Intrinsic Nonlinear Ferromagnetic Relaxation in Thin Metallic Films

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We propose an intrinsic mechanism for ferromagnetic relaxation in thin films that can dominate competing linear mechanisms even for rapidly relaxing metals. In particular, we use an analytic theory of four-magnon scattering to demonstrate rapid decay for technologically important systems involving high moment materials subject to large rotations. A micromagnetic simulation is used to verify the results.

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Determination of the mechanism for elimination of the magnetostatic energy from a system subject to a change in magnetic field is one of the fundamental problems of magnetism. This topic, labeled as ferromagnetic relaxation or damping, was first studied extensively in insulators, where the dominant mechanisms were identified in several important cases [1]. The situation in metals remains less clear owing to the high conductivity and the high defect density. Experimentally, most information about energy loss was derived from the ferromagnetic resonance (FMR) linewidth. However, this information is limited to small rotations of the magnetization, with less sensitivity to the rich collection of nonlinear phenomena associated with large rotations [2,3]. Furthermore, large rotations are technologically very important. In particular, magnetic recording heads and media are expected to change magnetization directions over large angles at ever increasing frequencies that may soon surpass 1 GHz.

In view of this, interest has recently shifted towards isolated switching experiments in thin metallic films [4–6]. In this Letter, we investigate theoretically ferromagnetic relaxation in thin films under conditions typical of these recent experiments. The dynamics of large angle magnetization rotation is found to be very different from that in the usual FMR regime. We will show that an important mechanism of ferromagnetic damping in this case (in contrast to the linear FMR case) is the intrinsic four-magnon scattering process, which transfers energy from initially excited uniform precession mode to $k \neq 0$ magnons. To quantitatively describe this mechanism we develop an analytic theory, related to that used for treatment of Suhl instabilities in high-power-level problems [7]. The results of the analytic theory are confirmed by micromagnetic simulations. We will demonstrate that the damping rate for large angle rotations in iron and iron-cobalt films is strongly enhanced with respect to the linear (FMR) values.

We consider a film of thickness D_z and infinite in-plane dimensions (x and y), made of a material with cubic (fourfold) magnetocrystalline anisotropy, with x, y, z aligned with the easy axes [100], [010], [001]. In our analytic theory we take the films to be sufficiently thin that the magnetization along z is uniform, i.e., no $k_z \neq 0$ magnons are excited. Initially ($t = 0$), the external magnetic field $\mathbf{H}_{\text{appl}}$ is applied in-plane to orient the initial equilibrium magnetization at an angle ϕ_0 with x axis. At $t = 0$ the direction of the field is suddenly switched to align with the x axis, and the evolution of magnetization is investigated.

Within the 2D macroscopic continuum approach the Zeeman, magnetostatic (dipole-dipole) and exchange contributions to the total energy can be conveniently expressed in terms of Fourier components $\mathbf{M}_\mathbf{k} \equiv \int d\mathbf{r}/V \exp(i\mathbf{k}\mathbf{r})\,\mathbf{M}(\mathbf{r})$ of magnetization:

$$E_Z = -V(\mathbf{H}_{\text{appl}}\cdot\mathbf{M}_0); \qquad E_D = V\sum_{\mathbf{k};\,\alpha\beta} \mathcal{T}_\mathbf{k}^{\alpha\beta} M_\mathbf{k}^\alpha M_{-\mathbf{k}}^\beta;$$
$$E_{\text{exc}} = V\sum_\mathbf{k} Ak^2\,(\mathbf{M}_\mathbf{k}\cdot\mathbf{M}_{-\mathbf{k}}). \tag{1}$$

Here V is the sample volume, k is a 2D magnon wave vector (k_x, k_y), the demagnetization tensor is given by [8]: $\mathcal{T}_\mathbf{k}^{zz} = 2\pi[1 - G(kD_z)]$; $\mathcal{T}_\mathbf{k}^{z\alpha} = \mathcal{T}_\mathbf{k}^{\alpha z} = 0$; $\mathcal{T}_\mathbf{k}^{\alpha\beta} = 2\pi\, G(kD_z)\cdot k^\alpha k^\beta/k^2$, where $\alpha, \beta = x, y$ and $G(x) \equiv 1 - (1 - e^{-x})/x$. The cubic anisotropy energy is given by $E_A = -K_1/2M_s^4 \cdot \int d\mathbf{r}\,(M_x^4 + M_y^4 + M_z^4)$. This continuum approach is valid until magnons with wavelength near the lattice constant become thermally excited close to the Curie temperature.

