Longitudinal resonance for thin film ferromagnets with random anisotropy

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At the microscopic level, individual spins in ferromagnets with random anisotropy tip transversely with distinct local angles relative to the magnetization $\vec{M}$. When driven by an rf field along the equilibrium $\vec{M}_0$, which changes $dM = \hat{M}_0 \cdot d\vec{M}$, the transverse tippings rotate about $\vec{M}_0$, corresponding to a new macroscopic collective angle ϕ about $\vec{M}_0$. The coupling of dM and ϕ leads to a new longitudinal mode that in bulk has a frequency that is largely independent of field H. A longitudinal mode has been observed in thin films of ferromagnets with random anisotropy, but its frequency is H dependent; for H at angle θ to the film normal and fixed resonator frequency f, $H \cos\theta$ was constant to angles of 80°, with H saturating for larger angles. When the demagnetization field is included the theory yields such an H vs θ for $\theta < 80°$, thus providing evidence for the predicted angle ϕ. However, lower frequency resonators are needed to manifest the predicted macroscopic anisotropy energy.

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I. INTRODUCTION

A ferromagnet with random anisotropy (FRA) should possess two macroscopic dynamical variables—both the longitudinal magnetization $\vec{M}_0$, and rotations by a macroscopic collective angle ϕ about $\vec{M}_0$, which decouple from the transverse components of the magnetization [1]. This leads to a different anisotropy-driven restoring torque and a different resonance, driven by an rf field $\vec{h}_{rf}$ along $\vec{M}_0$ [1]. Evidence has been found for such a longitudinal resonance (LR) [2,3], for $Co_{93-x}Zr_7(RE)_x$ (RE = Pr, Nd, Tb, and Dy) amorphous thin films, but with very different properties than predicted by the original theory, developed for bulk. The present work shows that by including the demagnetization field the data can be understood, and thus validates the predicted macroscopic collective angle. Note that a longitudinal resonance has been observed in spin glasses, where the origin of the macroscopic angles lies in random exchange [4,5].

We also observe that, by employing a lower resonance frequency [6], the effect of the random anisotropy-driven restoring torque, and its corresponding macroscopic energy, should be observable. Further, for large fields H at large angles θ to the film normal, all four magnetic variables—the three components of the magnetization $\vec{M}$ and the angle ϕ—couple. This will require further studies, both experimental and theoretical.

Section II reviews FRAs, both theory and experiment. Section III reviews the experiments on the longitudinal resonance (LR). Section IV presents the relevant dynamical variables, their macroscopic energies and their dynamics. Section V works out the consequences of the macroscopic energy on the equilibrium state. Section VI develops the detailed theory of longitudinal resonance, including the crucial effect of the demagnetization energy. Section VII presents a summary and conclusions.

II. FERROMAGNETS WITH RANDOM ANISOTROPY: EXPERIMENT AND THEORY

We begin with an introduction to FRAs, followed by an experimental review, of the LR mode, and then the theory.

A. Ripple and skew in ferromagnetic films

Early work on magnetic films, largely driven by magnetic recording issues, found two types of magnetic nonuniformity. At small scales the spins in different crystallites (dimension ⩽200 nm) pointed in slightly different directions: *small scale ripple*. At large scales the magnetization could vary, presumably due to nonuniform deposition: *large scale skew*. Citing experiments [7] establishing ripple, and work suggesting that ripple is due to local variations of the easy axis [8,9], Hoffman and others developed a theory for ripple in permalloy films [10–13].

This early theoretical work assumes that magnets are only weakly disordered by random axis anisotropy [10,12–14]. It finds that in a field $\vec{H}$ there is a transverse component of the magnetization, due to the randomly oriented anisotropy axes in randomly oriented crystallites [15]. Let the film normal be along $\hat{z}$. With saturation magnetization M_s (in A/m), macroscopic uniaxial anisotropy constant K_u (an energy density), and in-plane preferred axis $\hat{y}$ (determined by the cooling history), it is described as having a uniaxial energy density

$$\varepsilon_u = -\frac{K_u}{M_s^2}(\vec{M} \cdot \hat{y})^2. \quad (1)$$

Experiments indicate that, even when pulled out of plane, the magnetization magnitude M is always close to M_s.

