Relaxation Mechanism for Ordered Magnetic Materials

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We have formulated a relaxation mechanism for ferrites and ferromagnetic metals whereby the coupling between the magnetic motion and lattice is based purely on continuum arguments concerning magnetostriiction. This theoretical approach contrasts with previous mechanisms based on microscopic formulations of spin-phonon interactions employing a discrete lattice. Our model explains for the first time the scaling of the intrinsic FMR linewidth with frequency, and \( T \) temperature dependence and the anisotropic nature of magnetic relaxation in ordered magnetic materials, where \( M \) is the magnetization. Without introducing adjustable parameters our model is in reasonable quantitative agreement with experimental measurements of the intrinsic magnetic resonance linewidths of important class of ordered magnetic materials, insulator or metals.

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INTRODUCTION

Since the discovery of magnetic resonance, the physics community has been fascinated with possible mechanisms to explain the absorption linewidth or the relaxation time in magnetic materials. It was and still is a very challenging problem. Magnetic relaxation is so important to understand because it affects a number of technologies, including computer, microwave, electronics, nanotechnology, medical, etc. Ultimately, the physical limitation of any technology which incorporates magnetic materials of any size, shape and combinations thereof comes down to precise knowledge of the relaxation time of the magnetic material being utilized. The background of various calculations or formulations of magnetic relaxation for the past sixty years or so can be summarized briefly as follows: (i) The relaxation times in paramagnetic materials [1] is characterized by two parameters, \( T_1 \) and \( T_2 \), wherein \( T_2^{-1} \) describes the magnetic resonance linewidth and \( T_1 \) describes the time taken for the external magnetic field Zeemann energy density \( -M \cdot H_{ext} \) to relax into thermal equilibrium. These times have been modeled in terms of various coupling schemes, i.e. spin-spin and/or spin-lattice interactions [2]. Since the coupling between spins is relatively weak, as it should be in a paramagnetic material, the coupling to the lattice involves discrete spin sites rather than a collective cluster of spins. As such, paramagnetic coupling is necessarily microscopic in nature. For example, a microscopic coupling scheme was formulated [3] whereby a spin Hamiltonian was modulated by the lattice motion. Variants to this approach have been very successful in explaining relaxation in paramagnetic materials. (ii) The magnetic relaxation of ferrimagnetic or ferromagnetic resonance (FMR) linewidth is characterized by the Gilbert parameter \( \alpha \) or equivalently by Landau-Lifshitz parameter \( \lambda_L \). A distinguishing feature of the collective coherent magnetic moments in FMR is that the magnitude of the magnetization, \( M = |M| \) remains fixed which requires a magnetic resonance equation of the simple form

\[
\frac{dM}{dt} = \gamma M \times H_{tot} = \gamma M \times (H + H'),
\]

wherein the gyromagnetic ratio \( \gamma = ge/2mc \). The total magnetic intensity \( H_{tot} \) has a thermodynamic part \( H \) determined by the energy per unit volume \( u \),

\[
du = Tds + H \cdot dM,
\]

and a dissipative part \( H' \) determined by the Gilbert linear operator \( \hat{\alpha} \),

\[
H' = \left[ \frac{1}{\gamma M} \right] \hat{\alpha} \cdot \frac{dM}{dt}.
\]

Eqs. [1] and [3] imply that all components of the magnetization must relax simultaneously in a way which conserves the magnitude of the magnetization. Much of the successful microscopic approaches or formulations utilized in paramagnetic materials were transferred over to models [4] which attempted to explain Eqs. [1] and [3]. In some sense this presented a contradiction or paradox which was conveniently ignored. As it is well known that collective excitations in a ferri or ferromagnetic state can be adequately described in classical continuum terminologies, although microscopic descriptions remain perhaps more accurate [7]. To our knowledge very few or any microscopic models have been successful in explaining the origin of Eq. [3]. For example, much attention was given in the seventies to explain the FMR linewidth in YIG (\( Y_3Fe_5O_{12} \)), since its linewidth was the narrowest ever measured in a ferrimagnetic material [8]. Clearly, there was less to explain, and perhaps spin-lattice interactions
could be treated at discrete spin sites as in paramagnetic materials. These calculations contained many approximations and predicted an FMR linewidth about 1/10 to 1/100 of the measured linewidth. We believe that this is the best agreement between theory and experiment on relaxation in an ordered magnetic material. The purpose of this work is to improve upon the predictability of a theoretical model not only on a given material but in general for any ordered magnetic materials without restoring to any approximations and assumptions.

