Precessional effects in the linear dynamic susceptibility of uniaxial superparamagnets: Dependence of the ac response on the dissipation parameter

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It is shown that the low-frequency relaxation spectrum of the linear dynamic susceptibility of uniaxial single domain particles with a uniform magnetic field applied at an oblique angle to the easy axis can be used to deduce the value of the damping constant.

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A single domain ferromagnetic particle is characterized by an internal potential, having several local states of equilibrium with potential barriers between them. If the particles are small (~ 10 nm) so that the potential barriers are relatively low, the magnetization vector **M** may cross over the barriers due to thermal agitation. The ensuing thermal instability of the magnetization results in the phenomenon of superparamagnetism.[1] This problem is important in information storage, rock magnetism, and the magnetization reversal observed in isolated ferromagnetic nanoparticles.[2] The dynamics of the magnetization **M** of a superparamagnetic particle is usually described by the Landau-Lifshitz or Gilbert (LLG) equation[3,4]

$$2\tau_N \frac{d}{dt}\mathbf{M} = \beta(\alpha^{-1}M_s[\mathbf{M}\times\mathbf{H}] + [[\mathbf{M}\times\mathbf{H}]\times\mathbf{M}]), \quad (1)$$

where

$$\tau_N = \frac{\beta(1+\alpha^2)M_s}{2\gamma\alpha} \quad (2)$$

is the free Brownian motion diffusion time of the magnetic moment, α is the dimensionless damping (dissipation) constant, M_s is the saturation magnetization, γ is the gyromagnetic ratio, $\beta = v/(kT)$, v is the volume of the particle, and the magnetic field **H** consists of applied fields (Zeeman term), the anisotropy field $\mathbf{H}_a$, and a random white-noise field accounting for the thermal fluctuations of the magnetization of an individual particle. Here the internal magnetization of a particle is assumed homogeneous. Surface and ''memory'' effects are also omitted in Eq. (1). These assumptions are discussed elsewhere (e.g., Refs. 5–7). Furthermore, the description of the relaxation processes in the context of Eq. (1) does not take into account effects such as macroscopic quantum tunneling (a mechanism of magnetization reversal suggested in Ref. 1). These effects are important at very low temperatures[8,9] and necessitate an appropriate quantum-mechanical treatment, e.g., Refs. 10–12.

The various regimes of relaxation of **M** in superparamagnetic particles are governed by α. In general, α is difficult to estimate theoretically, although a few experimental methods of measuring α [such as ferromagnetic resonance (FMR) and the angular variation of the switching field, e.g., Refs. 7 and 8] have been proposed. Yet another complementary and potentially promising technique, viz., the *nonlinear* response of single domain particles to alternating (ac) stimuli, has recently[13] been suggested in order to evaluate α. In particular, it has been shown in Ref. 13 that for uniaxial particles having a strong ac field applied at an angle ψ to the easy (Z) axis, the nonlinear response truncated at terms cubic in the ac field is particularly sensitive to the value of α. On the other hand, the *linear* response to the ac field does not exhibit such behavior. The explanation of this is reasonably straightforward: the linear ac response may simply be calculated from the after effect solution following the removal of a weak uniform field applied at an angle ψ to the easy axis. Thus the superparamagnetic (greatest) relaxation time τ is that of a particle with simple uniaxial anisotropy, which is given by Brown's[4] expression

$$\tau \sim \tau_N \frac{\sqrt{\pi}}{2\sigma^{3/2}} e^{\sigma}, \quad \sigma \gg 1, \quad (3)$$

where $\sigma = \beta K$ is the barrier height parameter and K is the anisotropy constant. Equation (3) yields the approximate position of the peak in the imaginary part $\chi''(\omega)$ of the complex susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ in linear response. The most striking feature of Eq. (3) is that τ when normalized by τ_N is independent of α. The physical reason for this is the lack of coupling between the transverse and longitudinal modes in the linear response when a weak ac field alone is applied at an angle ψ to the Z axis. If one proceeds, however,

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to the next term in the response which yields the so-called third-order susceptibility $\chi^{(3)}(\omega)$, then a strong dependence of the imaginary part of $\chi^{(3)}(\omega)$ on α appears.[13] The explanation of this lies in the coupling between the longitudinal and transverse (or processional) modes in the nonlinear response. Thus one can evaluate α from measurements of the nonlinear ac response.