In order to diagonalize the Hamiltonian corresponding to the total energy $E_{\text{tot}} = E_Z + E_{\text{exc}} + E_D + E_A$, we apply the Holstein-Primakoff transformation [1] from magnetization $\mathbf{M}(\mathbf{r})$ to elliptical magnon variables $b_\mathbf{k}$ and their complex conjugate $b_\mathbf{k}^\dagger$:

$$M_z + iM_y = 2a\cdot\sqrt{\mu(M_s - \mu\, aa^\dagger)}, \qquad \mu \equiv \hbar\gamma/2V;$$
$$a(\mathbf{r}) = \sum_\mathbf{k}(u_\mathbf{k} b_\mathbf{k} + v_\mathbf{k} b_{-\mathbf{k}}^\dagger)\, e^{i\mathbf{k}\mathbf{r}}; \qquad u_\mathbf{k} = \cosh\xi_\mathbf{k},$$
$$v_\mathbf{k} = -\sinh\xi_\mathbf{k}, \qquad \tanh 2\xi_\mathbf{k} = B_\mathbf{k}/A_\mathbf{k};$$
$$A_\mathbf{k} = H_{\text{appl}} + 2K_1/M_s + 2A/M_s\cdot k^2 + M_s[\mathcal{T}_\mathbf{k}^{yy} + \mathcal{T}_\mathbf{k}^{zz}];$$
$$B_\mathbf{k} = M_s[\mathcal{T}_\mathbf{k}^{zz} - \mathcal{T}_\mathbf{k}^{yy}]. \tag{2}$$

The classical quantities $b_{\mathbf{k}}$ and $b_{\mathbf{k}}^{\dagger}$ correspond to quantum magnon annihilation and creation operators, thus the number of magnons with wave vector $\mathbf{k}$ is given by $N_{\mathbf{k}} = b_{\mathbf{k}} b_{\mathbf{k}}^{\dagger}$. Following Suhl's approach [7] we keep only the 4th order terms which couple $k = 0$ with $k \neq 0$ magnons:

$$\mathcal{H} = \hbar \sum_{\mathbf{k}} \omega'_{\mathbf{k}} b_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} + \Psi_{\mathbf{k}} b_0 b_0 b_{\mathbf{k}}^{\dagger} b_{-\mathbf{k}}^{\dagger} + \Psi_{\mathbf{k}}^{\dagger} b_0^{\dagger} b_0^{\dagger} b_{\mathbf{k}} b_{-\mathbf{k}}. \tag{3}$$

Here, $\omega'_{\mathbf{k}} = \omega_{\mathbf{k}} + \Lambda_{\mathbf{k}} N_0$. The $\Lambda_{\mathbf{k}}$ term describes the nonlinear correction to the linear magnon frequencies $\omega_{\mathbf{k}} = \gamma\sqrt{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}$:

$$\begin{aligned}
\frac{\Lambda_{\mathbf{k}}}{\mu^2 V} &= \sum_{\alpha} L_{\mathbf{k}}^{\alpha} (Ak^2 + \mathcal{T}_{\mathbf{k}}^{\alpha\alpha}) + 2\pi \tilde{L}_{\mathbf{k}}^{z} + \frac{K_1}{M_s^2} L_{\mathbf{k}}^{A}; \\
L_{\mathbf{k}}^{x} &= 8[u_0 u_{\mathbf{k}} + v_0 v_{\mathbf{k}}]^2 + 8[u_0 v_{\mathbf{k}} + v_0 u_{\mathbf{k}}]^2; \\
L_{\mathbf{k}}^{y(z)} &= -4[u_{\mathbf{k}} - (+) v_{\mathbf{k}}]^2 [u_0^2 - (+)\, u_0 v_0 + v_0^2]; \\
\tilde{L}_{\mathbf{k}}^{z} &= -4[u_0 + v_0]^2 [u_{\mathbf{k}}^2 + u_{\mathbf{k}} v_{\mathbf{k}} + v_{\mathbf{k}}^2]; \\
L_{\mathbf{k}\neq 0}^{A} &= -72[u_0^2 + v_0^2][u_{\mathbf{k}}^2 + v_{\mathbf{k}}^2] - 192 u_0 v_0 u_{\mathbf{k}} v_{\mathbf{k}}; \\
L_0^{A} &= -18[u_0^2 + v_0^2]^4 - 24 u_0^2 v_0^2.
\end{aligned} \tag{4}$$

The $\Psi_{\mathbf{k}}$ terms correspond to the four-magnon scattering process (two $k = 0$ magnons annihilate, k and $-k$ magnons are created):

$$\begin{aligned}
\frac{\Psi_{\mathbf{k}}}{\mu^2 V} &= \sum_{\alpha} P_{\mathbf{k}}^{\alpha} (Ak^2 + \mathcal{T}_{\mathbf{k}}^{\alpha\alpha}) + 2\pi \tilde{P}_{\mathbf{k}}^{z} + \frac{K_1}{M_s^2} P_{\mathbf{k}}^{A}; \\
P_{\mathbf{k}}^{x} &= 4[u_0 u_{\mathbf{k}} + v_0 v_{\mathbf{k}}]^2; \\
P_{\mathbf{k}}^{y(z)} &= -[u_{\mathbf{k}} - (+) v_{\mathbf{k}}][u_0^2 u_{\mathbf{k}} - (+) 2 u_0 u_{\mathbf{k}} v_0 \\
&\qquad + 2 u_0 v_0 v_{\mathbf{k}} - (+) v_0^2 v_{\mathbf{k}}]; \\
\tilde{P}_{\mathbf{k}}^{z} &= -[u_0 + v_0][u_0 u_{\mathbf{k}}^2 + 2 u_0 u_{\mathbf{k}} v_{\mathbf{k}} \\
&\qquad + 2 u_{\mathbf{k}} v_0 v_{\mathbf{k}} + v_0 v_{\mathbf{k}}^2]; \\
P_{\mathbf{k}}^{A} &= 6[u_0^2 v_{\mathbf{k}}^2 + v_0^2 u_{\mathbf{k}}^2 + 3 u_0^2 u_{\mathbf{k}}^2 + 3 v_0^2 v_{\mathbf{k}}^2 \\
&\qquad + 12 u_0 v_0 u_{\mathbf{k}} v_{\mathbf{k}}].
\end{aligned} \tag{5}$$

These terms are dominant in the Hamiltonian if $N_0 \gg N_{\mathbf{k}}$, which is true for most of the decay, because at $t = 0$ the number of $k \neq 0$ magnons depends on the temperature of the sample: $\overline{N}_{\mathbf{k}} \equiv N_{\mathbf{k}}(t = 0) = 1/[\exp(\hbar\omega_{\mathbf{k}}/T) - 1]$. The number of $k = 0$ magnons is determined by the initial deviation of magnetization from equilibrium: $\overline{N}_0 = V/(2\hbar\gamma M_s)[M_y(t = 0)/(u_0 - v_0)]^2$.