We have adopted a conventional continuum magneto-mechanical description of the magnetic and elastic states of the ferri or ferromagnetic crystal. The advantage of this description is that the microscopic spin-lattice coupling need not be formulated, since it has already been included in the continuum model which has been proved to be experimentally correct. We introduce a thermodynamic argument stating that the heat exchange between the magnetic and elastic systems must be the same. As such, Eq. (3) may be directly related to the elastic sound wave relaxation time and the coupling strength between the magnetic and elastic systems. Specifically, we will show that \( \alpha \) is proportional to the square of the magnetostriction constant. i.e. \( \lambda^2 \) and inversely proportional to \( \gamma M \tau \) wherein \( \tau \) the elastic relaxation time. In addition, the model predicts that \( \tilde{\alpha} \) cannot be presumed to be a scalar as it has been done in the past; i.e. \( \tilde{\alpha} \) is predicted to be anisotropic a second rank tensor in a single crystal material.

It is clear that one needs an interaction between phonons and electron spins to account for Gilbert damping parameter \( \alpha \). Suhl and Moodera have considered such coupling schemes. The Gilbert damping parameter can be thought of as a transport coefficient in much the same way as conductivity and/or viscosity are transport coefficients. Such transport coefficients describe heating processes by which otherwise long lived modes are damped. One can in fact relate the Gilbert damping parameter to conductivity and/or viscosity. For metallic ferromagnetic materials, conductivity as well as electron viscosity produces considerable amount of magnetic damping via eddy current heating. For magnetic insulators it is the viscosity which determines the magnetic damping. As it is well known, conductivity and viscosity can be non zero even in zero frequency limit. Hence, the implied Gilbert damping parameter is also non zero at zero frequency. In Suhl and Hickey and Moodera’s papers they find, in the limit of zero frequency and zero wave number, that the real part of \( \alpha \) is zero. This limiting case suggests that they have not included the zero frequency transport coefficients consistently in their theory. In our derivation the expected result at zero frequency occur naturally in our formalism. In general, we believe the very nature of discreetness (as in paramagnetic materials) gives rise to relatively long magnetic relaxation times. However, the magnetic relaxation time of a coherent collection of spins (as in FMR) implies shorter relaxation times, since it involves collective acoustic waves in the interaction scheme. Our present theoretical treatment takes this into account via the continuum magneto-mechanics.

**THEORETICAL MODEL**

From Eq. (3), it is evident that the heating rate per unit volume due to the dissipative magnetic intensity \( \mathbf{H}' \) obeys

\[
\dot{Q} = \frac{dM}{dt} \cdot \mathbf{H}'
\]

\[
\dot{Q} = \frac{1}{\gamma M} \frac{dM}{dt} \cdot \tilde{\alpha} \cdot \frac{dM}{dt}
\]

\[
\dot{Q} = \frac{M}{\gamma} N_i \alpha_{ij} \dot{N}_j \] wherein \( N = \frac{M}{M} \). \( \text{(4)} \)

and \( \tilde{\alpha} \) is a second rank tensor

\[
\tilde{\alpha} = \begin{pmatrix}
\alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\
\alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\
\alpha_{zx} & \alpha_{zy} & \alpha_{zz}
\end{pmatrix} \) \( \text{(5)} \)

The crystal displacement \( \mathbf{u} \) yields in elasticity theory the strain tensor

\[
\mathbf{e}_{ij} = \frac{1}{2} (\partial_i u_j + \partial_j u_i) \] \( \text{(6)} \)

In virtue of the magneto-elastic effect, a changing magnetization \( dM/dt \) will produce a changing strain \( d\epsilon/dt \). In detail, in terms of third rank magneto-elastic tensor \( \Lambda_{ijkl} \) one finds

\[
\mathbf{e}_{ij} = \Lambda_{ijkl} \dot{N}_k \dot{N}_l, \quad \mathbf{e}_{ij} = 2\Lambda_{ijkl} \dot{N}_k \dot{N}_l. \] \( \text{(7)} \)

Finally, the fourth rank crystal viscosity tensor, \( \eta_{ijkl} \) determines the heating rate per unit volume due to the time dependent strain

\[
\dot{Q} = \mathbf{e}_{ij} \eta_{ijkl} \mathbf{e}_{kl}. \] \( \text{(8)} \)

Employing Eqs. (7) and (8) and comparing the result to Eq. (4) yields the central result of our model.

