The damping of spin motions in ultrathin films: Is the Landau–Lifschitz–Gilbert phenomenology applicable? ☆

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Abstract

The Landau–Lifschitz–Gilbert (LLG) equation is used widely in device design to describe spin motions in magnetic nanoscale structures. The damping term in this equation plays an essential role in the description of the magnetization dynamics. The form of this term is simple and appealing, but it is derived through use of elementary phenomenological considerations. An important question is whether or not it provides a proper description of the damping of the magnetization in real materials. Recently, it was predicted that a mechanism called two magnon damping should contribute importantly to linewidths and consequently spin damping in ultrathin ferromagnetic films. This process yields ferromagnetic resonance (FMR) linewidths whose frequency dependence is incompatible with the linear variation expected from the Landau–Lifschitz equation. This prediction has now been confirmed experimentally. Furthermore, subsequent experimental and theoretical studies have demonstrated that the damping rate depends strongly on wave vector as well. It is thus clear that for many samples, the LLG equation fails to account for the systematics of the damping of the magnetization in ultrathin ferromagnets, at the linear response level. The paper will review the recent literature on this topic relevant to this issue. One must then inquire into the nature of a proper phenomenology to describe these materials. At the linear response level, the theory of the two magnon mechanism is sufficiently complete that one can describe the response of these systems without resort to LLG phenomenology. However, currently there is very great interest in the large amplitude response of the magnetization in magnetic nanostructures. In the view of the authors, it is difficult to envision a generally applicable extension of linear response theory into the large amplitude regime.

Keywords: Ferromagnetic; Films; Damping; Landau–Lifschitz–Gilbert; Two magnon

1. Introduction

Spin dynamics in magnetic nanostructures is a topic of great current interest. When the spin system is excited, of course, the spins precess in response to various torques exerted on them. These arise from externally applied magnetic fields, anisotropy fields of internal origin, dynamic dipole fields generated by the motions of the spins themselves, and finally torques generated by exchange interactions between the spins, if spatial gradients of the dynamics magnetization are present. Quite generally speaking, the origin and magnitude of such torques have been very well understood for decades, and our knowledge of these interactions in bulk magnetic materials provides us with knowledge sufficient to address their nature on the nanometer length scale.

It is critical to understand the nature of the damping of spin motions in such structures. There are practical reasons for this, in addition to interest from the perspective of fundamental physics. In the current era, major advances have resulted from devices which incorporate nanoscale magnetic components. The impact of GMR read heads on the technology of computer hard discs stands out as the most dramatic illustration of the usefulness of these new materials, and other applications are envisioned for the near future. All such devices depend for their operation on

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physical effects associated with the reversal of the magnetization, or on the response of the device to changes in orientation of the magnetization. The speed by which information may be read, written or extracted is controlled by the damping of spin motions within the device. The interest centers on nanoscale magnetic materials fabricated from the 3d transition metal ferromagnets or their alloys, since their Curie temperatures are well above room temperature, thus insuring that cooling is not required to exploit the magnetic degrees of freedom. More recently, ultrathin ferromagnets are employed in very high-frequency microwave devices whose performance characteristics are influenced importantly by spin damping [1].

Thus, a central question is the nature of spin damping in nanoscale samples of the metallic 3d ferromagnets. Can we simply extrapolate down to the nanoscale what we know about this from bulk materials, or do we encounter new mechanisms operative on small length scales not present in bulk matter? It is now clear that there are indeed new and fascinating mechanisms which operate at small length scales, and which are not evident in studies of bulk materials. These fall into two classes: (i) extrinsic mechanisms and (ii) intrinsic mechanisms which are present by virtue of the fact that a large fraction of the magnetic moments are in or near surfaces and interfaces. In this paper, we discuss the implications of our past work [2] on an extrinsic mechanism referred to as two magnon damping. At this time, we have an impressive body of data in hand which show that two magnon damping enters importantly in diverse samples fabricated from ultrathin ferromagnets. As we shall point out here, one consequence of the presence of this mechanism is that a well known and often used phenomenology breaks down qualitatively in samples where this mechanism is operative. In the view of the present authors, this has serious consequences, since the phenomenology to be described next is widely used in the magnetics community as the basis for device design.