The equations of motion $i\hbar\dot{b}_{\mathbf{k}} = \partial\mathcal{H}/\partial b_{\mathbf{k}}^{\dagger}$ can be easily solved under the assumption $|b_0| = \text{const} = |b_0(t = 0)|$. In this case, $N_{\mathbf{k}}(t) = \overline{N}_{\mathbf{k}} \exp[\Gamma_{\mathbf{k}} t]$ with

$$\Gamma_{\mathbf{k}} = \sqrt{16\,\overline{N}_0^2 |\Psi_{\mathbf{k}}|^2 - (2\omega'_0 - 2\omega'_{\mathbf{k}})^2}. \tag{6}$$

Note, that the second term under square root enforces conservation of energy in these processes, namely $2\omega'_0 = \omega'_{\mathbf{k}} + \omega'_{-\mathbf{k}} = 2\omega'_{\mathbf{k}}$. However, the energy conservation requirement is broadened by the first term, because scattering is allowed for all modes for which the first term is bigger than the second.

The decrease of $N_0(t)$ with time owing to this four-magnon scattering leads to ferromagnetic damping, i.e., alignment of the average magnetization with the applied field direction. Numbers of magnons with $\Gamma_{\mathbf{k}} > 0$ grow exponentially with time, and since the total number of magnons is conserved in the four-magnon process, $N_0(t) = \overline{N}_0 - \sum_{\mathbf{k}} N_{\mathbf{k}}(t)$. Considering $t \gg 1/\Gamma_{\mathbf{k}}$ and using the saddle point approximation we find

$$N_0(t) \approx \overline{N}_0 - V\overline{N}_{\tilde{\mathbf{k}}} C_{\tilde{\mathbf{k}}} t^{-1} \exp[\,\Gamma_{\tilde{\mathbf{k}}}\, t\,], \tag{7}$$

where $C_{\tilde{\mathbf{k}}}^{-2} \equiv 8\pi^2 D_z^2 \det[\partial^2 \Gamma_{\tilde{\mathbf{k}}}/\partial\tilde{k}_i \partial\tilde{k}_j]$, $\tilde{\mathbf{k}}$ is the wave vector which delivers the maximum $\Gamma_{\tilde{\mathbf{k}}}$.

Of course Eq. (7) is valid only while $\overline{N}_0 - N_0(t) \ll \overline{N}_0$; however, we can use it to estimate the $k = 0$ mode decay time with logarithmic accuracy:

$$\tau_{4m} \approx \frac{1}{\Gamma_{\tilde{\mathbf{k}}}} \ln \frac{\overline{N}_0}{V\overline{N}_{\tilde{\mathbf{k}}} C_{\tilde{\mathbf{k}}} \Gamma_{\tilde{\mathbf{k}}}}. \tag{8}$$

The dependence of decay time on initial $k \neq 0$ magnon occupation numbers (and thus on temperature) is very weak: $\tau_{4m} \sim \ln[\hbar\omega_{\tilde{\mathbf{k}}}/T]$, in contrast with the strong dependence in the theories which considered interaction of $k = 0$ and thermal magnons [9,10]. In these previous theories the number of thermally excited magnons was presumed constant, yielding a low relaxation rate, while in our theory the numbers of some $k \neq 0$ magnons grow very rapidly with time.

To verify our analytic theory results we performed micromagnetic simulations, in which the standard equations for magnetic precession $d\mathbf{M}/dt = -\gamma[\mathbf{M} \times \mathbf{H}]$, $\mathbf{H} = -\partial E_{\text{tot}}/\partial\mathbf{M}$ are integrated numerically. This approach is similar to that used in Ref. [11], the crucial difference, however, is that in the latter paper samples of small size were investigated, leading to nonintrinsic magnon-magnon scattering mediated by highly nonuniform demagnetization fields at the samples boundaries. In our simulations we use periodic boundary conditions, which allow us to get rid of these finite-size effects. As can be easily seen from the equation of motion, if a simulation in an infinite film starts with a strictly uniform magnetization, the magnetization will stay uniform throughout the simulation, thus there would be no magnon-magnon scattering. In reality, at nonzero temperatures thermally excited $k \neq 0$ magnons make the initial magnetization slightly nonuniform, which allows the relaxation. We model thermal magnons in our system using the Bose-Einstein distribution for the initial magnon occupation numbers. Thick films require calculation of interlayer interaction and Eq. (1) is no longer valid: we employ the method proposed by Ref. [8] to calculate the magnetostatic interaction.