Later, motivated by developments in critical phenomena, Imry and Ma found that in dimension $d = 3$ ordinary magnets

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subject only to short-range exchange and random *fields* do not have long-range order, the random fields causing decoherence over a large distance [16].

For the related and more practical problem of random axis anisotropy and exchange, Chudnovsky and Serota showed that the system also becomes disordered [17,18]. However, in a field or with some form of macroscopic anisotropy (as for permalloy), the system retains long-range order.

B. Longitudinal resonance (LR)

At the microscopic level, the local spins in ferromagnets with locally random anisotropy (but with an applied field or crystalline anisotropy to maintain long-range order) tip transversely to the net magnetization $\vec{M}$. As a whole they are characterized by a macroscopic angle variable ϕ about $\vec{M}$ that may be set to zero in equilibrium. This angle ϕ is a new dynamical degree of freedom that couples to the longitudinal magnetization, leading to a mode that in theory can be excited by an rf magnetic field $\vec{h}_{\rm rf}$ along the longitudinal direction [1].

The present work shows that, just as the demagnetization field in thin films changes the resonant frequency of the usual, transverse, ferromagnetic resonance mode (TR) [19], so the demagnetization field changes the resonant frequency of the longitudinal resonance mode (LR). Applied to resonance data on thin films the theory yields a LR that agrees well with experiment. More extensive experimental studies are desirable because, although the experiments provide evidence for the presence of the macroscopic angle ϕ, they are at too low a frequency to observe the effect of the macroscopic twisting energy associated with ϕ.

Specifically, theory can explain the following:

(1) the dependence on the direction α of the rf field relative to the anisotropy axis $\hat{y}$, as expected for an LR;

(2) the relatively low intensity of the LR, from the small dynamical susceptibility expected for the LR relative to the usual ferromagnetic resonance (TR);

(3) the unusual result that, for fields H at an angle θ relative to the normal, the LR satisfies $H \sim (\cos\theta)^{-1}$ for angles up to about 80°;

(4) the nearly linear relation between H and driving frequency f for the range $6 < f < 12$ (GHz) studied;

(5) the complex line shape at larger θ, due to the demagnetization field coupling the LR and the TR (meaning four coupled variables), which we do not further investigate in this work.

III. LONGITUDINAL RESONANCE EXPERIMENTS

In the 1980s Suran and coworkers began studying the static properties thin magnetic films doped with rare earths (REs) [20,21]. The spatial scale of the disorder is approximately the separation between the dopant atoms; perhaps 1–2 nm. As with permalloy studies of the 1950s, they found an in-plane uniaxial (not unidirectional) anisotropy along the cooling field direction. As with the early work largely on permalloy, basic mechanisms have been proposed but the phenomenon of uniaxial anisotropy is not definitively understood [15].

Following their static studies, they also studied the expected transverse ferromagnetic resonance (TR), more commonly called ferromagnetic resonance (FMR). Then, in a set of six papers, Suran and coworkers established that for certain magnetic thin film systems, typically amorphous $Co_{93-x}Zr_7RE_x$ thin films with RE = Pr, Nd, Tb, and Dy up to 4%, all support a previously unobserved longitudinal resonance [2,3,22–25]. (Only samples without long-range skew showed this resonance; high deposition temperatures gave skew.) An earlier theory for LR in the bulk [1] cannot explain these thin film results.

