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Effect of the Damping on the Magnetization of Antiferromagnetic Nanoparticles in a Presence of an Oblique Magnetic Field

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ABSTRACT

The magnetization of antiferromagnetic nanoparticles is investigated with the Fokker-Planck equation describing the evolution of the distribution function of the magnetization of an nanoparticle. By solving this equation numerically, the relaxation times, and dynamic susceptibility are calculated for dc field orientations across wide ranges of frequencies, amplitude of the fields and damping. Analytic equation for the dynamic susceptibility is also proposed. It is shown that the damping alters the magnetization in the presence of oblique field applied.

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Introduction

Ferromagnetic particles are categorized by thermal instability of their magnetization causing in natural change in their orientation from one metastable state to another by overcoming energy barriers, giving rise to superparamagnetism [1-3]. The thermal instability of magnetization in antiferromagnetic nanoparticles, may differ from that of ferromagnetic nanoparticles due to the intrinsic properties of antiferromagnetic materials [4, 5]. The theory of antiferromagnetic nanoparticles was advanced by Néel, who established that total magnetic compensation of the sublattices in antiferromagnetic nanoparticles is not possible for a number of reasons, namely, unequal numbers of spins in crystal planes, spin frustration near the surface, lattice defects, etc [6, 7]. Hence, an equilibrium magnetization should ensue in such particles, moreover, they should become superparamagnetic at a finite temperature just as ferromagnetic nanoparticles. According to Néel, the so-called superantiferromagnetism arises in a nanoparticle with an even number of sublattice planes, causing an appreciable increase in transverse susceptibility in comparison to that of a massive sample [6, 7]. An understanding the dynamics of the magnetization of the antiferromagnetic particles is essential owing to their role played in various areas of science and technology such as spintronics, biomedical applications, catalysis, etc [4].

The treatment of the fluctuations of the magnetization of nanoparticles due to Néel based on classical transition state theory was further developed by Brown and is therefore known as the Néel-Brown theory [1, 8, 9]. This tools utilizes the classical theory of Brownian motion with the Landau-Lifshitz-Gilbert equation augmented by white noise fields as Langevin equation governing the stochastic magnetization dynamics [10]. This equation is then used to obtain the Fokker-Planck equation describing the time evolution of the probability density function of magnetization orientations. At temperatures much lower than TN, this theory may be adapted to antiferromagnetic nanoparticles as has been suggested by Raikher and Stepanov in connection with the low-frequency magnetodynamics of antiferromagnetic nanoparticles suspended in a fluid by means of a kinetic model for the magnetization relaxation in the high magnetic anisotropy limit [4]. For an antiferromagnetic particle subjected to a dc magnetic field H, the magnetic moments of the sublattices and are given by [4].

$$\mathbf{m}_{1,2} = \mathbf{u}\left[vM_S \pm \mu/2 - v\chi_A(\mathbf{u}\cdot\mathbf{H})/2\right],$$

where MS is the sublattice magnetization in a bulk, is a parameter characterizing the induced magnetic moment of the particle, is the unit vector along the decompensation magnetic moment , and v is the particle volume. The free energy of the particle subjected to a dc magnetic field H applied at an angle to the easy axis is

$$U(\vartheta,\varphi) = \beta^{-1}\sigma\left[\sin^2\vartheta - 2h(\gamma_1\sin\vartheta\cos\varphi + \gamma_2\sin\vartheta\sin\varphi + \gamma_3\cos\vartheta)\right. \quad (1)$$
$$\left. + 2\sigma h^2\zeta(\gamma_1\sin\vartheta\cos\varphi + \gamma_2\sin\vartheta\sin\varphi + \gamma_3\cos\vartheta)^2\right],$$