For any crystal symmetry the Gilbert damping tensor due to magnetostriction coupling is rigorously given by

\[
\alpha_{ij} = \frac{4\gamma}{\lambda^2} (\Lambda_{nmpi} N_p) \eta_{nmrl} (\Lambda_{rlqj} N_q). \] \( \text{(9)} \)

The following properties of the Gilbert damping tensor Eq. (9) are worthy of note: (i) The Gilbert damping tensor \( \tilde{\alpha} \) is inversely proportional to the magnetization magnitude \( M \). (ii) The Gilbert damping tensor \( \tilde{\alpha} \) is proportional to the squares of the magnetostriction tensor elements. (iii) The tensor nature of \( \tilde{\alpha} \) dictates that the
magnetic relaxation is anisotropic. To a sufficient degree of accuracy, one may employ an average of the form
\[
\alpha = \frac{1}{3} \text{tr} \{ \tilde{\alpha} \} = \frac{\alpha_{xx} + \alpha_{yy} + \alpha_{zz}}{3}
\]
(10)
defining a scalar function \( \alpha \). (iv) The crystal viscosity tensor \( \eta_{\text{crystal}} \) may be employed to describe the acoustic wave damping [15]. For a mode label \( a \), e.g., a longitudinal (\( a = L \)) or a transverse (\( a = T \)) mode, the acoustic absorption coefficient at frequency \( \omega \) is given by [13]
\[
\tau_a^{-1} = \frac{\omega^2 \eta_a}{2 \rho v_a^2},
\]
(11)
wherein \( v_a \) is the acoustic mode velocity and \( \rho \) is the mass density. Finally, for a cubic crystal, there are only two independent magneto-elastic coefficients which may be defined
\[
\lambda_{xx} = \frac{3}{2} \lambda_{100} \quad \text{and} \quad \lambda_{xy} = \frac{3}{2} \lambda_{111}
\]
(12)
wherein the Cauchy three index magneto-elastic coefficients are \( \lambda_{ijk} \).

**COMPARISON WITH EXPERIMENT**

The Gilbert damping factor \( \alpha \) may be deduced from the measurement of the intrinsic FMR linewidth. However, the measurement of the intrinsic linewidth is, indeed, very difficult. The reason for this conclusion is that there are too many extrinsic effects that influence the measurement. For example, in ferromagnetic metals like Ni, Co and Fe the intrinsic linewidth contribution to the total linewidth measurement [16, 17] may be between 10% and 30%. The rest of the linewidth [18] may be due to exchange-conductivity effects.

However, there may be other contributions, such as magnetostatic excitations, surface roughness, volume defects [19], crystal quality, interfaces [20], size, etc. Similar conclusions apply to ferrites except there are no exchange-conductivity effects [18]. Thus, the reader should be mindful that when we quote or cite an intrinsic value of the linewidth it represents a maximum value for there can be some hidden extrinsic contributions in an experiment. However, we have relied on data well established over the years. The criteria that we have adopted in choosing an ensemble of intrinsic linewidth measurements are the ones exhibiting the narrowest linewidth ever measured in single crystal materials. In addition, we required full knowledge of their elastic, magnetic and electrical properties [16, 17, 18, 21]. The objective is not to introduce any adjustable parameters.

The experimental value of Gilbert damping parameter \( \alpha_{\text{exp}} \) may be deduced from the FMR linewidth \( \Delta H \) at frequency \( f \) as
\[
\alpha_{\text{exp}} = \sqrt{\frac{3}{2}} \left( \frac{\gamma \Delta H}{2\pi f} \right)
\]
(13)
The factor \( \sqrt{3}/2 \) assumes Lorentzian line shape of the resonance absorption curve. The theoretical Gilbert damping parameter \( \alpha_{\text{th}} \) value is expressed in terms of known [17] parameters so that there are no adjustable parameters in our comparison to experiments, as shown in TABLE I. The theoretical prediction for the Gilbert damping parameter is that
\[
\alpha_{\text{th}} = \frac{36 \rho \gamma}{MT} \left[ \frac{\lambda_{100}^2}{q_T^2} + \frac{\lambda_{111}^2}{q_T^2} \right],
\]
(14)
wherein \( \rho \) is the mass density, \( q_T \approx v_T M/2\gamma A \) is the transverse acoustic propagation constant, \( v_T \) is the transverse sound velocity, \( A \) is the exchange stiffness constant, \( \lambda_{100} \) and \( \lambda_{111} \) are magnetostriction constants for a cubic crystal magnetic material. The transverse acoustic propagation constant, was approximated on the basis that the relaxation process conserved energy and wave vector. Since the acoustic frequency is fixed in the process the longitudinal propagation constant may be also calculated to be \( q_L = q_T (v_L/v_T) \) for magnetic materials, wherein \( v_L \) is the longitudinal sound wave velocity.