Here unlike Ref. 13, we consider a uniaxial particle in a strong uniform field $\mathbf{H}_0$ applied at an angle ψ to the anisotropy axis of the particle. Hence the system in the absence of the ac perturbation unlike that of [13] is nonaxially symmetric, thus we expect precessional effects due to coupling of the transverse and longitudinal modes to appear even in the linear response to a small ac field $\mathbf{H}(t)$ superimposed on $\mathbf{H}_0$. Indeed, the limiting values of α, viz., $\alpha\to\infty$ and $\alpha\to 0$, correspond to the high-damping and the low-damping limits in the Kramers escape rate theory.[14] The coupling effect is made manifest in the formulas for the Kramers escape rate or inverse of the greatest relaxation time τ of the magnetization for both intermediate to high damping (IHD) and very low damping (VLD) which apply to nonaxially symmetric potentials of the magnetocrystalline anisotropy[5,6] (see below).

Now we recall that the Fokker-Planck equation (FPE) for the probability density distribution W of $\mathbf{M}$ (Ref. 4) corresponding to Eq. (1) is[4,15]

$$2\tau_N\frac{\partial}{\partial t}W=\Delta W+\beta\{\alpha^{-1}\mathbf{u}\cdot[\nabla V\times\nabla W]+\nabla\cdot(W\nabla V)\}, \tag{4}$$

where ∇ and Δ are the gradient and the Laplacian on the surface of the unit sphere, respectively, $\mathbf{u}$ is the unit vector directed along $\mathbf{M}$, and $V(\mathbf{M})$ is the free-energy density. Here, in the absence of the ac field, V is given by

$$\beta V=-\sigma[\cos^2\vartheta+2h(\sin\psi\cos\varphi\sin\vartheta+\cos\psi\cos\vartheta)], \tag{5}$$

where ϑ and φ are the polar and azimuthal angles, respectively, and $h=M_sH_0/(2K)$ is the dimensionless external field parameter. The free energy in Eq. (5) has a bistable structure with minima at $\mathbf{n}_1$ and $\mathbf{n}_2$ separated by a potential barrier containing a saddle point at $\mathbf{n}_0$.[15] If $(\alpha_1^{(i)},\alpha_2^{(i)},\alpha_3^{(i)})$ denote the direction cosines of $\mathbf{M}$ and $\mathbf{M}$ is close to a stationary point $\mathbf{n}_i$ of the free energy, then $V(\mathbf{M})$ can be approximated to second order in $\alpha^{(i)}$ as[4]

$$V=V_i+\frac{1}{2}[c_1^{(i)}(\alpha_1^{(i)})^2+c_2^{(i)}(\alpha_2^{(i)})^2]. \tag{6}$$

Substituting Eq. (6) into Eq. (5), the FPE may be solved near the saddle point yielding [4,15]

$$\tau=\tau_{\mathrm{IHD}}\sim\left\{\frac{\Omega_0}{2\pi\omega_0}[\omega_1e^{\beta(V_1-V_0)}+\omega_2e^{\beta(V_2-V_0)}]\right\}^{-1}, \tag{7}$$

where $\omega_i^2=\gamma^2M_s^{-2}c_1^{(i)}c_2^{(i)}(i=1,2)$ and $\omega_0^2=-\gamma^2M_s^{-2}c_1^{(0)}c_2^{(0)}$ are the squares of the well and saddle angular frequencies, respectively, and

$$\Omega_0=\frac{\beta}{4\tau_N}[-c_1^{(0)}-c_2^{(0)}+\sqrt{(c_2^{(0)}-c_1^{(0)})^2-4\alpha^{-2}c_1^{(0)}c_2^{(0)}}].$$

