Surface Roughness Induced Extrinsic Damping in Thin Magnetic Films

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The ferromagnetic relaxation caused by the surface roughness induced 2-magnon scattering is investigated. Approximate analytical solution predicts nonexponential decay of the uniform precession excitations of the form $\exp\{-|t/\tau_{3/2}|^{3/2}\}$. This behavior as well as the dependence of the decay time $\tau_{3/2}$ on roughness parameters are confirmed by micromagnetic simulations.

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Recent state-of-the-art switching experiments in ferromagnetic films [1–4] have fueled new interest in the problem of ferromagnetic relaxation. In these experiments, magnetization of a film is driven away from the equilibrium direction at the initial moment, and its following evolution is detected with a temporal resolution of a few picoseconds. In this Letter, we theoretically investigate roughness induced ferromagnetic damping under conditions typical of these recent experiments. It is widely believed that these single-switching experiments provide the same information about the ferromagnetic damping as the usual ferromagnetic resonance (FMR) experiments. In particular, it is expected that magnetization deviation from equilibrium decays exponentially with time: $\exp\{-t/\tau_1\}$. The decay time τ_1 is calculated using the Fermi golden rule and is the same as the time extracted from the FMR linewidth. In this work we will show that for surface roughness induced 2-magnon scattering, even in the small-angle (linear) regime, these intuitive conclusions are wrong. The decay of the uniform precession cannot be described by the Landau-Lifshitz-Gilbert (LLG) damping term, and it follows the faster than exponential $\sim \exp\{-|t/\tau_{3/2}|^{3/2}\}$ law. The decay time $\tau_{3/2}$ is not equal to the Fermi golden rule time τ_1, as measured by FMR. We will also demonstrate that for realistic magnitudes of the surface roughness its contribution to the ferromagnetic relaxation can dominate the intrinsic LLG damping.

It is well known that inhomogeneities in bulk ferromagnets induce 2-magnon scattering, which contributes to ferromagnetic relaxation [5]. In insulating yttrium iron garnet, the surface defect induced scattering was found to be the dominating damping mechanism in all but the best polished samples [6]. 2-magnon scattering, induced by spherical pits on the surface of thick films, was considered by Hurben and Patton [7]. The contribution of 2-magnon scattering to the FMR linewidth in thin films was discussed by McMichael *et al.* [8]; however, no explicit microscopic treatment of inhomogeneities was provided. Arias and Mills [9] considered contributions to the FMR response by small-size square-shape defects on the surfaces of thin films. In the present work, we study the ferromagnetic damping arising from the long range fluctuations of the film thickness. These fluctuations result in nonuniform dipole-dipole fields which couple the uniform mode to the nonuniform magnons, thus allowing the 2-magnon scattering.

The roughness of a film surface causes fluctuations of the z coordinate of the surface points (the z axis is perpendicular to the film plane): $z_s(x, y) = \langle z_s\rangle + \mathcal{Z}(x, y)$. Here $\langle z_s\rangle$ denotes the averaged z coordinate of the surface. The random variable $\mathcal{Z}(\mathbf{r})$ is described by a correlation function: $\langle \mathcal{Z}(\mathbf{r})(\mathbf{r}')\rangle = \sigma^2 f(|\mathbf{r} - \mathbf{r}'|/R)$. Here σ is the standard deviation of the surface z coordinate from the average, R is the correlation radius, and the function $f(x)$ is presumed to decay quickly with x. The correlation function can be measured, for example, in an atomic force microscopy experiment.