In FIG.1, we plot the experimental and theoretical values Gilbert damping constants as given by Eqs. (13) and (14). We note that the agreement between theory and experiment is remarkable in view of the fact that any of the cited parameters could differ from the ones listed in TABLE I by as much as 20-30%. For example, the linewidth reported in TABLE I may not be on the same sample where the elastic or magnetic parameters were cited. In a few cases we needed to extrapolate the value of \( A \), since there was no published value. In FIG.1 we did not present data on the ferromagnetic metals for lack of confidence on the linewidth data. For example, magnetostatic mode excitations have a deleterious effect on the dependence of the FMR linewidth on size. Most, if not all, previous FMR linewidth measurements have been performed on slabs, whiskers, etc., which can indeed support magnetoelastic mode excitations. Additional complications arise as a result of exchange-conductivity excitations in the linewidth data. Nevertheless, the agreement between theory and experiment is quite satisfactory.

**CONCLUSION**

Qualitative and quantitatively our model is in agreement with experimental observations of the intrinsic FMR linewidth reported over the years. Specifically, experimentally the most important characteristics of the intrinsic FMR linewidth, \( \Delta H \), measured on ordered magnetic
TABLE I: Calculated and measured Gilbert damping ($\alpha$) parameters

| Materials | $q_T$ ($10^{-6}$ cm$^{-1}$) | $\lambda_{100}$ ($10^{-6}$) | $\lambda_{111}$ ($10^{-6}$) | $M$ (G/4$\pi$) | $A$ ($10^{-6}$erg/cm) | $\Delta H$ (Oe) | $f$ (GHz) | $\tau$ ($10^{-13}$ sec) | $\alpha_{th}$ ($10^{-5}$) | $\alpha_{exp}$ ($10^{-5}$) |
|-----------|------------------|-----------------|-----------------|------------|----------------|----------------|-----|----------------|----------------|----------------|
| Y$_3$Fe$_5$O$_{12}$ | 3.8 | 1.25 | 2.8 | 139 | 0.40 | 0.33 | 9.53 | 4.4 | 5.56 | 9.0 |
| Y$_3$Fe$_4$GaO$_{12}$ | 1.46 | -1 | -1 | 36 | 0.28 | 3.0 | 9.53 | 4.4 | 51 | 76 |
| Li$_{10}$Fe$_{25}$O$_4$ | 8.6 | -8 | 0 | 310 | 0.40 | 2.0 | 9.50 | 1.5 | 26 | 50 |
| NiFe$_2$O$_4$ | 7.49 | -63 | -26 | 270 | 0.40 | 35 | 24.0 | 710 | 26 | 350 |
| MgFe$_2$O$_4$ | 9.30 | -10 | -1 | 90 | 0.1 | 2.3 | 4.9 | 1.5 | 120 | 120 |
| MnFe$_2$O$_4$ | 6.6 | -30 | -5 | 220 | 0.4 | 238 | 9.2 | 1.5 | 930 | 1040 |
| BaFe$_{12}$O$_{19}$ | 9.6 | 15 | 350 | 0.4 | 6 | 55 | 1.5 | 18 | 26 |
| Ni$^d$ | 6.3 | -46 | 25 | 484 | 0.75 | 102 | 9.53 | 1.8 | 770 | 2600 |
| Fe$^d$ | 8.75 | 20 | -20 | 1600 | 1.9 | 9 | 9.53 | 1.8 | 30 | 220 |
| Co$^d$ | 5.1 | 80 | 1400 | 2.78 | 15 | 9.53 | 1.8 | 530 | 370 |

$^a$Garnets $^b$Spinels $^c$Hexagonal Ferrite $^d$Ferromagnetic Materials

(Note: Longitudinal acoustic wave constant is $q_L = (v_T/v_L)q_T$)

FIG. 1: Shown are the experimental and theoretical values of the Gilbert damping constants as given by Eqs.(13) and (14).

materials (metal or insulator) for the past fifty years are that $\Delta H$ scales with frequency and $\frac{1}{\sqrt{f}}$. Indeed, these are the predictions of our theory. In addition, $\Delta H$ scales with the magnetostriction constant squared, see FIG.1. FIG.1 was plotted in a logarithmic scale only to be able to include all of the data in TABLE I. Another prediction of our theoretical work is that the Gilbert damping parameter $\tilde{\alpha}$ is not simply a scalar parameter but a tensor quantity. This implies that the FMR linewidth is intrinsically anisotropic in single crystals of ferri-ferrimagnetic materials. There was much controversy in the seventies about whether or not the intrinsic linewidth should be anisotropic or not. Poor quality of samples seemed to have incited the controversy. Improved or more accurate angular linewidth data [18,19] supports the notion of an anisotropic linewidth in ordered magnetic materials in agreement with our model. In summary, we believe that the comparison between theory and experiment is very encouraging in terms of continuing this continuum approach to explain intrinsic linewidths in ordered magnetic materials.

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