Simulation results are presented in Fig. 1. While decreasing the number of $k = 0$ magnons, magnon-magnon scattering conserves the total number of magnons

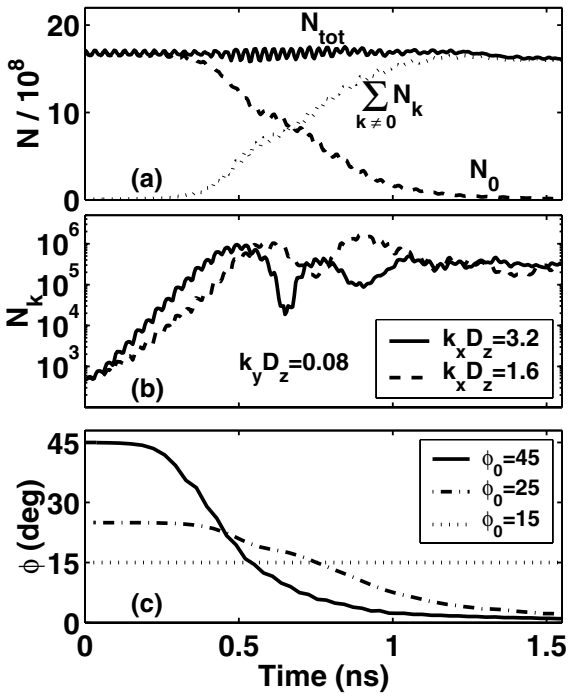

FIG. 1. Time evolution of (a) number of $k = 0$ magnons N_0, number of all $k \neq 0$ magnons $\sum_{\mathbf{k}\neq 0} N_{\mathbf{k}}$, and total number of magnons $N_{\text{tot}} \equiv \sum_{\mathbf{k}} N_{\mathbf{k}}$; (b) magnon numbers $N_{\mathbf{k}}$ with different $\mathbf{k} \neq 0$; (c) envelope of the angle ϕ between the average magnetization and the applied field. Iron film ($M_s = 1700$ emu/cm^3, $\gamma = 1.76 \times 10^7$(Oe·s)$^{-1}$, $A = 2 \times 10^{-6}$erg/cm^3, $K_1 = 4.8 \times 10^5$erg/cm^3); $D_z = 500$ Å, $H_{\text{appl}} = 1000$ Oe, $\phi_0 = 25°$, $T = 300$ K.

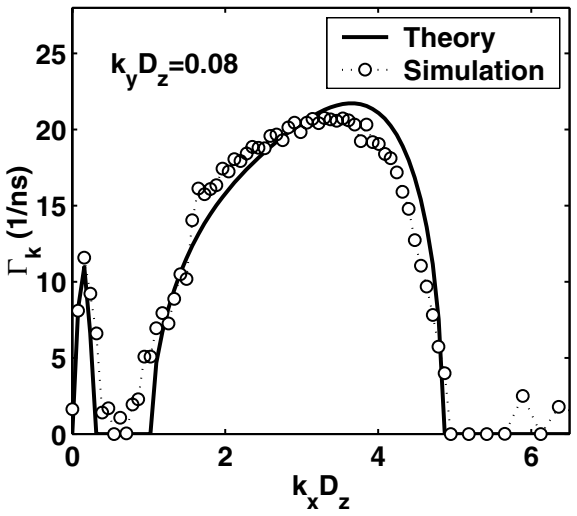

FIG. 2. Theoretical $\Gamma_{\mathbf{k}}^{\text{th}}$, calculated using Eqs. (2) and (4)–(6), and simulation $\Gamma_{\mathbf{k}}^{\text{sim}} \equiv \ln[N_{\mathbf{k}}(t_2)/N_{\mathbf{k}}(t_1)]/t$, $t_2 = 0.2$ ns, $t_1 = 0.1$ ns. Calculation parameters are the same as in Fig. 1.