We will consider the usual case $\sigma \ll R$. We also assume that our films are sufficiently thin, so that the film magnetization $\mathbf{M} = \mathbf{M}(x, y)$ is uniform throughout the film thickness D_z. The demagnetization field from the rough surfaces was calculated previously [10,11], and, for the top rough surface of the film, is given by

$$\tilde{H}^{\alpha}_{\mathbf{k}} = \sum_{\beta\,\mathbf{k}'} K^{\alpha\beta}_{\mathbf{k}'} M^{\beta}_{\mathbf{k}'\mathbf{k}-\mathbf{k}'}; \tag{1}$$

$$K^{\alpha\beta}_{\mathbf{k}} = -2\pi\frac{k_\alpha k_\beta}{k^2}\frac{k}{D_z}; \qquad K^{\alpha z}_{\mathbf{k}} = 2\pi i\frac{k_\alpha}{k}\frac{k}{D_z};$$

$$\alpha, \beta = x, y; \qquad K^{zz}_{\mathbf{k}} = 2\pi\frac{k}{D_z}; \qquad \mathcal{Z}_{\mathbf{k}} \equiv \int\frac{d^2\mathbf{r}}{L_x L_y}e^{i\mathbf{k}\mathbf{r}}\mathcal{Z}(\mathbf{r}).$$

Here we used $k\sigma \ll 1$, justified by the fact that $\sigma \ll R$, and, as we will show below, only long wavelength magnons $k \lesssim 1/R$ are excited by surface imperfections with correlation length R. The field from the bottom surface has a similar form.

We use geometry and initial conditions similar to the time-resolved magnetodynamics experiments [1–4]. Initially ($t < 0$), the external magnetic field $\mathbf{H}_0$ is applied in plane to orient the initial equilibrium magnetization at an angle ϕ_0 with the x axis. At $t = 0$, the direction of the field is suddenly switched to align with the x axis, and the time evolution of magnetization is investigated. In contrast with our previous work [12] where we investigated intrinsic nonlinear damping for large angle magnetization rotations, in this work we are interested in the linear response of the system; therefore we study only small-angle rotations of magnetization, typically $\phi_0 \lesssim 1°$.

The magnon Hamiltonian in a flat film can be diagonalized by the Holstein-Primakoff [13] transformation. The definitions that we use here are introduced in [12]. The surface roughness perturbation introduces terms of the type $b_{\mathbf{k}} b^{\dagger}_{\mathbf{k}'}$ into the Hamiltonian. These terms correspond to a 2-magnon process, in which one magnon is annihilated and another is created via interaction with surface defects. In experiments, the evolution of the average magnetization of the film, which corresponds to the $k = 0$ magnons, is observed. Therefore we keep only 2-magnon terms in the Hamiltonian, which scatter the uniform $k = 0$ magnons into the nonuniform $k \neq 0$ magnons:

$$\mathcal{H}/\hbar = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b_{\mathbf{k}} b^{\dagger}_{\mathbf{k}} + S_{\mathbf{k}} b_0 b^{\dagger}_{\mathbf{k}} + S^{\dagger}_{\mathbf{k}} b^{\dagger}_0 b_{\mathbf{k}}. \quad (2)$$

Thus we neglect all the so-called secondary processes [14] in which nonuniform magnons are scattered into other nonuniform magnons. Our micromagnetic simulations, which include all scattering processes, show that this approximation works very well in the present case.

The main contribution to the 2-magnon scattering element comes from the term with $K^{xx}_{\mathbf{k}}$ in Eq. (1). The $K^{yy}_{\mathbf{k}}$ and $K^{xy}_{\mathbf{k}}$ terms are small because they contain a $\sin\varphi_{\mathbf{k}}$ factor (where $\varphi_{\mathbf{k}}$ is the angle between $\mathbf{k}$ and the x axis), which, as we will see later, is small for $H_0 \ll 4\pi M_s$ because of the energy conservation requirement. The $K^{zz}_{\mathbf{k}}$, $K^{zx}_{\mathbf{k}}$, and $K^{zy}_{\mathbf{k}}$ terms are small because of the high ellipticity of the magnetization rotation in the present geometry: $M_z/M_y \sim \sqrt{(H_0/4\pi M_s)}$. Among the $K^{xx}_{\mathbf{k}} Z_{\mathbf{k}-\mathbf{k}'} M^x_{\mathbf{k}'} M^x_{-\mathbf{k}}$, the largest terms are those with $k' = 0$, thus $S_{\mathbf{k}} = -\gamma M_s K^{xx}_{\mathbf{k}} Z_{\mathbf{k}} (u_0 u_{\mathbf{k}} + v_0 v_{\mathbf{k}})$.