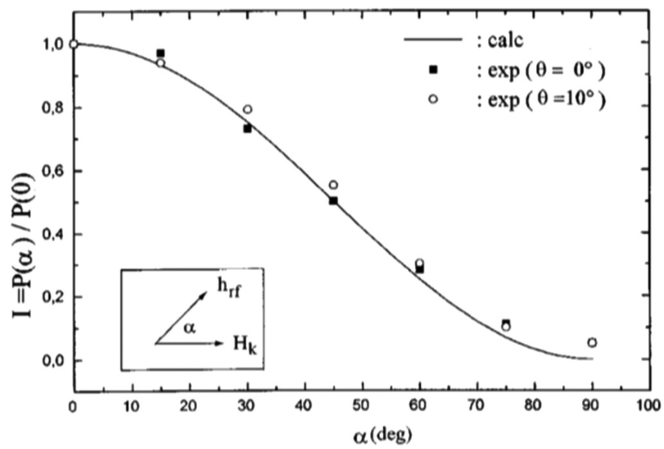

FIG. 1. Relative intensity I data from $I(\alpha)$ from Ref. [2], establishing longitudinal nature of new mode. Theoretical curve is $\cos^2\alpha$. Reproduced from G. Suran, E. Boumaiz, and J. Ben Youssef, J. Appl. Phys. **79**, 5381 (1996), with the permission of AIP Publishing.

We now highlight certain aspects of the experiments. Since demagnetization effects are significant the magnetization $\vec{M}$ largely lies in the plane.

(i) By definition, when $\vec{h}_{\rm rf}$ is in the film plane, it is at an angle α relative to the uniaxial anisotropy field $\vec{H}_u$, as in the inset to Fig. 1, taken from Ref. [2]. (We show below that $\vec{H}_u$ has magnitude proportional to $\hat{M}\cdot\hat{y}$, and thus is not a constant.) This figure shows that the intensity of the new line varies as $\cos^2\alpha$. Since for a longitudinal mode only the component of $\vec{h}_{\rm rf}$ along $\vec{H}_u$, or $\cos\alpha$, should be effective in driving the resonance, this result is consistent with a longitudinal mode. The measured $\vec{M}$ is nearly in-plane, so its out-of-plane component is small. This will be useful in later approximations.

Reference [2] also studied the resonant field H normal to the plane, or $H_\perp$, at a number of resonator frequencies from 6 to 11 GHz (see Fig. 2). With θ the angle between H and the normal, this corresponds to $\theta = 90°$. The data points are taken from Ref. [2]. Reference [3] reported similar results. Below we show that this nearly linear H vs f behavior can be understood if the zero-field longitudinal resonance $f_0 = \omega_0/2\pi$ is at perhaps 2–4 GHz. Figure 2 is drawn for $f_0 = 2$ GHz.

(ii) Reference [22] studied the resonance field H for $\alpha = 0$ and various angles θ to the normal to the plane. Frequencies of 9.8, 17.9, and 35.7 GHz were studied. At 35.7 GHz their Fig. 2 shows that, for θ up to 80°, $H_\perp \equiv H\cos\theta$ is independent of θ, so $H \sim (\cos\theta)^{-1}$. (The vertical axis reads H, but the text discusses $H_\perp$, which is consistent with the later works.) Ref. [23] reported similar results.

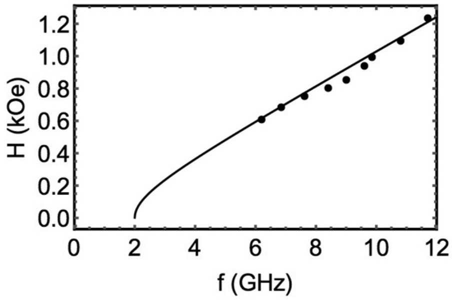

FIG. 2. Data points for ω vs H measurements from Ref. [2] for $\alpha = 0$ and $\theta = 90^\circ$. Theoretical curve from Eq. (26) for $f_0 = 2$ GHz, $M = 1.0 \times 10^6$ A/m, and $\chi_L = 3.6 \times 10^{-3}$.

(iii) Reference [24] gave detailed line shapes for a number of angles at $f = 9.8$ GHz, and plotted field H vs θ. For θ up to 80°, $H \sim (\cos\theta)^{-1}$, but $H(90^\circ)/H(0)$ saturated near 15 (see Fig. 3). The authors remark without specifics that for large θ the LR develops a transverse component.