2. Some background

Long wavelength spin motions in diverse ferromagnetic structures are described by a commonly used phenomenological equation referred to as the Landau–Lifschitz–Gilbert (LLG) equation. It describes the system in macroscopic language, in terms of the magnetization per unit volume $\vec{M}(\vec{r},t)$. The LLG equation takes the form

$$\frac{\mathrm{d}\vec{M}(\vec{r},t)}{\mathrm{d}t} = \gamma\left[\vec{H}_{\mathrm{eff}} \times \vec{M}(\vec{r},t)\right]\partial + \frac{G}{\gamma M_{\mathrm{S}}^2}\left[\vec{M}(\vec{r},t) \times \frac{\partial \vec{M}(\vec{r},t)}{\partial t}\right]. \quad (1)$$

The first term on the right-hand side of Eq. (1) is the torque term, and the various effective fields mentioned in Section 1 are incorporated into the operator $\vec{H}_{\mathrm{eff}}$ and, as mentioned earlier, these are well understood. The second term is a damping term introduced in a phenomenological manner many decades ago. Clearly, the magnetization must be time dependent for damping to occur. Hence the proportionality to $\partial\vec{M}/\partial t$. In ferromagnets, it is assumed that the length of the magnetization is conserved locally as it relaxes to its equilibrium position. This is insured by the structure of the damping term. We note that even within the framework of the original phenomenology, it has been pointed out recently [3] that the form of the second term in Eq. (1) is overly restrictive. Its form assumes the equation of motion is form invariant under arbitrary rotations of the coordinate system, whereas in any ferromagnet the equation only need be form invariant under rotations about the z-axis, along which the magnetization points (in the presence of anisotropy, the symmetry is lower yet). Thus, symmetry allows a generalized form of the damping term on the right-hand side of Eq. (1). While one must keep this matter in mind, this is not the issue of primary concern in this paper.

If we accept the commonly used LLG equation, then one strong prediction follows, when it is applied to a ferromagnetic film magnetized in plane, or perpendicular to the plane. This is that the ferromagnetic resonance (FMR) linewidth must scale linearly with the FMR frequency. We refer the reader to Ref. [3] for a review of applications of the LLG equation and an expanded discussion of this point, along with related matters.

Early FMR data on ultrathin films taken for three frequencies in the 10–36 GHz range were indeed interpreted [4] in terms of a picture where a linear variation with frequency was imposed on a “zero field linewidth”, a constant independent of frequency assumed to have its origin in sample inhomogeneities. It is the case that the value of G extracted from such fits to FMR data on ultrathin films is consistently larger than appropriate to macroscopic crystals of the same material, and this quantity was also found to depend on growth conditions.

In the early days of FMR studies of ferrites, it was also found that linewidths were systematically larger than expected from the theory of intrinsic processes. The linewidth was also sensitive as well to aspects of sample preparation [5]. In a classic paper [6], a mechanism called two-magnon damping was introduced, and shown to account nicely for the data. It can be said that the understanding of the origin of this extrinsic contribution to the linewidth allowed it to be eliminated through appropriate sample preparation, and this made the first ferrite devices possible. The physical picture is that in the spherical samples used in the experiments, the FMR mode (a spin wave or magnon of infinite wavelength) was degenerate in frequency with a band of short wavelength spin waves (wavelengths short compared to the sphere radius). Hence surface defects could scatter energy from the uniform mode to the short wavelength degenerate manifold of magnons, thus providing an extrinsic mechanism for damping the FMR mode.

A few years ago, we argued that that the two-magnon mechanism can be operative in the ultrathin film environment

[2], since by virtue of the contribution of dipolar interactions to the spin wave dispersion relation at long wavelengths. In quasi-two-dimensional ferromagnets one realizes short wavelength spin wave modes degenerate with the main FMR mode. We developed the theory of this source of damping, which shortly thereafter was extended by Rezende and his collaborators [7]. We now have in hand several experiments which nicely confirm the predictions of theory for such systems. We direct the reader to Ref. [3] for a discussion.

A consequence is that in small magnetic structures where the two-magnon mechanism is operative, the LLG equation provides a qualitatively incorrect account of the damping of the spin motions. We discuss why this is so and its consequences in the next section.

We note that a second damping mechanism not present in bulk materials also is operative in ultrathin ferromagnets. This is referred to as the spin-pumping mechanism [8]. When the spins precess coherently in an ultrathin metallic ferromagnetic adsorbed on a metallic substrate, spin current is transmitted across the interface into the substrate, where the angular momentum it carries off from the film is dissipated. This is an intrinsic damping mechanism, it should be noted, operative in the ultrathin film environment and absent from bulk materials. The spin pumping mechanism leads to a contribution in the FMR linewidth which scales linearly with frequency, and thus within macroscopic phenomenology is compatible with the LLG equation, though it raises the value of the effective damping constant G above that appropriate to bulk matter. For the first experimental study of this phenomena see Ref. [8]. This paper contains references to earlier theoretical work which predicted the phenomenon. For more recent theoretical work see Refs. [9–11]. A different but equivalent viewpoint is found in Refs. [12–14]. A new approach, and quantitative account of data is given in Ref. [15].