The classical equations of motion for magnon amplitudes, derived from the Hamiltonian (2) using $i\hbar \dot{b}_{\mathbf{k}} = \partial\mathcal{H}/\partial b^{\dagger}_{\mathbf{k}}$, yield the following integral equation for $B_0 \equiv b_0 \exp(i\omega_0 t)$:

$$\frac{\partial B_0}{\partial t} = -\sum_{\mathbf{k}} |S_{\mathbf{k}}|^2 \int_0^t dt' B_0(t') e^{i[\omega_{\mathbf{k}} - \omega_0](t'-t)}. \quad (3)$$

Equation (3) describes the decay of the uniform precession mode, resulting in the alignment of magnetization with the applied field direction, which constitutes the ferromagnetic relaxation.

An analytic solution of this equation can be obtained for initial stages of the decay, when the change in $B_0(t)$ is small, and thus it can be moved outside the integral at $t' = t$:

$$\left|\frac{B_0(t)}{B_0(0)}\right| \approx \exp\left\{-\sum_{\mathbf{k}} \langle |S_{\mathbf{k}}|^2 \rangle \frac{1 - \cos([\omega_{\mathbf{k}} - \omega_0]t)}{[\omega_{\mathbf{k}} - \omega_0]^2}\right\}. \quad (4)$$

The averaged matrix element $\langle |S_{\mathbf{k}}|^2 \rangle$ is given by

$$\langle |S_{\mathbf{k}}|^2 \rangle \approx (\gamma M_s 2\pi k^2 \cos^2\varphi)^2 \frac{\sigma^2 R^2}{L_x L_y} \frac{\pi M_s}{H_0} F(kR), \quad (5)$$

where the Fourier transformation of the roughness correlation function $F(kR) \equiv \int d^2\mathbf{x} f(x) e^{-iR\mathbf{k}\mathbf{x}}$. Note that the function $F(kR)$ is a quickly decaying function of its argument. For example, in the case of Gaussian correlation function $[f(x) = \exp(-x^2)]$, we have $F(kR) = \pi \exp(-k^2R^2/4)$. It is clear that only the magnons with $k \lesssim 1/R$ contribute to the 2-magnon scattering.

The sum over $\mathbf{k}$ in (4) can be calculated analytically for intermediate times defined by

$$1 \ll \omega_0 t \ll \frac{H_0 M_s R^2}{A}, \quad (6)$$

$$\frac{H_0}{4\pi M_s} \ll \omega_0 t \frac{D_z}{4R} \ll 1. \quad (7)$$

This limits the analytic theory to long correlation lengths and weak fields, both of which are typical in time-resolved experiments. The argument of the cosine in sum (4) oscillates rapidly in time; therefore for long time (6) only magnons with $|\omega_{\mathbf{k}} - \omega_0| \ll \omega_0$ will contribute to the sum. This allows us to approximate the magnon frequencies:

$$\omega_{\mathbf{k}} - \omega_0 \approx a_1 k \sin^2\varphi_{\mathbf{k}} - a_2 k + a_3 k^2; \quad (8)$$

$$a_1 = 4\pi^2\gamma^2 M_s^2 D_z/\omega_0, \quad a_2 \equiv D_z/4, \quad a_3 \equiv 4\pi\gamma^2 A/\omega_0.$$

In correspondence to our assumption of long correlation lengths, $kD_z \sim D_z/R \ll 1$. Because of the left-hand side of (7), the $\sin^2\varphi_{\mathbf{k}}$ term in the argument of cosine in (4) oscillates rapidly with $\varphi\mathbf{k}$; thus only small angles $\varphi \sim \sqrt{H_0 R/(\pi M_s D_z \omega_0 t)}$ contribute to the sum. For these angles, the contribution of the first term in the magnon spectrum (8) dominates over the second and third terms because of the right-hand sides of (6) and (7), and the sum in the Eq. (4) can be evaluated:

$$|B_0(t)/B_0(0)| = \exp\left\{-\left|\frac{t}{\tau_{3/2}}\right|^{3/2}\right\}. \quad (9)$$