(iv) Reference [25] studied different films (a-$Fe_{100-x-y}Co_xZr_y$). For a-$Fe_{72}Co_{15}Zr_{13}$, detailed line shapes are given for a number of angles at $f = 9.8$ GHz. For θ up to 80°, $H \sim (\cos\theta)^{-1}$, but $H(90^\circ)/H(0)$ saturated near 6. From their information (Gaussian units) we estimate that $4\pi M_s \approx 12.5$ kOe, or (SI units) $M_s \approx 10^6$ A/m. See Fig. 4, where the solid line is theory, given below. The present work employs their coordinate system.

The first four of the above works refer to the new longitudinal resonance as the LR, but the last two refer to it as the RAR (random anisotropy resonance).

Below we show that, for θ not too large, this $H \sim (\cos\theta)^{-1}$ dependence at fixed f is expected theoretically. Further, if f is constant the tipping angle β of $\vec{M}$ is the same for all resonances. However, changing f also changes β, making comparison to experiment more complex.

IV. THEORY: ENERGY AND DYNAMICS

Consider a thin film ferromagnet with random anisotropy and normal $\hat{z}$. Take it to have uniaxial anisotropy given by Eq. (1), and temporarily restrict the external field $\vec{H}$ to lie in the y-z plane, with

$$\vec{H} = H(\hat{z}\cos\theta + \hat{y}\sin\theta). \tag{2}$$

Then in equilibrium $\vec{M}$ will lie in the y-z plane and, moreover, for the longitudinal mode, $d\vec{M}$ will also lie in the y-z plane. Following Suran, let $\vec{M}$ lie in this plane at an angle β to $\hat{y}$ (not to $\hat{z}$), so

$$\vec{M} = M(\hat{z}\sin\beta + \hat{y}\cos\beta). \tag{3}$$

We employ SI units but, as noted above, the experiments employed Gaussian units. A clear tutorial discussion of the units conversion is given by Ref. [26].

We now discuss the various energies:

(i) The Zeeman energy is $-\mu_0\vec{H}\cdot\vec{M}$.

(ii) The thin film demagnetization energy is $(\mu_0/2)(\vec{M}\cdot\hat{z})^2$.

(iii) The energy that, for zero field, ensures both that $M \approx M_s$ in equilibrium and that the system has magnetic susceptibility χ_L is $(\mu_0/2\chi_L)(M - M_s)^2$. Since the system is near saturation, the longitudinal susceptibility $\chi_L \ll 1$. Not only does χ_L affect the resonance, it is precisely the susceptibility for the longitudinal rf field; a small value of χ_L can explain the lower intensity of the longitudinal resonance than the usual transverse resonance.

(iv) Denoting by ϕ the macroscopic angle associated with rotations about $\vec{M}$, the energy of such rotations is taken to be $(K/2)\phi^2$.

Summing these terms gives the total energy density (using SI units, where M and H are in A/m):

$$\varepsilon = \frac{K}{2}\phi^2 - \mu_0\vec{H}\cdot\vec{M} - \frac{K_u}{M_s^2}(\vec{M}\cdot\hat{y})^2 + \frac{\mu_0}{2\chi_L}(M - M_s)^2 + \frac{\mu_0}{2}(\vec{M}\cdot\hat{z})^2. \tag{4}$$

Reference [1] employed $K/2$ for the energy associated with ϕ, whereas Suran employed K_u for the uniaxial anisotropy. We expect K_u to be associated with the smallest energy in the problem, and we later will drop this term.

To study the longitudinal resonance, we let M vary in magnitude by dM along $\vec{M}$, and we let ϕ vary. We take the gyromagnetic ratio $\gamma > 0$, so $\vec{M}$ is opposite the spin angular

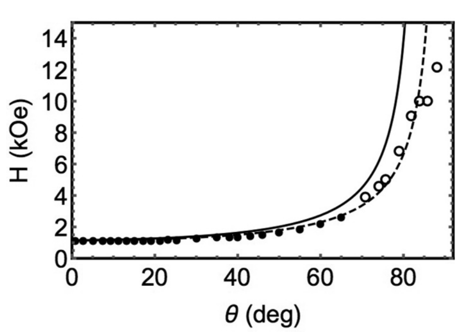

FIG. 3. Data points from Ref. [24], for $\alpha = 0$. Open circles correspond to data with perhaps 25% uncertainty. Theoretical curves from Eq. (14) (solid) and Eq. (15) (dashed) with $M = 10^6$ A/m and $\sin\beta = 0.092$. θ is measured relative to the normal to the plane.