[Fig. 1(a)], which is consistent with the four-magnon scattering considered above. The simulation performed at different temperatures $30\ \text{K} < T < 600\ \text{K}$ (affecting the initial magnons occupation numbers) show little difference, thus confirming the predicted weak temperature dependence of the four-magnon damping mechanism. The numbers of $k \neq 0$ magnons increase exponentially with time [Fig. 1(b)] in accordance with the predictions of the analytic theory. Figure 2 shows a very good agreement between theoretical and simulation values of instability increments $\Gamma_{\mathbf{k}}$. This agreement demonstrates the validity of the Suhl Hamiltonian [Eq. (3)] for angles as large as 25° and corroborates the findings of our analytic theory.

Note, that our analytic theory describes only initial growth of $k \neq 0$ magnons and decay of the $k = 0$ mode [$t < 0.5$ ns in Figs. 1(a) and 1(b)]. Beyond this initial period $k \neq 0$ magnons do not grow exponentially, however the decay of the $k = 0$ mode (and thus the intrinsic damping) continues at a high rate, because at this second relaxation step $k = 0$ magnons scatter off the exponentially large number of $k \neq 0$ magnons excited in the first step. This scattering is the same four-magnon process, since simulations show that the total number of magnons is still conserved.

Intrinsic relaxation for different magnetization rotation angles is presented in Fig. 1(c). The drastic decrease of the damping time with the magnetization rotation angle reflects closely the strong dependence of $\Gamma_{\mathbf{k}}$ [Eq. (6)] and τ_{4m} [Eq. (8)] on the $k = 0$ mode initial occupation number $\overline{N}_0$. This surprising result (starting farther from equilibrium the system reaches equilibrium faster) is a manifestation of the highly nonlinear nature of magnetization dynamics. Note that the linear (small angle) relaxation time for high-quality iron films is $\tau_{\text{FMR}} \approx 1.2$ ns, as measured in FMR experiments [12]. For $\phi_0 > 20°$, damping time of the four-magnon mechanism is shorter than τ_{FMR}, thus this mechanism dominates in the relaxation for large magnetization rotations.

The variation of the damping time τ_d, defined as $[M_x(\infty) - M_x(0)]/[M_x(\infty) - M_x(\tau_d)] = \exp(1)$, with simulation parameters is presented in Fig. 3. The three-magnon splitting process becomes dominant in thicker films and bulk samples, and is responsible for the sharp drop of τ_d in iron for $D_z > 1000$ Å [Fig. 3(a)]. For the three-magnon scattering energy conservation requires $\omega_0 = 2\omega_{\mathbf{k}}$, which is satisfied in low applied fields ($H_{\text{appl}} < 4\pi/3 \cdot M_s$) in bulk slablike samples. For thin films this requirement cannot be satisfied and thus three-magnon processes are prohibited.

The damping time for different materials is presented in Fig. 3(b). The damping time is reduced relative to the FMR value (1.2 ns) for FeCo [13]. The increase of the damping time in permalloy and nickel with respect to iron shows the importance of the magnetostatic interaction for the magnon-magnon scattering. First, the magnetostatic interaction distorts the magnon dispersion to allow energy conservation $\omega_0 = \omega_{\mathbf{k}}$ for $k \neq 0$; second, it provides the main contribution to the magnon-magnon matrix element $\Psi_{\mathbf{k}}$. It is worth noting that technologically important recording head materials are high moment materials.