Equations for $c_i^{(j)}$ and V_i are given elsewhere.[15] Equation (7) is similar to the IHD formula derived by Kramers[14] and applies when the energy loss per cycle at the saddle point energy of the motion of the magnetic moment $\Delta E\gg kT$. If $\Delta E\ll kT$(VLD), we have for the escape from a single well[5,16]

$$\tau=\tau_{\mathrm{LD}}\sim\frac{\pi kT}{\omega_1\Delta E}e^{\beta(V_0-V_1)}. \tag{8}$$

[Here instead of numerical evaluation of ΔE, we have used an approximation $\Delta E\approx\alpha v|V_0|$ (Ref. 5)]. The IHD and VLD limits correspond to $\alpha\geq 1$ and $\alpha\leq 0.01$, respectively. However, for crossover values of α (about $\alpha\approx 0.1$) neither the IHD formula (7) nor the VLD, Eq. (8), can yield reliable quantitative estimates. Thus a more detailed analysis is necessary.[17]

Equations (7) and (8) applied to the potential given by Eq. (5) yield the greatest relaxation time τ in the appropriate limits (IHD, VLD) for a strong uniform field $\mathbf{H}_0$ applied at an angle ψ to the Z axis [8,18]; τ is effectively identical to the integral relaxation time (in linear response, the correlation time)[19] if the strength of $\mathbf{H}_0$ is smaller than the reduced critical field h_c at which depletion of the shallower of the two potential wells of the bistable potential occurs (for example, $h_c\approx 0.17$ in the axially symmetrical case[19]). As shown in Refs. 8 and [17], the asymptotes (7) and (8) are in excellent agreement with the exact numerical results from the FPE (4). Equations (7) and (8) can also successfully reproduce the experimental angular variation of the switching field for individual Co and BaFeCoTiO particles and thus allows one to evaluate α.[8]

Equations (7) and (8) for τ, which now exhibit strong α dependence, suggest that the frictional dependence of the relaxation process may also be observed and used for the evaluation of α in the linear ac response of the system. In order to verify our conjectures concerning the α dependence of the linear response to a small ac field $\mathbf{H}(t)$ (i.e., assuming $\beta M_sH\ll 1$), we have calculated using linear-response theory the complex magnetic susceptibility $\chi(\omega)$ of the system. The susceptibility was calculated by using a matrix continued-fraction solution[20,21] of the system of moments [the expectation values of the spherical harmonics $\langle Y_{l,m}\rangle(t)$] governing the kinetics of the magnetization $\mathbf{M}$ (the moment system can be obtained either from the FPE or from the LLG equation[22]). The details of the calculation can be found elsewhere[20,21]; it is assumed that $\mathbf{H}(t)$ is directed along $\mathbf{H}_0$. The plots of $\mathrm{Re}\{\chi(\omega)\}$ and $\log_{10}[-\mathrm{Im}\{\chi(\omega)\}]$ vs $\log_{10}(\omega\tau_N)$ are shown in Figs. 1–3 for a wide range of frequency, bias field strength, and damping (the calculations were carried out for $v\beta M_s^2N_0=1$; N_0 is the number of particles per unit volume). The results indicate that a marked dependence of $\chi(\omega)$ on α exists and that three distinct dispersion bands appear in the spectrum. Furthermore, the characteristic frequency and the half-width of the low-frequency relaxation band (LRB) are determined by the characteristic frequency $\omega_{ob}\sim\tau^{-1}$ of

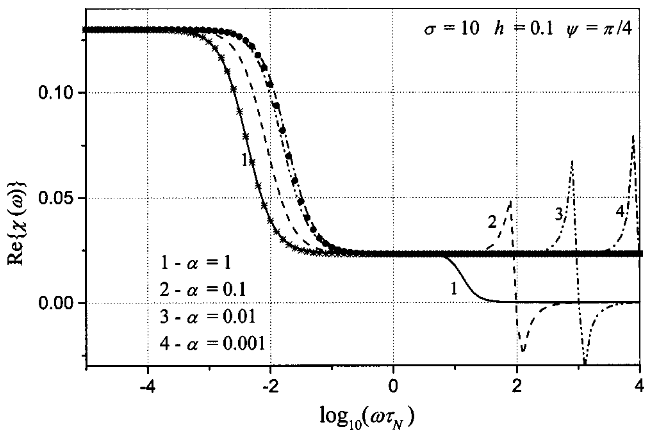

FIG. 1. Re$\{\chi(\omega)\}$ vs $\log_{10}(\omega\tau_N)$ from the IHD ($\alpha=1$) to the VLD ($\alpha=0.001$) limits for $\sigma=10$, $h=0.1$, and $\psi=\pi/4$. Curves 1–4: exact numerical calculations of $\chi(\omega)$ based on the results of Refs. 20 and 21. Stars and filled circles: Eq. (9) with $\Delta\chi_{\rm hf}\approx 0.023$ and τ from Eqs. (7) and (8), respectively.

