Large Nonlinear Dynamical Response of Superparamagnets: Interplay between Precession and Thermoactivation in the Stochastic Landau-Lifshitz Equation

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The nonlinear dynamical response of classical spins governed by the stochastic Landau-Lifshitz equation is found to be large and very sensitive to the damping in the medium-to-weak damping regime. This result is interpreted in terms of a cooperation, induced by the driving field, between the precession of the spin and its thermoactivation over the potential barrier. The large damping dependence (absent in the linear response) can be used to determine the evasive damping coefficient in superparamagnets, so clarifying the nature of the spin-environment interaction in these systems.

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The Landau-Lifshitz equation for the dynamics of classical spins [1] is an important equation in nonlinear physics, possessing a rich variety of exact solutions and being a practically inexhaustible object for numerical analysis. In statistical physics, its stochastic partner (or, equivalently, the associated Fokker-Planck equation) is the minimal tool for the study of *noninertial* Brownian rotation of dipoles (much as the original Langevin equation is to ordinary Brownian motion). For instance, it constitutes the starting point in the efforts to extend Kramers' theory of thermoactivated barrier crossing to such rotational systems. In addition, if an oscillating field is included, one can find what in modern jargon is called "stochastic resonance" but in rotationally multistable potentials.

This equation also plays an important role in condensed matter physics, being used to describe a variety of electric and magnetic phenomena. For instance, the equations underlying the Debye theory of noninertial dielectric relaxation can *formally* be obtained from the stochastic Landau-Lifshitz equation in the limit of zero precession. In magnetism, the Landau-Lifshitz equation describes spin dynamics in classical XY and Heisenberg models, thin films, and superparamagnets (nanoscale solids or clusters whose net spin $S \sim 10^2 - 10^5$ rotates thermally activated in the anisotropy potential). Furthermore, the Landau-Lifshitz equation may provide an approximate description of high spin ($S \sim 10$) magnetic molecular clusters; it can also be used to incorporate finite-temperature effects in first principles calculations, and it is employed as well in modeling of technological applications as information storage and processing devices.

The study of the *linear* response associated with the stochastic Landau-Lifshitz equation goes back to the above-mentioned Debye theory of dielectric relaxation, and is to some extent an established subject in superparamagnets. The extension of these studies to the *nonlinear* response is important in both the context of the general progress in nonlinear physics and on account of the greater sensitivity of the nonlinear susceptibilities (or hyperpolarizabilities) to important characteristics of the underlying system. Because of its relative complexity, however, this field has progressed at a slower pace.

The rigorous derivation of the nonlinear dynamical susceptibility of isotropic dipoles was carried out in the context of the Debye theory by Coffey and Paranjape [2]. Further developments included the study of anisotropically polarizable molecules and the effects of strong superimposed electric fields [3,4]. The corresponding problem in magnetism has been addressed [5] under the assumptions of *uniaxial* magnetic anisotropy and *overdamped* dynamics (precession term disregarded) [6]. Such a simple symmetry of the anisotropy potential, recently confirmed in an important nanoparticle system [7], suffices to shed some light on this complex dynamical problem. In contrast, although the damping coefficient λ is poorly known, the most reliable determinations [8] indicate that it lies in the medium or weak damping regimes ($\lambda \sim 0.5-0.01$). Moreover, the intrinsic temperature or field dependences of λ [9,10] could bring the system from one damping regime to another. Nevertheless, an investigation of the role of arbitrary damping in this problem is still lacking.

To fill this gap in our knowledge of the dynamics of the stochastic Landau-Lifshitz equation, in this article we study the nonlinear response of spins governed by this equation, fully accounting for the spin precession. We obtain an unexpectedly large dependence of the low-frequency nonlinear susceptibility on the damping coefficient, due to the interplay between the precession of the spin and its thermoactivation over the anisotropy barrier. Such a sensitivity to the damping coefficient, which has no analog in the low-frequency linear response, suggests that λ can experimentally be extracted from the nonlinear susceptibility. This would facilitate the determination of the intrinsic dependences of the damping in superparamagnets (and congeneric systems), thus clarifying its microscopic origin.