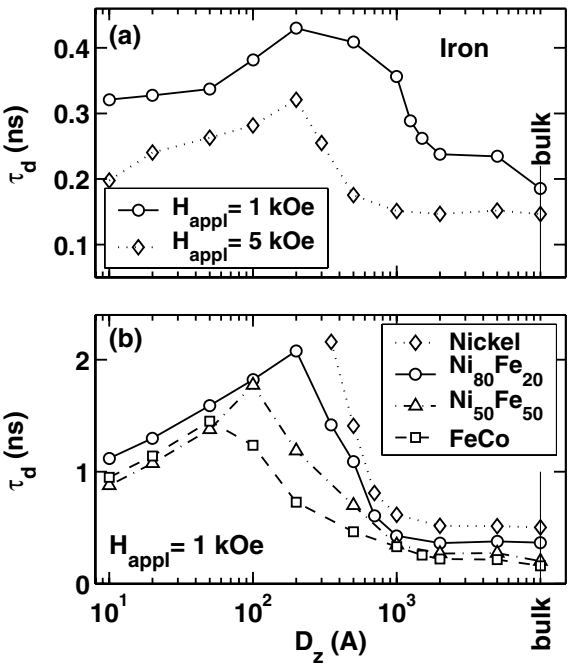

FIG. 3. Damping time τ_d calculated from simulations for $\phi_0 = 45°$ switching angle as a function of film thickness D_z.

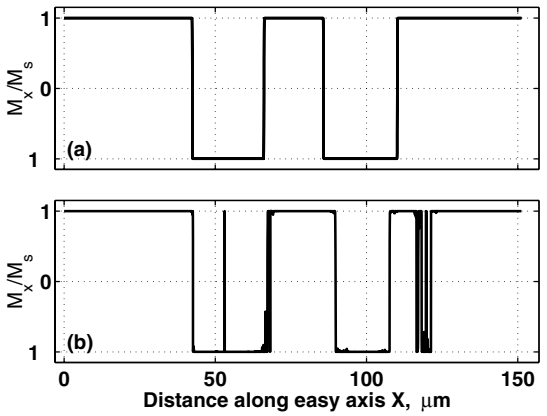

FIG. 4. Switching pattern in the conditions of experiment [4] with (a) $\alpha = 0.037$ and no magnons excitation allowed; (b) $\alpha = 0.005$ and magnon-magnon scattering included. Pattern is shown along easy direction x, for $y = 0$ in terms of paper [4]. $x = 0$ corresponds to the origin in [4]. Initially, $M_x/M_s = 1$; after the field pulse, regions with $M_x/M_s = -1$ have been switched. Note that the rapid fluctuations would likely be experimentally unresolvable.

Back *et al.* [4] observed an effective increase of damping for large magnetization rotation in cobalt films under pulse magnetic field. Our four-magnon scattering mechanism can allow systems with small damping parameter α to appear as though they have much larger α in the large angle regime. In Fig. 4 we present results of our simulations in the conditions of experiment [4]. We find that the four-magnon process with realistic small $\alpha = 0.005$ gives a very similar pattern to $\alpha = 0.037$ (value used to explain experimental results [4]) without four-magnon scattering.

The four-magnon scattering process transfers the initial energy excess of the $k = 0$ mode to $k \neq 0$ magnons, but it does not change the total energy of the magnetic system. To complete ferromagnetic relaxation we need another damping mechanism which would transfer energy away from the magnetic system. A likely energy dissipation mechanism for the thicker films in our study ($D_z > 300$ Å) was proposed by Abrahams [14], in which $k \neq 0$ magnons are strongly scattered by conduction electrons. The use of realistic assumptions about the conductivity, Fermi wave vector, and electron mass yields decay rates of order 1 ns for the magnons excited in our calculations. Other possible mechanisms are magnon-phonon scattering and Kambersky-Korenman-Prange spin-orbit mediated scattering [15]. Although the relaxation time of a single switching experiment is defined by the four-magnon damping time, the energy dissipation mechanism could manifest itself in repeated high-speed switching, where a slower decay rate of excited $k \neq 0$ magnons may become a bottleneck for high switching frequencies.

In conclusion, we propose an intrinsic mechanism of nonlinear ferromagnetic relaxation in thin metallic films. Using analytic theory as well as micromagnetic simulations, we show that the four-magnon scattering process is responsible for a rapid decay of the $k = 0$ mode into $k \neq 0$ magnons. This mechanism dominates competing linear relaxation mechanisms and governs large angle magnetization reversal in technologically important systems.

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