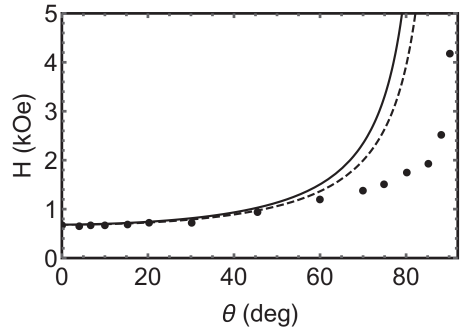

FIG. 4. Data points from Ref. [25]. Theoretical curves from Eq. (14) (solid) and Eq. (15) (dashed) with $M = 10^6$ A/m and $\sin\beta = 0.054$.

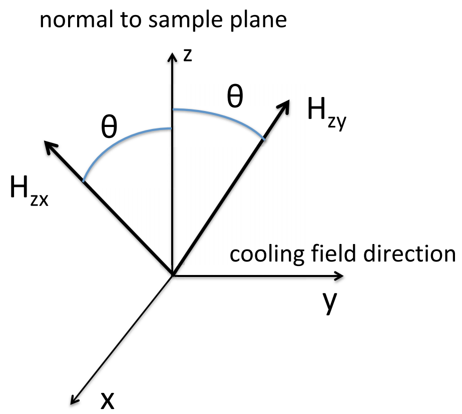

FIG. 5. Geometry for field H_{zy} with cooling field component and field H_{zx} with no cooling field component.

momentum density $\vec{S}$. The equations of motion then are given by [1,27]

$$\frac{\partial\phi}{\partial t} = -\gamma\frac{\partial\varepsilon}{\partial M}, \tag{5}$$

$$\frac{\partial M}{\partial t} = \gamma\frac{\partial\varepsilon}{\partial\phi}. \tag{6}$$

V. EQUILIBRIUM

The experiments considered the two cases where $\vec{H}$ sweeps in the y-z plane from z to y (the cooling field direction), at an angle θ to z [22,23], and where $\vec{H}$ sweeps in the x-z plane from z to x, at an angle θ to z [24,25]. We consider them in succession. Figure 5 illustrates both uses.

$\vec{H}$ in y-z plane. The equilibrium angle β is obtained from the condition that $\vec{M}$ be parallel to the net field $\vec{H}_{\rm net}$ acting on $\vec{M}$. We define

$$H_k = \frac{2K_u}{M_s}. \tag{7}$$

Now, with $\vec{M}$ nearly in the plane (due to the demagnetization field), and nearly along $\hat{y}$ (due to the uniaxial anisotropy), the easy axis field $\vec{H}_u$ satisfies

$$\vec{H}_u = \frac{2K_u}{\mu_0 M_s^2}(\vec{M}\cdot\hat{y})\hat{y} = \frac{2K_u}{\mu_0 M_s}\frac{\vec{M}\cdot\hat{y}}{M_s}\hat{y} \equiv H_k\frac{M}{M_s}\cos\beta\,\hat{y}. \tag{8}$$

Then

$$\vec{H}_{\rm net} = \vec{H} + \vec{H}_{\rm demag} + \vec{H}_u$$
$$= H(\hat{z}\cos\theta + \hat{y}\sin\theta) - (\vec{M}\cdot\hat{z})\hat{z} + \frac{H_k}{M_s}(\vec{M}\cdot\hat{y})\hat{y}. \tag{9}$$

Comparison with Eq. (3) gives

$$\tan\beta = \frac{H\cos\theta - M\sin\beta}{H\sin\theta + H_k\frac{M}{M_s}\cos\beta}. \tag{10}$$