where ϑ and φ are the polar and azimuthal angles of the spherical coordinate system, $\beta = (kT)^{-1}$, k is Boltzmann's constant, T is the temperature in Kelvin, $\sigma = v\beta K$ is the barrier, K is the anisotropy constant, $h = \mu H/(2vK)$ is the applied field parameter, $\zeta = v\chi_A/\beta\mu^2$ is the "antiferromagnetic" parameter, and γ_1, γ_2, γ_3 are the direction cosines of the vector **H**. As long as the magnetic field **H** is much weaker than the exchange field, the only possible motion of the vector **μ** is rotation which may be treated using the Brown model [8, 9]. Thus the magnetization dynamics are governed by a Fokker-Planck equation for the probability density function $W(\boldsymbol{\mu}, t)$ of **μ**, viz.,

$$2\tau_N \frac{\partial}{\partial t} W = \mathrm{L}_{FP} W \qquad (2)$$

where L_{FP} is the Fokker-Planck operator given in ref, τ_N is the free diffusion time of the magnetization, and α is the dimensionless damping parameter [8].

When a dc magnetic field parallel to the easy axis of the particle, i.e., $\gamma_1 = \gamma_2 = 0$, $\gamma_3 = 1$, the free energy Eq. (1) is independent of the azimuthal angle φ. Due to axial symmetry, no dynamical coupling between the longitudinal and the transverse modes of motion exists so that for the longitudinal relaxation Eq. (2) reduces to a single-variable Fokker-Planck equation for the distribution function $W(\vartheta, t)$ namely [4, 11],

$$\frac{\partial}{\partial t} W = \frac{1}{2\tau_N \sin\vartheta} \frac{\partial}{\partial \vartheta} \left[\sin\vartheta \left(\frac{\partial W}{\partial \vartheta} + \beta W \frac{\partial V}{\partial \vartheta} \right) \right]. \qquad (3)$$

This axially symmetric case has been considered recently by Raikher and Stepanov; by solving Eqs. (2) and (3), they have calculated numerically the longitudinal and transverse dynamic susceptibilities and corresponding integral relaxation times [4].

The goal of the present paper is to study the relaxation of the magnetization of antiferromagnetic nanoparticles subjected to a Oblique dc magnetic field applied. We present results of calculations of the longitudinal complex magnetic susceptibility $\chi(\omega)$ of a antiferromagnetic particle and characteristic relaxation times of the magnetization in broad temperature and damping ranges. In particular, we calculate the reversal, effective, and integral relaxation times which characterize, respectively, the long, short, and overall behavior of the magnetization. The calculations are mainly accomplished by using the matrix continued fractions, however, simple analytic equations for the low and high-frequency parts of the spectrum $\chi(\omega)$ and relaxation times are also obtained [10]. In the low temperature limit, our numerical calculation for the reversal time of the magnetization τ agrees with analytic estimates of Ouari et al [11]. who have evaluated τ of antiferromagnetic nanoparticles by adapting the Kramers escape rate theory to fine ferromagnetic particles given by Coffey et al [12-14]. We remark in passing that in the limiting case ζ=0, the free energy from Eq. (1) reduces to that of uniaxial superparamagnets; this case has been treated in Refs [10,15-19].

Basic Equations

A Strong theoretical description of the relaxation of the magnetization in antiferromagnetic nanoparticles can be given by linear response theory. Here it is supposed that a particle in the presence of a strong uniform magnetic field H is subjected in addition to a small probe field $\mathbf{H}_1$ [$\beta(\boldsymbol{\mu}\cdot\mathbf{H}_1) \ll 1$] parallel to $\mathbf{H}$. Then the decay of the longitudinal component of the averaged magnetization $\langle M_\parallel\rangle(t) = v^{-1}\langle\mu_\parallel\rangle(t)$ of the particle, when the field $\mathbf{H}_1$ has been switched off at time $t = 0$, is [10]

$$\left\langle M_\square \right\rangle(t) - \left\langle M_\square \right\rangle_0 = \chi C_\square(t) H_1, \qquad (4)$$

where $C_\square(t)$ is the normalized relaxation (correlation) function of the longitudinal component of the magnetization defined as

$$C_\square(t) = \frac{\left\langle M_\square(0) M_\square(t) \right\rangle_0 - \left\langle M_\square(0) \right\rangle_0^2}{\left\langle M_\square^2(0) \right\rangle_0 - \left\langle M_\square(0) \right\rangle_0^2} = \sum_{k=1}^{k=\infty} c_k e^{-\lambda_k t}, \qquad (5)$$