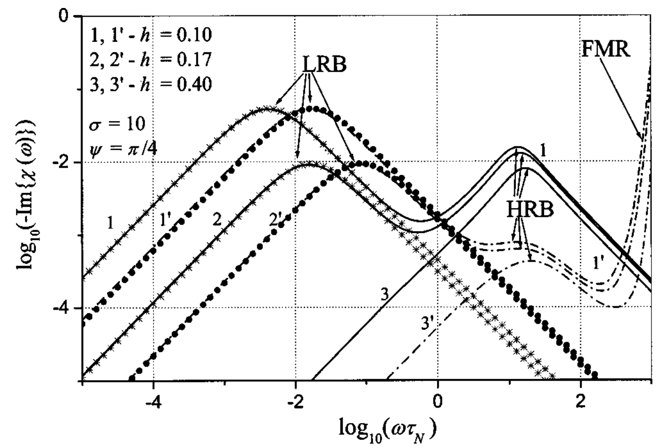

FIG. 3. $\log_{10}[-\text{Im}\{\chi(\omega)\}]$ vs $\log_{10}(\omega\tau_N)$ for $\sigma=10$, $\psi=\pi/4$, $\alpha=1.0$ (IHD: solid lines 1, 2, and 3), and $\alpha=0.01$ (low damping: dashed-dotted lines $1'$, $2'$, and $3'$). Lines 1, $1'(h=0.01)$, 2, $2'(h=0.17)$, and 3, $3'(h=0.4)$ are exact numerical calculations. Stars and filled circles: Eqs. (9) with τ from Eqs. (7) and (8), respectively.

the overbarrier relaxation mode. As α decreases, this peak shifts to higher frequencies and reaches its limiting value τ_{LD}^{-1}. In addition, a far weaker second relaxation peak appears at high frequencies (HF). This HF relaxation band (HRB) is due to the intrawell modes [for $\psi=0$ and $\sigma\gg 1$, the characteristic frequency of this relaxation peak is $\omega_{\rm well}\approx 2\sigma(1+h)/\tau_N$ (Ref. 19)]. The third FMR peak due to the excitation of transverse modes having frequencies close to the precession frequency $\omega_{\rm pr}$ of the magnetization appears only at low damping and strongly manifests itself at HF. As α decreases, the FMR peak shifts to higher frequencies since $\omega_{\rm pr}\sim\alpha^{-1}$. Moreover, at $\psi=0$ or $\psi=\pi$, the FMR peak disappears because the transverse modes no longer take part in the relaxation process. The dependence of the linear response on the bias-field strength is demonstrated in Fig. 3. Here, the effect of the depletion[19,23] of the shallower of the two potential wells of a bistable potential (5) by a bias field is apparent: at fields above the critical field h_c at which the depletion occurs, it is possible to make the LF peak disappear (curves 3 and $3'$). Such behavior of $\chi(\omega)$ implies that if one is interested solely in the low-frequency ($\omega\tau\lesssim 1$) part of $\chi(\omega)$, where the effect of the HF modes may be completely ignored (so that the relaxation of the magnetization at long times may be approximated by a single exponential with the characteristic time τ), then the Debye-like relaxation formula, viz.,

$$\chi(\omega)=\frac{\chi_{\rm st}-\Delta\chi_{\rm hf}}{1+i\omega\tau}+\Delta\chi_{\rm hf}, \tag{9}$$

