components \( (M_{y_0} \simeq M_z) \):

\[ \frac{d}{dt} \left( \begin{array}{c} M_{x_0} \\ M_{y_0} \end{array} \right) = \left( \begin{array}{cc} -\alpha H_{x_0} & -\gamma H_{y_0} \\ \gamma H_{x_0} & -\alpha H_{y_0} \end{array} \right) \left( \begin{array}{c} M_{x_0} \\ M_{y_0} \end{array} \right) + \gamma M_s \left( \begin{array}{c} h_{y_0}(t) \\ -h_{x_0}(t) \end{array} \right). \] (44)
We see that the non-diagonal terms in Eq. (44) and (39), as expected, coincide with each other, respectively. The diagonal terms, responsible for relaxation, in general, are different \((H_{x_0} \neq H_{y_0})\). These damping terms are equal only in the special case when \(H_{x_0} = H_{y_0}\) (see, Appendix A).

The Eq. (44) can be rewritten in terms of normal modes using (9) as

\[ dc/dt + \eta c + \tilde{\eta} c^* = -i\omega_0 e + f(t), \tag{45} \]

where

\[ \eta = \alpha \gamma (H_{x_0} + H_{y_0})/2, \quad \tilde{\eta} = \alpha \gamma (H_{x_0} - H_{y_0})/2. \tag{46} \]

We see that Eq. (45) contains one extra fast oscillating term \(\tilde{\eta} c^*\). As shown in Ref. [9], this term changes the power spectral shape. However, in the case when the thermal term is negligible, the fast oscillation term \(\tilde{\eta} c^*\) effectively vanishes by averaging. In those cases the LLG dynamics can be used as an approximation.

The most important influence of this parasitic fast oscillation appears in the case when we study the effect of a random force \(f(t)\) in Eq. (45); this term gives effectively higher magnetization noise in the system. Only, in the case when the thermal bath produces both magnetization relaxation and thermal fluctuations whose level is defined by fluctuation-dissipation theorem. Both damping and thermal noise appear as a result of microscopic spin fluctuations, therefore a connection of phenomenological dynamic magnetization equations with microscopic physics is necessary. We can quote van Kampen [31]: “The moral is that one cannot give a stochastic description of a physical system simply by taking one of the familiar equations for the sure case and declaring some of the coefficients to be random. One has to start from the fundamental equations...”.

VI. ANALYSIS OF FMR EXPERIMENTAL DATA

FMR linewidth measurements have been widely utilized to determine the relaxation rate for low-level linear excitations [32]. In this section we will use the relaxation rates for microscopic mechanisms analyzed in Secs. III and IV to analyze FMR data in soft thin films (NiFe, permalloy).

For small relaxation compared to the resonant frequency the linewidth is given by (e.g., [3]):

\[ \Delta \omega = 2\eta \tag{47} \]

for all microscopic processes. Usually in FMR experiments, the field swept linewidth \(\Delta H\) is measured. There is a simple relation between \(\Delta H\) and frequency linewidth \(\Delta \omega\):

\[ \Delta H = \frac{\Delta \omega}{\partial \omega_0/\partial H_0}, \tag{48} \]

where from (8)

\[ \partial \omega_0/\partial H_0 = \gamma^2 (H_{x_0} + H_{y_0})/2\omega_0, \tag{49} \]

which is valid for any direction of the applied magnetic field, including those parallel and perpendicular to the film plane.

Let us summarize the linewidths obtained in Sections III and IV. Using Eqs. (24), (37), (48) and (49), we can obtain the field swept linewidths:

\[ \Delta H_{m-c} \simeq \frac{4c_{def} \omega_0 m^3 V^2_0 f^2 \epsilon_F}{4\pi^3 \gamma \hbar^3}, \tag{50} \]

\[ \Delta H_{sr} \simeq \frac{4c_{imp} S (z_{imp} B)^2}{\hbar \gamma} \frac{\exp(h\Omega_0/k_B T)}{[\exp(h\Omega_0/k_B T) + 1]^2} \frac{\omega_0 \Gamma_{||}}{\omega_0}. \tag{51} \]

Note that the linear frequency dependence for \(\Delta H_{m-c}\) was predicted in Ref. [23].