λ_k are the eigenvalues of the Fokker-Planck operator LFP in Eq. (2), $\sum_{k=1}^{k=\infty} c_k = 1$, $\chi = v\beta\left[\left\langle M_\square^2(0)\right\rangle_0 - \left\langle M_\square(0)\right\rangle_0^2\right]$, the static susceptibility of the particle, and the brackets $\langle\ \rangle$ and $\langle\ \rangle_0$ denote the nonequilibrium and equilibrium ensemble averages, respectively. The equilibrium ensemble averages are defined as

$\langle A\rangle_0 = Z^{-1}\int_0^{2\pi}\int_0^{\pi} A(\vartheta,\varphi) e^{-\beta V(\vartheta,\varphi)} \sin\vartheta d\vartheta d\varphi$ (Z is the partition function). Having determined, $C_\square(t)$ one can calculate the longitudinal dynamic susceptibility of the particle $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ given by [10].

$$\frac{\chi(\omega)}{\chi} = 1 - i\omega\int_0^{\infty} e^{-i\omega t} C_\square(t) dt = \sum_{k=1}^{k=\infty} \frac{c_k}{1 + i\omega/\lambda_k} \qquad (6)$$

The dynamic susceptibility characterize the response of the particle to a weak ac probe field $H_1(t) = H_1 \mathrm{Cos}\ \omega t$ viz.,

$$\left\langle M_\square \right\rangle(t) - \left\langle M_\square \right\rangle_0 = H_1\left[\chi'(\omega)\cos\omega t + \chi''(\omega)\sin\omega t\right].$$

Both $\chi'(\omega)$ and $\chi''(\omega)$ can be measured experimentally.

The asymptotic behavior of $\chi(\omega)$ in the extremes of very low and very high frequencies is

$$\frac{\chi(\omega)}{\chi} \sim \begin{cases} 1 - i\omega\tau_{cor} + \ldots, & \omega \to 0, \\ -i\left(\omega\tau_{ef}\right)^{-1} + \ldots, & \omega \to \infty. \end{cases}, \qquad (7)$$

where

$$\tau_{cor} = \sum_{k=1}^{k=\infty} c_k / \lambda_k \ \text{ and } \ \tau_{ef} = \left(\sum_{k=1}^{k=\infty} c_k \lambda_k\right)^{-1} \qquad (8)$$

We remark that the relaxation times so defined τ_{cor} and τ_{ef} parameterize the time behavior of $C(t)$. The integral relaxation time τ_{cor} which can be also defined as the area under $C_\square(t)$, viz., [10]

$$\tau_{cor} = \int_0^{\infty} C_\square(t) dt, \qquad (9)$$

characterizes the overall behavior of $C(t)$ while the effective relaxation time τ_{ef} yields precise information on the initial decay of $C(t)$, namely,

$$\tau_{ef} = -1/\dot{C}_\square(0). \qquad (10)$$

The relaxation times τ_{cor} and τ_{ef} contain contributions from all the eigenvalues λ_k of the Fokker-Planck operator LFP. The smallest nonvanishing eigenvalue λ_1 is associated with the slowest interwell (or overbarrier) relaxation mode and so with the reversal time of the magnetization $\tau = 1/\lambda_1$; the other eigenvalues λ_k characterize high-frequency "intrawell" modes. The dependences of the effective relaxation time τ_{ef} on the model parameters (external field, anisotropy constants) may differ considerably from that of τ_{cor} and τ as τ_{ef} is not governed by λ_1. The effective relaxation time τ_{ef} can also be expressed in terms of equilibrium averages as [10].