In Section 3, we focus on those aspects of two-magnon damping incompatible with the LLG equation, and discuss the consequences.

3. Two-magnon damping and consequences for the LLG equation

As remarked above, a key prediction of the LLG equation is that, for the commonly employed FMR geometry (magnetization in plane, for example), the FMR linewidth should scale linearly with the FMR frequency ω_{FMR}. For in-plane magnetized films, the two-magnon mechanism provides a very different behavior. Under the conditions outlined in Ref. [2], commonly encountered in experiments, this theory provides an expression which may be written as follows:

$$\Delta H^{(2)} = \Gamma \sin^{-1}\left[\frac{\left\{\left(\frac{\omega_0}{2}\right)^2 + \omega_{\mathrm{FMR}}^2\right\}^{1/2} - \left(\frac{\omega_0}{2}\right)}{\left\{\left(\frac{\omega_0}{2}\right)^2 + \omega_{\mathrm{FMR}}^2\right\}^{1/2} + \left(\frac{\omega_0}{2}\right)}\right]^{1/2}. \qquad (2)$$

Here, Γ is the square of a matrix element whose detailed form depends on the nature of the defects responsible for activating the two-magnon mechanism. Then $\omega_0 = \gamma(4\pi M_S + H_S)$, with H_S the strength of the surface anisotropy field, taken positive when the normal to the surface is a hard axis.

If $\omega_{\mathrm{FMR}} \ll \omega_0$, then Eq. (2) predicts that the linewidth should vary linearly with frequency, very much as the prediction of the LLG equation. However, under typical conditions, $\omega_{\mathrm{FMR}} \sim \omega_0$, and in this regime Eq. (2) yields strong deviations from linear behavior. A plot of Eq. (2) shows that the linewidth should increase much more slowly with frequency than expected from the linear law, for experimentally accessible FMR frequencies.

How, then, does one reconcile the comments just made with the analyses such as those described in Ref. [4]? It was argued in Ref. [2] that data taken only for the three frequencies commonly used in FMR studies, 10, 24 and 36 GHz are in fact compatible with Eq. (2), which when plotted over a narrow frequency range can be fitted approximately by a straight line with finite intercept at zero frequency. The frequency range covered is not sufficiently large so that one can detect the negative curvature expected from Eq. (2). In a remarkable series of experiments, Baberschke and his colleagues measured linewidths of Fe/V superlattices from 1 to 80 GHz. Their data are fitted beautifully by the form in Eq. (2), supplemented by a linear term associated with classical Gilbert damping. Their data provides no evidence for the "zero-field linewidth" discussed in Ref. [4]. It is intriguing that these data also shows a dramatic fourfold in plane anisotropy associated with the two-magnon contribution to the linewidth: when the magnetization is aligned along a [11] direction, the two-magnon contribution is very small. In Ref. [2], it was argued that (i) the dominant contribution to the prefactor Γ in Eq. (2) has its origin in the perturbation of surface anisotropy by defects, and (ii) it was argued that the defects consist of rectangular structures of random aspect ratio with sides parallel to the [10] directions. Remarkably, the coefficient Γ then vanishes identically when the magnetization is aligned along a [11] direction [16]. In subsequent experiments on films in which the two-magnon mechanism is active, the Heinrich group has verified that the defects in the film have symmetry compatible with this picure through their STM studies of the samples [17].

It is established that the two-magnon mechanism is active in diverse ultrathin films, very much as proposed in Ref. [2]. A consequence is that the linear frequency dependence predicted from the LLG phenomenology is qualitatively incorrect for real materials. This has important consequences for device design: extrapolation of linewidths inferred from FMR data to higher frequencies will provide a misleading estimate of damping rates at higher frequencies. This can have serious consequences for the design of devices which will operate at high frequencies or short time scales.