Let us begin with a brief discussion of the dynamics of a subsystem of interest (a spin in our case) accounting for its interaction with the surrounding "medium" (lattice

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vibrations, conduction electrons, nuclear spins, etc.). In a variety of systems, this interaction, after the elimination of the explicit dependences on the environment dynamical variables, can be separated into a time-dependent modulation of the subsystem by the proper modes of the environment (fluctuating term), and the back reaction on the subsystem of its action on the surrounding medium (relaxation or damping term). This approach was particularized phenomenologically by Brown [11] and Kubo and Hashitsume [12] to classical spins (superparamagnets), by introducing the *stochastic Landau-Lifshitz equation:*

$$\frac{1}{\gamma}\frac{d\vec{S}}{dt} = \vec{S}\wedge[\vec{B}_{\rm eff} + \vec{b}_{\rm fl}(t)] - \frac{\lambda}{S}\vec{S}\wedge(\vec{S}\wedge\vec{B}_{\rm eff}). \quad (1)$$

In this (Stratonovich) stochastic differential equation, $\vec{B}_{\rm eff} = -\partial\mathcal{H}/\partial\vec{S}$ is the deterministic effective field, the double vector product is the damping term, which rotates $\vec{S}$ towards the potential minima (preserving $|\vec{S}|$), and $\vec{b}_{\rm fl}(t)$ is a fluctuating field with white noise properties:

$$\langle b_{{\rm fl},i}(t)\rangle = 0, \quad \langle b_{{\rm fl},i}(t)b_{{\rm fl},j}(t')\rangle = \frac{2\lambda k_{\rm B}T}{\gamma S}\,\delta_{ij}\delta(t-t').$$

The dimensionless damping coefficient λ measures the relative importance of the relaxation and precession terms and controls the intensity of the fluctuations (so that fluctuation-dissipation relations are obeyed).

For dipoles with the simplest uniaxial magnetic anisotropy in an arbitrarily directed driving field $\Delta\vec{B}(t)$, the Hamiltonian reads

$$\mathcal{H} = -D(S_z/S)^2 - \vec{S}\cdot\Delta\vec{B}(t).$$

The anisotropy term has two minima at $S_z = \pm S$ (the "poles") with a barrier between them at $S_z = 0$ (the "equator"). Let us introduce the spherical harmonics $X_\ell^m(z,\varphi) = e^{{\rm i}m\varphi}P_\ell^m(z)$, where $z = S_z/S$ and φ is the azimuth of $\vec{S}$. The dynamical equations for the X_ℓ^m, averaged over realizations of the fluctuating field, can be obtained *directly* from Eq. (1) [13] and constitute an infinite system of coupled equations:

$$\begin{aligned}
2\tau_{\rm D}\frac{d}{dt}X_\ell^m &+ \left[\ell(\ell+1) - 2\sigma\frac{\ell(\ell+1)-3m^2}{(2\ell-1)(2\ell+3)}\right]X_\ell^m + 2\sigma\frac{\rm i}{\lambda}m\left[\frac{\ell+m}{2\ell+1}X_{\ell-1}^m + \frac{\ell-m+1}{2\ell+1}X_{\ell+1}^m\right] \\
&- 2\sigma\left[\frac{(\ell+1)(\ell+m-1)(\ell+m)}{(2\ell-1)(2\ell+1)}X_{\ell-2}^m - \frac{\ell(\ell-m+1)(\ell-m+2)}{(2\ell+1)(2\ell+3)}X_{\ell+2}^m\right] \\
&= \Delta\xi_z\left[\frac{(\ell+1)(\ell+m)}{2\ell+1}X_{\ell-1}^m - \frac{\rm i}{\lambda}mX_\ell^m - \frac{\ell(\ell-m+1)}{2\ell+1}X_{\ell+1}^m\right] \\
&+ \frac{\Delta\xi_+}{2}\left[\frac{(\ell+1)(\ell+m-1)(\ell+m)}{2\ell+1}X_{\ell-1}^{m-1} + \frac{\rm i}{\lambda}(\ell-m+1)(\ell+m)X_\ell^{m-1}\right. \\
&\left. + \frac{\ell(\ell-m+1)(\ell-m+2)}{2\ell+1}X_{\ell+1}^{m-1}\right] \\
&+ \frac{\Delta\xi_-}{2}\left[-\frac{\ell+1}{2\ell+1}X_{\ell-1}^{m+1} + \frac{\rm i}{\lambda}X_\ell^{m+1} - \frac{\ell}{2\ell+1}X_{\ell+1}^{m+1}\right]. \qquad (2)
\end{aligned}$$

Here, $\sigma = D/k_{\rm B}T$, $\Delta\vec{\xi} = S\Delta\vec{B}/k_{\rm B}T$, $\Delta\xi_\pm = \Delta\xi_x \pm {\rm i}\Delta\xi_y$, and $\tau_{\rm D}$ is the relaxation time in the isotropic ($\sigma \to 0$) limit (the counterpart of the Debye time in dielectrics)

$$\tau_{\rm D} = \frac{1}{\lambda}\frac{S}{2\gamma k_{\rm B}T}. \quad (3)$$

If the Gilbert form is used instead of Eq. (1), one has *only* to replace λ by $\lambda/(1+\lambda^2)$ in $\tau_{\rm D}$.