The temperature dependence of \(\Delta H\) in permalloy thin films with the static magnetic field in the film plane has been measured by Patton and Wilts [11]. As seen in Fig. 2, \(\Delta H\) exhibits a strong frequency dependence with a maximum in the vicinity of \(T_{\text{max}} \approx 80\) K. The temperature maximum shifts to slightly higher temperatures with increasing FMR frequency. Such a non-monotonic temperature dependence of \(\Delta H\) is typical for slow relaxing impurities (see, [11], [29], [33]). However, the slow-relaxing impurity process alone cannot describe the experiment [11]. One can assume that the magnon-electron confluence process plays a role of a relaxation rate “baseline”. Thus for analysis we combine the slow-relaxing impurity and magnon-electron confluence processes together:

\[ \Delta H = \Delta H_{sr}(T, \omega_0, c_{imp}) + \Delta H_{m-c}(\omega_0, c_{def}). \tag{52} \]

According to Eq. (51), the temperature \(T_{\text{max}}\) of linewidth maximum is given by

\[ \Gamma_{||}(T_{\text{max}}) \simeq \omega_0. \tag{53} \]

The impurity damping \(\Gamma_{||}\) is expected to increase monotonically with temperature [29]. Thus \(T_{\text{max}}\) will increase with increasing \(\omega_0\) in agreement with experiment. The solid lines in Fig. 2 represent a theoretical fit with Eq. (52). We see that the temperature (and frequency) dependence [23] describe well the range from about the peak to about room temperature.

For the slow relaxation mechanism the fit gives
\[ \Delta H_{sr}[\text{Oe}] \approx \frac{1.9 \cdot 10^4}{T[\text{K}]} \cdot \frac{\exp \left( \frac{100}{T[\text{K}]} \right)}{[\exp \left( \frac{100}{T[\text{K}]} \right) + 1]^2 + \left( \frac{\omega}{\Gamma} \right)^2}, \]

where typical parameters for slow-relaxing impurity were used: \( \hbar \omega_0/k_B = 100 \) K, \( z_{imp}B/k_B = 25.3 \) K, \( S \approx 1 \) and \( c_{imp} = 10^{-3} \). The spin-lattice relaxation rate variation with temperature was assumed to be \( \Gamma_{\parallel}(T) = c_{\parallel} T^2 \) with a fit of \( c_{\parallel}/2\pi = 0.8\cdot10^{-3} \) GHz/K\(^2\). The impurity interactions with conduction electrons are likely to be responsible for such a temperature dependence. The spin-lattice relaxation time at room temperature \( 1/T_{\parallel}(300 \text{ K}) \approx 2.2 \text{ psec} \) agrees with typical experimental data [23].

For the magnon-electron confluence process the fit gives

\[ \Delta H_{m-e}[\text{Oe}] \approx 2.1 \left( \omega_0/2\pi[\text{GHz}] \right). \]  

Substituting \( m \approx 10^{-27} \) g, \( V_0 \approx 10^{-22} \) cm\(^3\) and \( \gamma = 1.76 \times 10^7 \) s\(^{-1}\) Oe\(^{-1}\) into Eq.(54) and comparing with Eq.(53), we obtain \( c_{def}/h \delta \omega \approx 7 \cdot 10^{-38} \) erg\(^3\). Taking defect concentration \( c_{def} = 10^{-3} \) and Fermi energy \( \epsilon_F = 3 \) eV, one gets an estimate for the interaction amplitude \( \hbar \delta \omega \approx 2.4 \) eV, which is typical for ferromagnetic metals [34,35].

Patton et al. [22] have studied the frequency dependence of \( \Delta H \) in thin permalloy films (17-48 nm) at room temperature (see, Fig.3). They have found that the field linewidth has a linear frequency dependence \( \Delta H \propto \omega_0 \) for the case when the external magnetic field is parallel to the film plane (open circles in Fig.2). For the perpendicular case (black circles) the linear dependence is valid just for high FMR frequencies \( \omega_0 > 8 \) GHz and it is saturated at smaller frequencies. Recent data [36] show similar results except that for the field perpendicular to the film plane the saturation region occurs only at extremely small frequencies (\( \omega_0/2\pi < 2 \) GHz). Essentially for a wide range of frequencies the field swept linewidths are identical for the two field orientations and strictly linear with resonant frequency. It is clear that this linearity can be explained by the magnon-electron confluence process (55). At room temperature the impurity relaxation rate is \( \Gamma_{\parallel}/2\pi \simeq 72 \) GHz so that this mechanism yields a linewidth (54) that is also linear with frequency at least to about 50 GHz. Taking \( \Gamma_{def} \), \( \Gamma_{rel} \) and \( \Gamma_{sat} \) at \( T = 300 \) K we obtain:

\[ \Delta H[\text{Oe}] \approx 2.3 \left( \omega_0/2\pi[\text{GHz}] \right). \]

This linear dependence is shown in Fig.3 by a solid line and demonstrates good agreement with experiment.

It is interesting that Eq.(52) can be also used for interpretation of experimental field linewidth in ultra-thin permalloy films [22]. The thickness dependence at room temperature for two different FMR frequencies is shown in Fig.4. Assuming that the surface magnetic atoms behaves as impurities, the concentration \( c_{imp} \) increases with decreasing film thickness as \( c_{imp} \propto d_0/d \), where \( d_0 \sim 3 \) Å is the linear size of the single-atomic magnetic layer. The theoretical thickness dependence for typical microscopic parameters (as above) can be written as

\[ \Delta H[\text{Oe}] = \left( 1.3 + 53/d[\text{Å}] \right) \left( \omega_0/2\pi[\text{GHz}] \right). \]  

The solid lines in Fig.4 by plot Eq.(57) and demonstrate good agreement with experimental data. This is an alternative interpretation of [36], where two-magnon scattering on surface defects (with thickness dependence \( 1/d^2 \)) plus a baseline thickness independent process were suggested. In order to evaluate these two interpretations, FMR linewidth should be measured at lower temperatures, where the role of slow-relaxing impurities increases.

A. Comment: data fitting by the LLG equation

The Landau-Lifshitz-Gilbert equations were originally introduced (a) for small magnetization motions (small deviations from the equilibrium) and (b) for the case of high magnetic symmetry (axial symmetry) with an isotropic damping fitting parameter \( \alpha \) (or “damping constant”) [19,21]. Nevertheless both “a” and “b” conditions are usually violated; these equations have been used for large magnetization motions such as magnetization reversal and for a description of magnetization dynamics in systems with lower magnetic symmetry (e.g., in thin films).

In the LLG phenomenology the linewidth is given by (e.g., [19]):

\[ \Delta \omega = \alpha \gamma (H_{ex} + H_{yy}). \]

From Eqs. (43) and (48) we find that the corresponding field swept linewidth is:

\[ \Delta H = \alpha \omega_0/\gamma. \]  

This formula has been used to obtain the phenomenological damping parameter \( \alpha \) from experimental data \( \Delta H \) and frequency \( \omega_0 \).

Assuming a constant \( \alpha \) leads, according to (59), to a linear frequency dependence for \( \Delta H \), in agreement with the room temperature data shown in Fig.3. This agreement has been used to claim the validity of the LLG equations even for an anisotropic system, e.g. a thin film [23]. This dependence is obviously coincidental. The LLG approach is purely phenomenological and can be related to physical processes only in the case of axial symmetry. As we have shown in this paper, detailed microscopic analysis yields dynamic equations which depend on the system symmetry. Microscopic analysis with appropriate relaxation mechanisms explain a wide variety of experimental data and, as well, can be used to predict dynamic magnetization phenomena.
VII. DISCUSSION

In this paper we have examined the effect of general forms of microscopic relaxation mechanisms on magnetization dynamics. Only the case of linear relaxation has been considered when the deviations of magnetization from equilibrium are relatively small. We have shown for two broad classes of relaxation mechanisms that dynamic relaxation may be expressed in terms of a damped harmonic oscillator equation driven by thermal fluctuations. The coordinates of the harmonic oscillator reflect a transformation of the transverse magnetization coordinates to circular motion. The transformation directly reflects the magnetic anisotropy. The relaxation time or damping parameter contains details of the microscopic relaxation mechanisms and also, in general, information, via the transformation, about the magnetic anisotropy. It is important to note that by anisotropy we do not mean the anisotropy that yields a tensor form is macroscopic and arises, for example, from the sample shape (e.g., a thin film) or the crystalline symmetry.