$$\tau_{ef} = 2\tau_N \frac{\left\langle u_\square^2 \right\rangle_0 - \left\langle u_\square \right\rangle_0^2}{1 - \left\langle u_\square^2 \right\rangle_0}, \qquad (11)$$

where $\mu = \cos\psi\cos\vartheta + \sin\psi\sin\vartheta\cos\varphi$. Here and below without loss of generality it is supposed that the field $\mathbf{H}$ is in the xz plane

so that the direction cosines in Eq. (1) are $\gamma_1 = \sin\psi$, $\gamma_2 = 0$, and $\gamma_3 = \cos\psi$, where ψ is the angle between $\boldsymbol{H}$ and the Z axis is taken as the easy axis of the particle.

The reversal and integral relaxation times can be used to evaluate the low-frequency dynamics of the magnetization using a single mode approximation [10]. According to this approximation, the dynamic susceptibility $\chi(\omega)$ given as an infinite series of Lorenzians, Eq. (6), may be approximated at low frequencies by a single Lorentzian [10].

$$\frac{\chi(\omega)}{\chi} \approx 1 - \frac{i\omega\tau_{cor}}{1+i\omega\tau} \qquad (12)$$

guaranteeing the correct asymptotic behavior of $\chi(\omega)$ at low frequencies $\omega\tau \leq 1$ [cf. Eq. (7)].

Calculation of the Observables

By applying the method of solution of the Fokker-Planck equation (2) developed in Ref., one can obtain 25 terms differential-recurrence equation for the relaxation functions governing the $c_{l,m}(t) = \langle Y_{l,m}\rangle(t) - \langle Y_{l,m}\rangle_0$ dynamics of the magnetization, viz [20].,

$$\frac{d}{dt}c_{l,m}(t) = \sum_{r,s=-2}^{2} d_{l,m,l+r,m+s}\, c_{l+r,m+s}(t), \qquad (13)$$

where $Y_{l,m}(\vartheta, \varphi)$ are the spherical harmonics and $d_{l,m,l',m'}$ are the matrix elements of the Fokker-Planck operator in Eq. (2). Details of the derivation of Eq. (13) for an arbitrary free energy $V(\vartheta, \varphi)$ are given in Refs. 10 and 20. The $d_{l,m,l',m'}$ for $V(\vartheta, \varphi)$ from Eq. (1) are listed in Appendix A.

Equation (13) can be solved exactly for the one-sided Fourier transforms $\tilde{c}_{l,m}(\omega) = \int_0^\infty c_{l,m}(t)e^{-i\omega t}dt$ by matrix continued fractions (see Appendix A). Having determined $\tilde{c}_{l,m}(\omega)$, we can evaluate the spectrum $\tilde{C}_\parallel(\omega) = \int_0^\infty e^{-i\omega t}C_\parallel(t)dt$ of the longitudinal relaxation function $C_\parallel(t)$ as

$$\tilde{C}_\parallel(\omega) = \frac{\sqrt{2}\gamma_3\tilde{c}_{1,0}(\omega) - (\gamma_1 - i\gamma_2)\tilde{c}_{1,1}(\omega) + (\gamma_1 + i\gamma_2)\tilde{c}_{1,-1}(\omega)}{\sqrt{2}\gamma_3 c_{1,0}(0) - (\gamma_1 - i\gamma_2)c_{1,1}(0) + (\gamma_1 + i\gamma_2)c_{1,-1}(0)} \qquad (14)$$

as well as the dynamic susceptibility $\chi(\omega)$ from Eq. (6), where the static magnetic susceptibility χ is given by

$$\chi = (\mu/H_1)\sqrt{4\pi/3}\left\{\gamma_3 c_{1,0}(0) - \sqrt{2}\,\mathrm{Re}\left[(\gamma_1 - i\gamma_2)c_{1,1}(0)\right]\right\}.$$

Moreover, by using matrix continued fractions, we can also evaluate the integral relaxation time

$$\tau_{cor} = \tilde{C}_\parallel(0) \qquad (15)$$

and the smallest nonvanishing eigenvalue λ_1 of the Fokker-Planck operator and consequently the reversal time $\tau = 1/\lambda_1$ (see Appendix A).