To solve this we define two dimensionless variables

$$\chi \equiv \frac{M}{H}, \qquad s \equiv \frac{H_k}{M_s}, \tag{11}$$

where χ may be thought of as the transverse susceptibility. We expect that $\chi \gg 1$ and $s \ll 1$. Then Eq. (10) becomes

$$\tan\beta = \frac{\cos\theta - \chi\sin\beta}{\sin\theta + \chi s\cos\beta}, \tag{12}$$

from which

$$\chi = \frac{1}{1+s}\left[\frac{\cos\theta}{\sin\beta} - \frac{\sin\theta}{\cos\beta}\right] = \frac{1}{1+s}\frac{\cos(\theta+\beta)}{\sin\beta\cos\beta}. \tag{13}$$

For $s \approx 0$ we have

$$\frac{H}{M} = \frac{1}{\chi} \approx \frac{1}{\frac{\cos\theta}{\sin\beta} - \frac{\sin\theta}{\cos\beta}}. \tag{14}$$

Largely due to the demagnetization field, $\vec{M}$ tips more toward the plane than does $\vec{H}$, so $\sin\beta < \cos\theta$. A stringent test of this equilibrium theory would be to measure H, M, β, and θ directly.

For small β, and θ not near 90°, (14) approximates to

$$\frac{H}{M} = \frac{1}{\chi} \approx \frac{\sin\beta}{\cos\theta}. \tag{15}$$

The next section, on dynamics, shows that at fixed frequency f, $\sin\beta$ is fixed. Therefore, for small β and θ not near 90°, Eq. (15) yields $H\cos\theta \approx M\sin\beta = $ const, in agreement with experiment, a central result of this work.

$\vec{H}$ in x-z plane. This situation is simpler than the previous case, because by symmetry there is no component in the y direction ($\beta = 90°$), and since $M_y = 0$, by (8) the uniaxial anisotropy field $\vec{H}_u = \vec{0}$ in equilibrium. (We consider small uniaxial anisotropy, but note that a large enough uniaxial anisotropy would lead to spontaneous symmetry breaking with $M_y \neq 0$.) Hence the results of the previous section also apply, with reinterpretation of θ and β, but with s literally set to zero in (13). That means for a sweep of increasing angle θ in the x-z plane the field H diverges less rapidly as θ increases than for a sweep of θ in the y-z plane.

Figure 3 for H vs θ shows that the theoretical curve of Eq. (24) compares favorably with the experimental data. Likewise, Fig. 4 for H vs θ shows that the theoretical curve of Eq. (24) compares favorably with the experimental data. In each figure, M is taken to be 10^6 A/m—a reasonable value for ferromagnets—and $\sin\beta$ is taken to fit the experimentally observed value of H at $\theta = 0$.

VI. LONGITUDINAL RESONANCE THEORY

We now derive the crucial result that at fixed resonator frequency $\sin\beta$ is constant, and therefore by (15) that $H\cos\theta$ is approximately constant—except for large θ, where (15) breaks down.

The equation of motion for M follows from (4) and (6):

$$\frac{\partial M}{\partial t} = \gamma K\phi. \tag{16}$$

The equation for ϕ is somewhat more complex. For simplicity we neglect the uniaxial anisotropy. Since we require

$\partial\varepsilon/\partial M$, we need consider only

$$\varepsilon(M) = -\mu_0\vec{H}\cdot\vec{M} + \frac{\mu_0}{2\chi_L}(M-M_s)^2 + \frac{\mu_0}{2}(\vec{M}\cdot\hat{z})^2$$
$$= -\mu_0\sin(\theta+\beta)HM + \frac{\mu_0}{2\chi_L}(M-M_s)^2 + \frac{\mu_0}{2}\sin^2\beta M^2. \quad (17)$$

In equilibrium, $\partial\varepsilon(M)/\partial M = 0$, which gives

$$M_{\rm eq} \approx M_s + \chi_L[H\sin(\theta+\beta) - M_s\sin^2\beta] \approx M_s. \quad (18)$$

The second derivative of (17) gives $\partial\varepsilon/\partial M$ when $M = M_{\rm eq} + dM$, with

$$\frac{\partial\varepsilon(M)}{\partial M} = \mu_0\left(\frac{1}{\chi_L} + \sin^2\beta\right)dM. \quad (19)$$