yields an accurate description of the LF spectra (see Figs. 1–3). Here τ is given by Eqs. (7) and (8) in the IHD and VLD limits, respectively, $\chi_{\rm st}=\chi(0)$ is the static susceptibility, and $\Delta\chi_{\rm hf}$ is the contribution of the HF transverse and longitudinal modes. The values of $\chi_{\rm st}$ and $\Delta\chi_{\rm hf}$ depend on ξ, ψ, and σ and can be measured experimentally, calculated numerically, and/or estimated theoretically (an example of such theoretical estimations of $\chi_{\rm st}$ and $\Delta\chi_{\rm hf}$ for $\psi=0$ has been given by Garanin [19]). Our calculations indicate that Eqs. (7)–(9) yield an adequate description of the LF spectra for $\sigma\geqslant 3$.

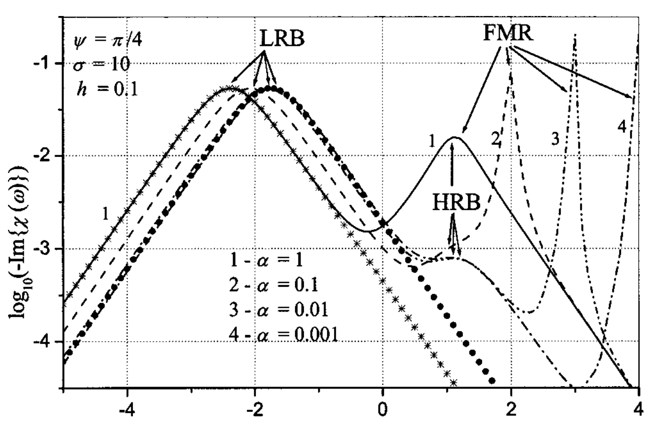

FIG. 2. The same as in Fig. 1 but for $\log_{10}[-\text{Im}\{\chi(\omega)\}]$.

We have demonstrated that it is unnecessary to resort to the nonlinear response in order to observe large precessional effects in the relaxation processes of uniaxial superparamagnets. All that is required is to superimpose a strong bias field $\mathbf{H}_0$ at an angle ψ to the easy axis of the uniaxial particle, thus ensuring that the system is nonaxially symmetric, and then to calculate the linear response to a perturbing ac field $\mathbf{H}(t)$. It follows that the nonaxial symmetry causes the various damping regimes (IHD and VLD) of the Kramers problem to appear unlike in an axially symmetric potential, where the formula for τ [for example, Eq. (2)] is valid for all α because τ/τ_N is independent of α. We remark that the intrinsic α dependence of $\chi(\omega)$ for the oblique field configuration serves as a signature of the coupling between the longitudinal and precessional modes of the magnetization. Hence, it should be possible to determine the evasive damping coefficient from measurements of the *linear* response, e.g., by fitting the theory to the experimental LF dependence of $\chi(\omega)$ on the angle ψ and the bias strength H_0, so that the sole fitting parameter is α. Just as in the nonlinear response,[13] α can be determined at different T, yielding its temperature dependence. This is of importance because of its implications in the search for other mechanisms of magnetization reversal of **M** (e.g., macroscopic quantum tunneling [9,27]), as a knowledge of α and its T dependence allows the separation of the various relaxation mechanisms. Moreover, such experiments are much more easily accomplished than those for the nonlinear response of Ref. 13. The results we have obtained suggest that the experimental measurements of linear and nonlinear susceptibility of fine particles (e.g., Refs. 24–26) should be repeated for a strong bias-field configuration.

The results we have presented pertain to noninteracting superparamagnetic particles with easy axes oriented along the Z axis of the laboratory system of coordinates. If the easy axes are randomly distributed in space, further averaging must be carried out in order to calculate $\chi(\omega)$. In the calculations, we have also assumed that all the particles are identical; in order to account for polydispersity, one must average $\chi(\omega)$ over the appropriate distribution function (e.g., over the particle volumes; see for details Refs. 25, 26, and 28). Furthermore, the neglect of interparticle interactions in the present model suggests that the results we have obtained are applicable for systems where the effects of the dipole-dipole and exchange interactions may be ignored, such as individual nanoparticles (e.g., Refs. 2 and 8) and diluted solid suspensions of nanoparticles (e.g., Ref. 26).

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