In very lovely experimental studies accompanied by new theory, the Rezende group has set forth a second critical observation. These authors compared FMR linewidths with spin wave linewidths measured on precisely the same sample by means of Brillouin light scattering (BLS). Remarkably, the BLS linewidths are larger than those measured by roughly a factor of five. These authors argue that the two-magnon mechanism is dominant in their samples, and they set forth calculations which demonstrate that the damping increases strongly with wave vector. In FMR, one excites a mode whose wave vector is very close to zero, whereas in BLS one excites modes with wave vectors in the range of those of the visible photons used in the experiment, $\sim 10^5\,\mathrm{cm}^{-1}$. These authors obtain a most impressive quantitative account of their data as follows. From the FMR linewidth, and an expression such as that in Eq. (2), they obtain a value for the prefactor Γ. Then through use of their theory, given this value of Γ they are able to obtain an excellent and fully quantitative account of the large linewidths seen in BLS, with no further adjustable parameters. We remark that examination of the expressions found in Ref. [2] shows that the wave vector scale on which substantial variations can be expected is $k_C \approx 4\pi M_S d/D$, with d the film thichness, and D the exchange stiffness, expressed in $\mathrm{G/cm^2}$. For the 3d transition metal ferromagnets, one estimates that $k_C \sim 2 \times 10^5\,\mathrm{cm}^{-1}$, which indeed lies in the range probed by BLS.

Thus, two matters are now very clear from the experimental data on ultrathin film structures, when the two-magnon mechanism is operative: (i) there are very large deviations from the linear frequency dependence of the FMR linewidth predicted by the LLG equations, and (ii) the linewidth exhibits a strong dependence of the wave vector of the spin wave, even for wavelengths long compared to the underlying lattice constant (the scale of the wave vector dependence r is set by k_C), whereas the LLG equation predicts the linewidth to be independent of wave vector.

It is thus the case that the LLG phenomenology fails badly to account for the spin damping observed in ultrathin ferromagnets. If it fails in these systems, we can expect failures for diverse forms of nanoscale magnetic matter.

Can we then write down a phenomenological equation of motion to replace the LLG equation of motion in such structures? This appears to be a major challenge, in the view of these writers. Ultrathin films are the most straightforward example of a magnetic nanostructure, so in what follows we consider only films.

First suppose we consider the small amplitude, linear response of the spin system, and we wish to write an equation of motion for its magnetization in real space. Then even for this simple case, the fact that the linewidth is not simply linear in frequency means the damping term would have the form of an integral over the past history of the magnetization motions in the system, i.e. the damping term will be nonlocal in the time domain rather than simply proportional to $\partial \vec{M}/\partial t$. Similarly, the strong wave vector dependence discussed in Ref. [7] would require a damping term nonlocal in space as well. Thus, the very simple LLG equation would have to be replaced by a rather sophisticated integrodifferential equation. However, in the linear response regime, Ref. [2] introduces and develops a formalism in frequency/wave vector space which allows one to obtain a complete description of the film response. One requires a microscopic picture of the specific mechanism which activates the two-magnon scattering, and from there one can carry through a complete description of the response of the system in terms of frequency and wave vector-dependent susceptibilities supplemented by, to use the language of many-body theory, a spin wave self-energy matrix. Such an analysis may not be simple in all situations. However, we may say that the problem of describing the linear response of an ultrathin film is solved in principle, through the formalism erected in Ref. [2].

In the current era, there is very great interest indeed in obtaining a complete description of the magnetization dynamics in magnetic nanostructures, under conditions where the deviations from equilibrium are very large in amplitude. The rapidly growing literature on torques induced by spin polarized transport currents injected into films and other structures are an illustration of a circumstance where such a phenomenology is needed. In virtually all studies, the LLG equation forms the basis of the analysis of the spin motions. We have seen that in real materials, it proves quite inadequate even for a discussion of the linear response, to the extent that damping in the spin system is of concern. The question of how to extend the LLG phenomenology into regime of large amplitude spin motions under conditions where two-magnon scattering is operative is, at the time of this writing, a most difficult question to address meaningfully. One might suppose the Blombergen equation of magnetic resonance, which distinguishes between longitudinal and transverse relaxation processes might serve as a starting point. However, the strong wave vector dependence of the damping rate observed in Ref. [7] is omitted from such an approach, as is the frequency dependence associated with the two-magnon mechanism.

Thus, in view of the remarks of the previous paragraph, we presently lack a phenomenology which provides us with a realistic description of spin motions in ultrathin film ferromagnets and by extension diverse magnetic nanostructures, most particularly when we enter the regime of large amplitude motions of the magnetization. We perceive this as a serious difficulty which will limit our ability to correctly predict device performance, most particularly when we are concerned with high frequencies and short length scales.

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