Then (19) and (5) yield

$$\frac{\partial\phi}{\partial t} = -\gamma\mu_0\left(\frac{1}{\chi_L} + \sin^2\beta\right)dM. \quad (20)$$

We now define

$$\omega_0^2 \equiv \frac{\gamma^2\mu_0 K}{\chi_L}, \quad (21)$$

neglect the uniaxial anisotropy, and assume that both ϕ and dM vary as $e^{i\omega t}$. Then, combining the two dynamical equations gives

$$\frac{\partial^2(dM)}{\partial t^2} = -\omega_0^2(1+\chi_L\sin^2\beta)(dM). \quad (22)$$

Using the small β approximation of (15) gives the resonance frequency

$$\omega^2 = \omega_0^2(1+\chi_L\sin^2\beta) = \omega_0^2(1+\chi_L\sin^2\beta). \quad (23)$$

Hence, for fixed ω, as in most of the experiments, $\sin\beta$ is fixed. By (14) with small $\sin\beta$ (and θ not near 90°), we then have

$$\omega^2 = \omega_0^2\left[1 + \chi_L\left(\frac{H\cos\theta}{M}\right)^2\right]. \quad (24)$$

At fixed ω, this gives $H \sim (\cos\theta)^{-1}$, agreeing with experiment.

To compare with experiments for $\theta = 0$ ($\vec{H}$ normal to the plane, as in Ref. [20]), we note that (15) leads to $\sin\beta \approx \chi^{-1} = H/M$, so that (23) leads to

$$\left(\frac{H}{M}\right)^2 = \frac{1}{\chi_L}\left(\frac{\omega^2}{\omega_0^2} - 1\right), \quad (25)$$

$$H = \frac{M}{\sqrt{\chi_L}}\sqrt{\frac{\omega^2}{\omega_0^2} - 1}. \quad (26)$$

To compare with the data of Fig. 2 above, note that there are two free parameters: $M/\sqrt{\chi_L}$ and ω_0 (due to the macroscopic random anisotropy). If the system were fully characterized, then the first would be determined by static measurements. Using Eq. (26) the form $H = C\sqrt{f^2 - f_0^2}$, where $C = M/(\sqrt{\chi_L}f_0)$, the values $C = 105$ kOe/GHz and $f_0 = 2$ GHz give a reasonable representation to the data. (However, $f_0 = 3$ GHz also works.) Use of $f_0 = \omega_0/(2\pi)$ and a characteristic value [2] $M = 1.0\times10^6$ A/m then gives $\chi_L = 3.6\times10^{-3}$.

The above discussion neglects any coupling between the transverse component $d\vec{M}_\perp$ and the LR degrees of freedom. In fact, $d\vec{M}_\perp$ contributes to $\partial\varepsilon/\partial M$, and thus to $d\phi/dt$, via the associated change in demagnetization field. Moreover, the longitudinal component of dM couples to $d\vec{M}_\perp/dt$ via the associated change in demagnetization field. This leads to coupled modes; such a possibility was indicated in the experiments [24,25].

VII. SUMMARY AND CONCLUSIONS

We have summarized the experimental work of Suran and co-workers on thin films of ferromagnets with random anisotropy, which established the fundamental result that such ferromagnets can support a longitudinal resonance. The theory for the bulk longitudinal resonance does not apply to their results. However, including the effect of the demagnetization field yields good agreement with the experimental result that $H \sim (\cos\theta)^{-1}$ for $\theta \sim\geqslant 80°$, and we reproduce the general trend of H saturating for $80° \leqslant \theta \sim\leqslant 90°$.

As noted earlier, for large angles the present theory does not apply because the four variables (all three components of $\vec{M}$, and the angle ϕ) are coupled. This is due to the dynamical demagnetization field.

Better sample characterization would permit even more stringent tests of the theory. Additional experiments, (1) reproducing the general trends, (2) studying H very accurately at large θ, (3) detailing the shape of the longitudinal resonance line, and (4) studying the resonance at different temperatures, would shed light on this fundamental aspect of ferromagnetism.

Finally, we note that there are additional unexplained aspects to the experiments, largely because the properties of the FRAs studied seem to be nearly independent of the dopant and its concentration.

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