The most important feature of Eq. (9) is that the decay of the uniform mode owing to the roughness induced scattering is nonexponential, with the decay time

$$\frac{1}{\tau_{3/2}} = \gamma M_s \left(\frac{M_s}{H_0}\right)^{1/2} \frac{\sigma}{R} \left(\frac{\sigma}{D_z}\right)^{1/3} C, \quad (10)$$

where C is a numerical constant: $C \equiv 2(2\pi)^{3/2} \times \int_0^\infty dy/\sqrt{y} \int_y^\infty dx F(x)$. For example, for the Gaussian correlations $C = [4\sqrt{2}\pi^{7/4}\Gamma_E(3/4)]^{2/3} \approx 13.8$, where Γ_E is the Euler gamma function. Note that the shape of the roughness correlation function does not affect either the character of the uniform mode decay (9) or the scaling (10) of the decay time $\tau_{3/2}$ with the system parameters.

The demagnetization field (1) from the rough surface can be easily introduced into our micromagnetic simulations [12] based on the fast Fourier transformation (FFT) approach [15]. The simulation results for an FeCo film with a Gaussian roughness correlation function are presented in Fig. 1. The decay of the uniform mode follows the $\exp\{-|t/\tau_{3/2}|^{3/2}\}$ law, while the angle decreases by an order of magnitude. Although Eq. (4), which led to the nonexponential dependence (9), was derived under the assumption $|B_0(t) - B_0(0)| \ll B_0(0)$, the simulation results suggest that the theory is applicable over much longer times. The discrepancy between the fit and simulation at $t > 1$ ns is due to the to the violation of this assumption, as well as of the conditions (6) and (7), used in the derivation of (9).

The decay (9) is symmetric with respect to the time reversal because the 2-magnon scattering is elastic and does not involve interaction with the external thermal reservoir. This momentum relaxation reversibility was recently discovered in YIG films [16].

To check the scaling (10) of the decay time with film parameters, we performed a series of micromagnetic simulations, varying applied field H_0, film thickness D_z, roughness standard deviation σ, and correlation length R. The dependence of the decay time $\tau_{3/2}$ on the applied field H_0 is presented in Fig. 2. The increase of the decay time with the applied field $\tau_{3/2} \propto H_0^{1/2}$ can be understood in terms of competition between dipole-dipole energy (which contributes to the roughness induced 2-magnon scattering) and Zeeman energy (which is proportional to applied field). As the applied field increases, it suppresses the roughness induced damping. Also shown is the dependence of the decay time $\tau_{3/2}$ on the film thickness D_z. The decrease of the decay time with the decrease of the film thickness (for a fixed roughness amplitude σ) reflects the fact that the surface becomes increasingly important in thinner films. The dependence of the decay time $\tau_{3/2}$ on the roughness standard deviation σ is shown in Fig. 2 (lower panel). Not surprisingly, the decay time decreases as the amplitude of roughness fluctuation increases. Also shown is the dependence of the decay time $\tau_{3/2}$ on the roughness correlation length R. The roughness induced damping time increases as the roughness correlation radius increases, making the film surface flatter. Overall, our approximate analytic theory is in very good quantitative agreement with our micromagnetic simulations.