The equations for the X_ℓ^m can be solved perturbatively in $\Delta\vec{B}$ [5]. The right-hand side of Eq. (2), at a specific order, will be a given function of time depending on the results of the preceding order [denoted by $F_\ell^m(t)$]. Then, on introducing the 2 vectors,

$$\mathbf{C}_\ell = \begin{pmatrix} X_{2\ell-1}^m \\ X_{2\ell}^m \end{pmatrix}, \qquad \mathbf{F}_\ell = \begin{pmatrix} F_{2\ell-1}^m \\ F_{2\ell}^m \end{pmatrix},$$

and the 2×2 matrices,

$$\mathbf{Q}_\ell^- = 2\sigma\begin{pmatrix} -\frac{2\ell(2\ell+m-1)(2\ell+m-2)}{(4\ell-1)(4\ell-3)} & \frac{\rm i}{\lambda}m\frac{2\ell+m-1}{4\ell-1} \\ 0 & -\frac{(2\ell+1)(2\ell+m)(2\ell+m-1)}{(4\ell-1)(4\ell+1)} \end{pmatrix},$$

$$\mathbf{Q}_\ell = 2\sigma\begin{pmatrix} \frac{\ell(2\ell-1)}{\sigma} - \frac{2\ell(2\ell-1)-3m^2}{(4\ell-3)(4\ell+1)} & \frac{\rm i}{\lambda}m\frac{2\ell-m}{4\ell-1} \\ \frac{\rm i}{\lambda}m\frac{2\ell+m}{4\ell+1} & \frac{\ell(2\ell+1)}{\sigma} - \frac{2\ell(2\ell+1)-3m^2}{(4\ell-1)(4\ell+3)} \end{pmatrix},$$

$$\mathbf{Q}_\ell^{+} = 2\sigma \begin{pmatrix} \frac{(2\ell-1)(2\ell-m)(2\ell-m+1)}{(4\ell-1)(4\ell+1)} & 0 \\ \frac{\mathrm{i}}{\lambda}\, m\, \frac{2\ell-m+1}{4\ell+1} & \frac{2\ell(2\ell-m+1)(2\ell-m+2)}{(4\ell+1)(4\ell+3)} \end{pmatrix},$$

the equations for the X_ℓ^m can be cast into the form of an inhomogeneous three-term differential-recurrence relation (in the index ℓ with fixed m):

$$2\tau_{\mathrm{D}} \frac{d\mathbf{C}_\ell}{dt} + \mathbf{Q}_\ell^{-}\mathbf{C}_{\ell-1} + \mathbf{Q}_\ell \mathbf{C}_\ell + \mathbf{Q}_\ell^{+}\mathbf{C}_{\ell+1} = \mathbf{F}_\ell(t).$$

This type of equation can efficiently be solved by using *matrix* continued fraction methods [14]. Then, the average dipole moment (response) of the system is obtained via $\langle S_z \rangle / S = X_1^0$ and $\langle S_x + \mathrm{i}S_y \rangle / S = X_1^1$. Finally, in the presence of an oscillating driving field $\Delta\xi(t) = \frac{1}{2}\Delta\xi(e^{+\mathrm{i}\Omega t} + e^{-\mathrm{i}\Omega t})$, the time-dependent part of the field projection $(\langle \vec{S} \rangle \cdot \Delta\vec{B}/\Delta B)$ of the stationary response can be expanded in Fourier series as follows:

$$\Delta M(t) = \sum_{k=1}^{\infty} \left(\frac{\Delta B}{2}\right)^k (\chi^{(k)} e^{+\mathrm{i}k\Omega t} + \chi^{(k)*} e^{-\mathrm{i}k\Omega t}),$$

which defines the linear $\chi^{(1)}(\Omega)$ (or simply χ) and nonlinear susceptibilities $\chi^{(k)}(\Omega)$, $k = 2, 3 \ldots$.