Specific mechanisms of slow relaxing impurities and magnon-electron coupling were analyzed. It was shown that a sum of these two effects explains very well FMR magnon-electron coupling were analyzed. It was shown that a sum of these two effects explains very well FMR frequency and film thickness. The fitting parameters that a sum of these two effects explains very well FMR thin film) or the crystalline symmetry.

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APPENDIX A: ELLIPTIC MAGNETIZATION MOTION

Consider the following energy density:

$$E_p/V = K_L \sin^2 \theta + K_\perp (M_x/M_s)^2 - M \cdot H_0.$$  \hfill (A1)

Here $K_L \geq 0$ and $K_\perp \geq 0$ are the “easy” ($z$) and “hard” ($x$) uniaxial anisotropy constants, $\theta$ is the angle between the magnetization $M=(M_x, M_y, M_z)$ and the easy axis of anisotropy. $H_0 = H_0(\theta, \cos \theta_H)$ is the external magnetic field. The equilibrium magnetization state $\theta = \theta_0$ is defined by the condition $\partial E_p/\partial \theta = 0$, or,

$$H^{(l)}_K \sin 2\theta_0 = 2H_0 \sin(\theta_H - \theta_0),$$  \hfill (A2)

where $H^{(l)}_K \equiv 2K_L/M_s$. In order to describe small magnetization oscillations it is necessary to transform the coordinate system and magnetization as follows

$$y = y_0 \cos \theta_0 + z_0 \sin \theta_0, \quad z = -y_0 \sin \theta_0 + z_0 \cos \theta_0, \quad x = x_0,$$

and

$$M_y = M_{y_0} \cos \theta_0 + M_{z_0} \sin \theta_0, \quad M_z = -M_{y_0} \sin \theta_0 + M_{z_0} \cos \theta_0, \quad M_x = M_{x_0}.$$  \hfill (A4)

The equilibrium state in these coordinates is $M = (0, 0, M_z)$. Taking into account transformation (A4) and relations $M^2 = M^2_x - M^2_y - M^2_z$ and $M_{z_0} \approx M_z - (M^2_x + M^2_y)/2M_x$, the energy (A1) can be written in the form \(8\), where

$$H_{s_0} = H^{(l)}_K \cos^2 \theta_0 + H^{(l)}_K + H_0 \cos(\theta_H - \theta_0),$$

$$H_{y_0} = H^{(l)}_K \cos 2\theta_0 + H_0 \cos(\theta_H - \theta_0),$$  \hfill (A5)

and $H^{(l)}_K \equiv 2K_L/M_s$. From (A4) and (A2) it follows that $H_{s_0} = H_{y_0}$ only in two special cases: 1) the case of spherical symmetry and 2) the case of uniaxial symmetry when the external magnetic field and equilibrium magnetization are oriented along the easy axis.
APPENDIX B: MAGNON-ELECTRON SCATTERING WITHOUT MOMENTUM CONSERVATION

The interaction of the uniform magnetic precession (a, a*) with electrons is assumed to be of the form:

\[ \mathcal{H}_I = \left(D^a + Da^*\right)/\sqrt{N}, \]  

(B1)

where

\[ D = \frac{1}{N} \sum_{k,k',j} f_{kk'}(r_j) d_k^d d_{k'}^c. \]  

(B2)

Here \( f_{kk'}(r_j) = |f_{kk'}| \exp[i\phi(r_j)] \) describes the scattering process in the vicinity of crystal defect at the point \( r_j \). \( \phi(r_j) \) is the phase, \( d_k^d \) and \( d_k^c \) are the fermion operators describing creation and annihilation of electron with wave number \( k \) and frequency \( \omega_{ek} \).

Using transformation (3) to a normal mode, we can rewrite \( \mathcal{H}_I \) as:

\[ \mathcal{H}_I/\hbar = \frac{1}{N^{3/2}} \sum_{k,k',j} \left[ \Psi_{kk'}(r_j) d_k^d d_{k'}^c + \Psi_{kk'}^*(r_j) d_k^c d_{k'}^* \right], \]  

(B3)

where

\[ \Psi_{kk'}(r_j) = uf_{kk'}(r_j) + vf_{kk'}^*(r_j) \]  

(B4)

is the amplitude of magnon-electron scattering.