Next, we analyze a time-resolved experiment [17] in CoFeHfO films, which are promising for high-frequency magnetic sensors. The deviation of the Fourier transformation of the inductive response from the Lorentzian shape [Fig. 2(b) from [17]] indicates nonexponential uniform mode decay with time. To check whether this discrepancy can be explained by the surface roughness induced damping, we performed simulations without intrinsic LLG damping ($\alpha = 0$). Thus the relaxation in our simulation is solely due to the roughness induced 2-magnon scattering. We chose the Gaussian surface roughness parameters $\sigma = 100$ Å and $R = 3$ μm, which cause decay similar to that observed in experiment. Simulation results are presented in Fig. 3. In the frequency domain (Fig. 3), the simulation with surface roughness agrees very well with the experimental data,

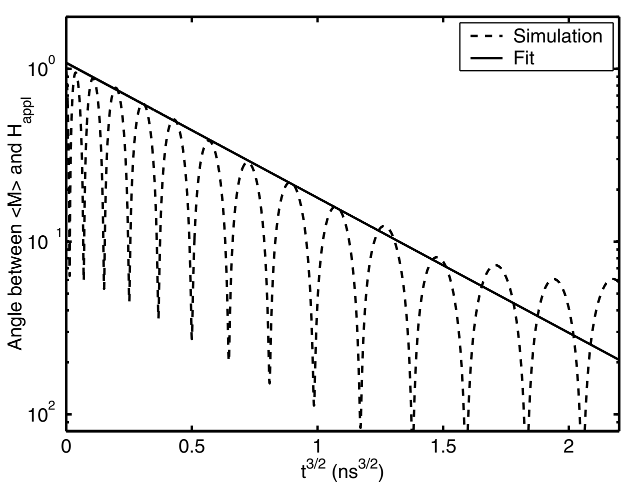

FIG. 1. Time evolution of the angle between the average magnetization of the film and the applied field direction. Calculation parameters: $M_s = 1950$ emu/cm^3, $\gamma = 1.76 \times 10^1$(Oe s)$^{-1}$, $A = 2.4 \times 10^{-6}$ erg/cm^3, $H_K = 0$; $D_z = 100$ Å, $H_0 = 200$ Oe, $\phi_0 = 1°$; $\sigma = 10$ Å, $R = 5000$ Å.

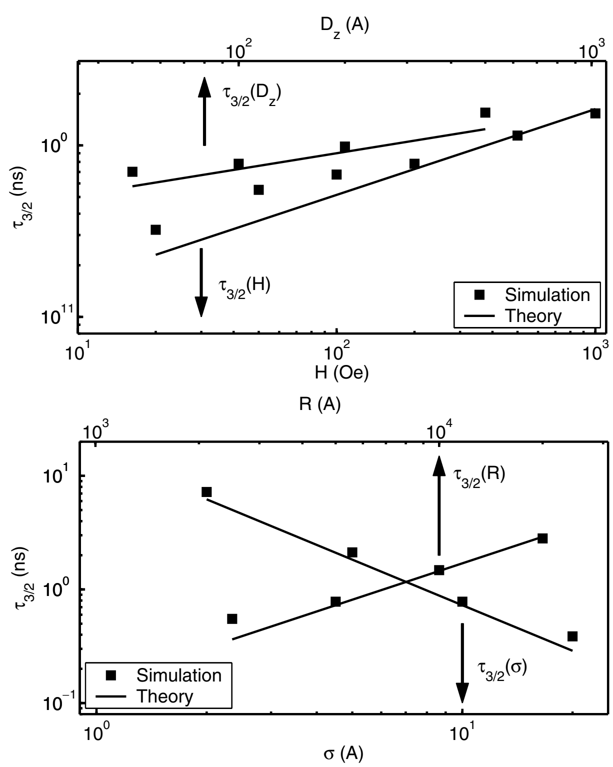

FIG. 2. The dependence of the decay time $\tau_{3/2}$ on the applied field H_0, film thickness D_z, surface roughness standard deviation σ, and correlation length R. Calculation parameters same as in Fig. 1.