In the absence of a bias field, the leading nonlinear term is $\chi^{(3)}$, henceforth referred to as the nonlinear susceptibility. Figure 1 displays the "low-frequency" ($\Omega\tau_{\mathrm{D}} \lesssim 1$) nonlinear susceptibility spectra of an ensemble of spins with collinear anisotropy axes and driving field parallel and perpendicular to the axes. The features of $\chi_\parallel^{(3)}(\Omega)$ are *qualitatively* similar to those in the isotropic dipole case [2]: the real part $\chi_\parallel^{(3)\prime}$ equals the thermal-equilibrium susceptibility at low Ω, then it decreases with increasing Ω, changes sign, exhibits a peak, and finally tends to zero at high frequencies. The imaginary part, $\chi_\parallel^{(3)\prime\prime}$, is approximately traced by the logarithmic derivative $-(\pi/2)(\partial \chi_\parallel^{(3)\prime} / \partial \log \Omega)$. Concerning the transverse response, it is dominated by fast ($\sim\tau_{\mathrm{D}}$) intra-potential-well relaxation modes, so that $\chi_\perp^{(3)}(\Omega)$ practically coincides with the corresponding equilibrium susceptibility in the whole $\Omega\tau_{\mathrm{D}} \lesssim 1$ range. Note that, when present, the peaks of the susceptibility curves always have heights that are a fraction of the equilibrium $\chi^{(3)}$. Note also that the susceptibilities are independent of the damping coefficient, or rather, λ enters only through the isotropic relaxation time τ_{D} [Eq. (3)] shifting the curves along the frequency axis, and this dependence collapses onto a single master curve when representing the spectra as a function of the natural variable $\Omega\tau_{\mathrm{D}}$.

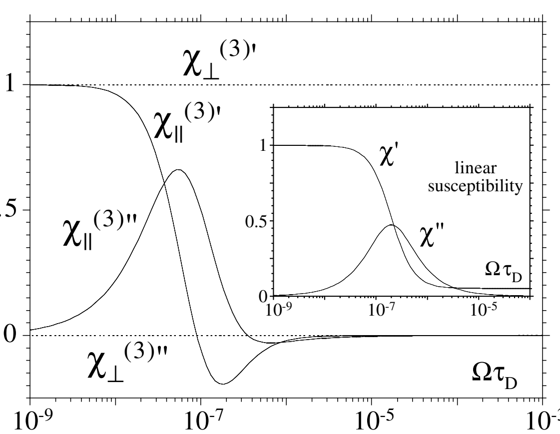

FIG. 1. Longitudinal (solid lines) and transverse (dotted lines) *nonlinear* susceptibility spectra of classical spins with collinear anisotropy axes at the temperature $k_{\mathrm{B}}T/D = 0.05$ ($\sigma = 20$). Inset: *Linear* susceptibility of spins with randomly distributed anisotropy axes. In both cases, results for different values of the damping coefficient λ (1, 0.1, 0.03, and 0.01) are displayed to show their coincidence. The susceptibilities have been divided by their equilibrium values, to single out variations due to dynamical effects.

Let us now turn our attention to the more general situation of a driving field at an oblique angle to the anisotropy axis. Although the linear response to such a field is a linear combination of the longitudinal and transverse susceptibilities, additional terms appear in the nonlinear response. Figure 2 displays $\chi^{(3)}(\Omega)$ in the experimentally most common case of anisotropy axes distributed at random. Note the large dependence of $\chi^{(3)}$ on λ, a dependence that was absent in both the *linear* susceptibility for the same axes distribution (inset of Fig. 1), as well as in the longitudinal and transverse *nonlinear* susceptibilities themselves (Fig. 1). Note also that, in this case, the heights of the susceptibility peaks are by no means only a fraction of the equilibrium values.

In order to interpret physically these results, it is convenient to rewrite the Landau-Lifshitz equation (1) with the time measured in units of τ_{D}, so that the frequency shift of the spectra is scaled out. In this representation, both the relaxation and the fluctuating terms become independent of λ, which enters *only* as a factor $1/\lambda$ multiplying the deterministic precession term. The dependence of $\chi^{(3)}$ on λ can then be interpreted as the result of the time-dependent saddle point created by the oblique driving field in the anisotropy potential barrier. This saddle favors inter-potential-well jumps [15] that would be unlikely if the field were in the linear range (weakly deformed barrier), and, hence, leads to an increase of the magnitude of the response. To illustrate, let us assume that the spin, after a "favorable" sequence of fluctuations, reaches a point P close to the top of the barrier but does not cross it [inset of Fig. 2(b)]. In the subsequent spiralling down back to the bottom of the potential well, a weakly damped spin executes more rotations ($\sim 1/\lambda$) about the anisotropy axis, so that it can approach the saddle area created by $\Delta B_\perp(t)$ (shaded area in that inset), where the probability of over-barrier crossing is larger.