The relaxation rate is defined by the golden Fermi rule as

\[ \eta_{m-e} = \frac{\pi}{N^3} \sum_{k,k',j} \left| \Psi_{kk'}^2 (\vec{m}_k - \vec{m}_{k'}) \delta(\omega_{ek'} - \omega_{ek} - \omega_0) \right| \]

\[ = \frac{\pi}{N^3} \sum_{k,k',j} (|u|^2|f_{kk'}|^2 + |v|^2|f_{kk'}^*|^2 + uv|f_{kk'}f_{kk'}^*| \cos[2\phi(r_j)]) \]

\[ \times (\vec{m}_k - \vec{m}_{k'}) \delta(\omega_{ek'} - \omega_{ek} - \omega_0). \]  

(B5)

Here \( \vec{m}_k \equiv \langle d_k^d d_k^c \rangle \) is the Fermi occupation number If we assume that the scattering phases \( \phi(r_j) \) are random and \( |f_{kk'}| = |f_{kk'}^*| \), then the \( uv \) term vanishes and Eq. (B3) becomes

\[ \eta_{m-e} = \frac{\pi c_{def} \sum_{k,k'} |f_{kk'}|^2}{N} \]

\[ \times (\vec{m}_k - \vec{m}_{k'}) \delta(\omega_{ek'} - \omega_{ek} - \omega_0). \]  

(B6)

Here \( c_{def} = N_{def}/N \), \( N_{def} \) is the total number of defects. After the following simplifications: \( \vec{m}_k - \vec{m}_{k'} = -\omega_0 \partial \vec{m}_k / \partial \omega_{ek} \) and \( \omega_0 \ll \omega_0 \), we obtain Eq. (B3).

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APPENDIX C: INTERACTIONS WITH IMPURITIES

Let us consider two-level impurities as effective spins \( s_j = 1/2 \). The anisotropic exchange Hamiltonian between the impurities and the neighboring host spins can be written in a general form:

\[ \mathcal{H}_{ex} = \sum_{j,\nu} \sum_{a,\alpha} B_{a,j,a}(R_j, r_\nu) s_{a,j}(R_j) S_a(R_j + r_\nu). \]  

(C1)

Here \( B_{a,j,a}(R_j, r_\nu) \) are the anisotropic exchange integrals, \( a_j = x_j, y_j, \tilde{z}_j \) are the local principal axes for the \( j \)-th impurity “spin” \( s_j(R_j) = \{ s_{x_j}(R_j), s_{y_j}(R_j), s_{z_j}(R_j) \} \) located at \( R_j \), and \( S(R_j + r_\nu) = \{ S_{x_j}(R_j + r_\nu), S_{y_j}(R_j + r_\nu), S_{z_j}(R_j + r_\nu) \} \) are the components of the host spin located in the vicinity of \( j \)-th impurity.

The Hamiltonian describing the impurity level modulation follows from Eq. (C1):

\[ \mathcal{H}_{ex,slow} = \frac{1}{\hbar} \sum_{j,\nu} \left[ B_{z_j,0}(R_j, r_\nu) S_{z_0}(R_j + r_\nu) \right. \]

\[ + B_{z_j,0}(R_j, r_\nu) S_{z_0}(R_j + r_\nu) \]

\[ \left. \right] \delta(\omega_{ek} - \omega_{ek'} - \omega_0). \]  

(C2)

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Figure captions

Fig.1 Relaxation processes: a) confluence of magnon and conduction electron into conduction electron in the presence of defect, b) ‘slow-relaxation’ mechanism via modulation of impurity levels by magnetization oscillation.

Fig.2 The linewidth temperature dependence for three FMR frequencies. Points are experimental data of Ref. [13]. Solid lines demonstrate theoretical fit with (54) and (55).

Fig.3 FMR frequency dependence at room temperature. Open circles correspond to parallel and black point to perpendicular field orientations. Solid line demonstrate theoretical fit (56).

Fig.4 The linewidth thickness dependence for two FMR frequencies at room temperature. Solid lines demonstrate theoretical fit (57).