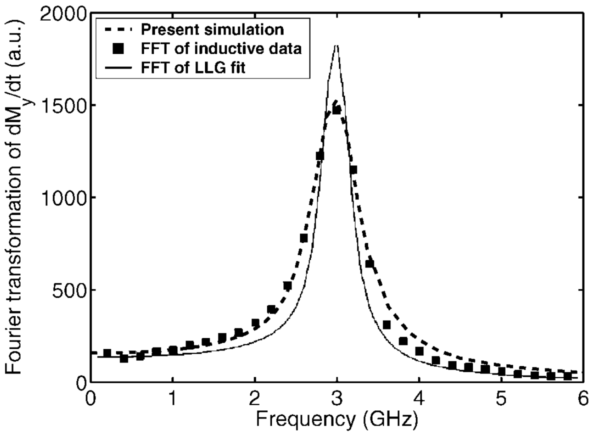

FIG. 3. Fourier transformation of the simulated time dependence of the inductive response in the CoFeHfO film superimposed on the experimental data from [17]. The simulation curve is normalized to match experimental data at low fields. Simulation parameters: $D_z = 100$ nm, $g = 2.15$, $M_s = 800\ \mathrm{emu/cm^3}$, $H_{\mathrm{appl}} = 40.2$ Oe, $H_K = 56.5$ Oe, $\alpha = 0$, $\sigma = 100$ Å, $R = 3\ \mu$m.

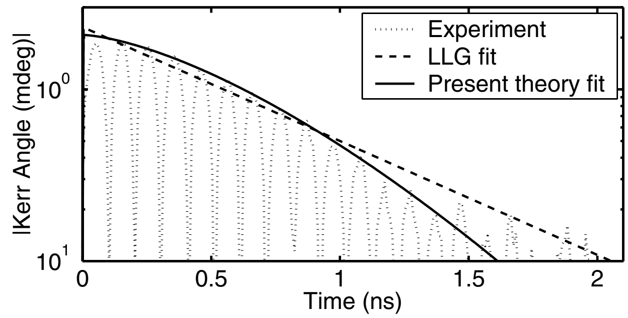

FIG. 4. LLG ($\alpha_{\mathrm{LLG}} \approx 0.0073$) and present theory ($\tau_{3/2} \approx 0.77$ ns) fits to the experimental data. $Fe_{0.5}Co_{0.5}$ film, $D_z = 185$ Å, $M_s = 1900\ \mathrm{emu/cm^3}$, $H_{\mathrm{appl}} = 110$ Oe.

providing a much better fit than the LLG damping term. In addition, the surface roughness parameters that we have chosen are consistent with physically measured values for sputtered thin films.

The time-resolved experimental data for a molecular-beam epitaxy grown $Fe_{0.5}Co_{0.5}(100)/GaAs(100)$ thin film [18], as well as LLG and our theory fits, are presented in Fig. 4. The LLG fit does not follow the experimental decay very well, while our $\sim\exp\{-|t/\tau_{3/2}|^{3/2}\}$ law is much closer to experimental data over a long time interval, until the excitation is reduced by an order of magnitude. The observed decay (the fitted value $\tau_{3/2} \approx 0.77$ ns) can be described with the only damping, induced by the surface roughness with $\sigma = 10$ Å and $R = 0.5\ \mu$m. The experimental correlation length R was determined to be $0.5\ \mu$m by analyzing the height-height correlation function generated over a $5\ \mu$m scan by an atomic force microscope in the "tapping mode" [18]. The experimental roughness σ was 13 Å, which, however, included some conformal roughness (which does not contribute substantially to the 2-magnon scattering). As might be expected from Eq. (10), application of higher fields to the same sample makes the $t^{3/2}$ law progressively more difficult (or impossible) to detect, as the proposed mechanism becomes less important.

In conclusion, we demonstrated that the surface induced 2-magnon scattering contributes substantially to ferromagnetic damping in thin films. The decay of the uniform mode is found to be nonexponential: $\sim\exp\{-|t/\tau_{3/2}|^{3/2}\}$. This surprising behavior is confirmed by our micromagnetic simulations in a film with realistic roughness parameters. Our mechanism may explain nonexponential decay in several time-resolved experiments. Furthermore, the predicted variations with the surface roughness and other film properties provide the opportunity for the other aspects of the theory to be systematically tested experimentally.

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