We can see that the precession-assisted barrier crossing picture is consistent with the results obtained: (i) The

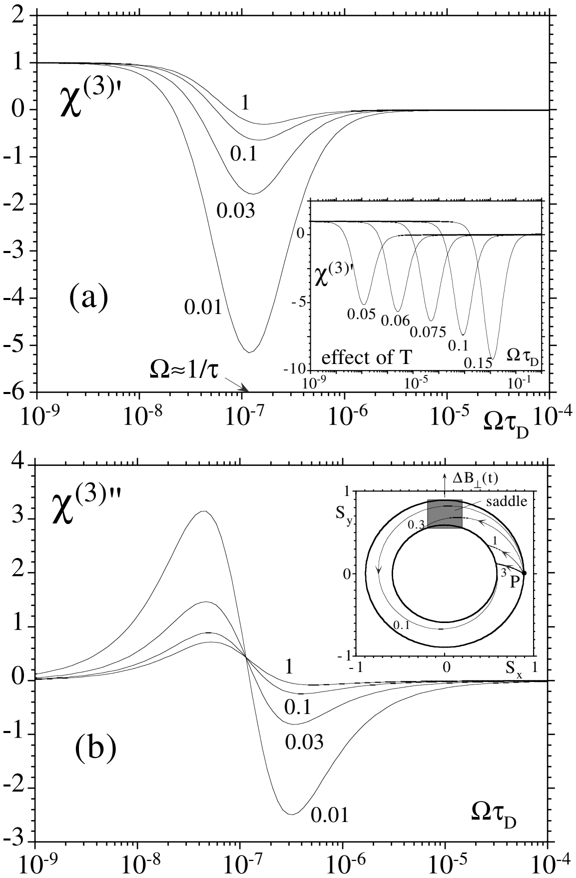

FIG. 2. Nonlinear susceptibility spectra (normalized by the equilibrium susceptibility) of classical spins with randomly distributed anisotropy axes at the temperature $k_B T/D = 0.05$ ($\sigma = 20$), for various values of the damping coefficient λ ranging from the overdamped to the underdamped regime. (a) Real part $\chi^{(3)\prime}$ (inset: results for $\lambda = 0.01$ at various temperatures). (b) Imaginary part $\chi^{(3)\prime\prime}$ (inset: top view of the deterministic decaying trajectories during a time interval τ_D/σ for various λ; shaded area: region with a temporarily lower barrier due to the driving field).

effect is naturally absent if there is no anisotropy barrier to be modified (as it follows from the analytical expression for $\chi^{(3)}$ of isotropic dipoles [2]). (ii) The coupling mechanism is not effective if the barrier is weakly perturbed (independence of the *linear* response on λ; inset of Fig. 1). (iii) The effect is absent if the driving field is either exactly parallel to the anisotropy axis ($\Delta B_\perp = 0$), since the barrier is then modified the same amount everywhere, or exactly perpendicular, since the barrier crossing plays a secondary dynamical role in the transverse projection of the response (dominated by the fast intra-potential-well modes). (iv) The relevance of the mechanism increases with decreasing λ, as the efficiency of the precession to bring the spins close to the saddle is then larger [inset of Fig. 2(b)]. (v) The effect is magnified by the temperature [inset of Fig. 2(a)], as the "energy levels" (orbits) close to the top of the barrier are thermally more populated.

The dependence of $\chi^{(3)}$ on λ can be used to determine λ experimentally, avoiding the methods in which λ is extracted from the preexponential factor τ_0 ($\propto \tau_D$) in the relaxation time τ of uniaxial spins. This is important since (i) τ_0 is typically obtained from the analysis of the position of the peaks of the χ vs T curves, so that what one gets is an average of λ over the temperature range involved; (ii) the errors in the determination of τ_0 are frequently at the level of order of magnitude. In contrast, the dependence of $\chi^{(3)}$ on λ does not rely on that of τ_D, circumventing the above-mentioned problems. In particular, λ can be found at different temperatures, allowing the study of its intrinsic dependence on T.

In summary, we have investigated the effect of the damping on the nonlinear dynamical response of spins governed by the stochastic Landau-Lifshitz equation. It has been found that the coupling of the thermoactivated overbarrier dynamics with the precession of the spin, via the driving field, leads to a large dependence (absent in the linear response) of the nonlinear susceptibility on the damping coefficient. We can confidently conjecture the existence of this effect in more complex systems (interacting spins, other symmetries of the anisotropy, etc.), as long as the essential ingredients—precession, potential barrier, and strong driving field—are present. The dependence found is, moreover, larger in the important medium-to-weak damping regime. Exploiting it, the evasive damping coefficient in superparamagnets could be determined, clarifying the mechanisms coupling the spin to the internal degrees of freedom.

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