Now the smallest nonvanishing eigenvalue λ_1 characterizes the slowest overbarrier relaxation mode and, hence, the long-time behavior of the magnetization. In order to find a low temperature (high barriers) asymptotic estimate for λ_1 of the Fokker-Planck operator L_{FP} in Eq. (2), Brown and Smith and De Rozario adapted to magnetic relaxation an ingenious method originally proposed by Kramers for thermally activated escape of point Brownian particles from a potential well [9, 21, 12, 13]. Thus they estimated the superparamagnetic relaxation time in the so-called intermediate-to-high damping (IHD) limit ($\alpha \geq 1$). Later, Klik and Gunther derived the corresponding formula for τ in the very low damping limit ($\alpha \geq 1$) [22, 23]. Finally, Coffey et al. have obtained the asymptotic formula for τ which is valid all values of damping [14]. The results of Coffey et al. agree closely with numerical solutions of the Fokker-Planck Eq. (2) and Langevin dynamics simulations of the magnetization reversal time for a variety of magnetocrystalline anisotropy potentials (cubic, biaxial, etc.); they also have been successfully compared with experiments on the angular variation of the switching field for individual Co and BaFeCoTiO particles [14, 18, [24-28]]. For antiferromagnetic nanoparticles with the free energy, Eq. (1), the reversal time of the magnetization τ have been estimated analytically by Ouari et al. [11] using the approach of Coffey et al. [14] as

$$\tau \sim \tau_{IHD}\frac{A(\alpha S_1 + \alpha S_2)}{A(\alpha S_1)A(\alpha S_2)}, \qquad (16)$$

where τ_{IHD} is the reversal time in the IHD limit, $\alpha \geq 1$, $S_{1,2}$ are the dimensionless actions, and A (δ) is the so called depopulation factor (equations for τ_{IHD}, A, and $S_{1,2}$ are given in Ref.) [14, 11].

For the axially symmetric case, $\gamma_1 = \gamma_2 = 0$, $\gamma_3 = 1$, all equations for the relaxation times can be simplified. So using the mean first passage method, Ouari et al. have derived from Eq. (3) the analytic equation for the reversal time in the low temperature limit, viz [11],

$$\tau \sim \frac{\tau_N\sqrt{\pi}e^{\sigma'(1-\xi/2\sigma')^2}}{\sigma'^{3/2}\left[1-(\xi/2\sigma')^2\right]\left[1-\xi/2\sigma'+(1+\xi/2\sigma')e^{-2\xi}\right]}. \qquad (17)$$

where $\sigma' = \sigma - \xi^2\zeta/2$ is an effective anisotropy constant. For axial symmetry, Eq. (16) is no longer valid. If the departures from axial symmetry are small, the nonaxially symmetric asymptotic Eq. (16) for the reversal time may be smoothly connected to the axially symmetric results Eq. (17) by means of suitable bridging integrals [14]. Yet another method of treatment of the uniaxial-nonuniaxial crossover, which does not need bridging integrals, was proposed by Usov [29]. Now, since the dynamics of the system are governed by a single variable ϑ, the integral relaxation time $\tau_{cor} = \tilde{C}_\parallel(0)$ can also be calculated from the analytic equation as (see, Ref. 10, Chap. 2, Sec. 2.10 for details)

$$\tau_{cor} = \frac{2\tau_N}{Z\left(\langle\cos^2\vartheta\rangle_0 - \langle\cos\vartheta\rangle_0^2\right)}\int_{-1}^{1}\left[\int_{-1}^{z}(z' - \langle\cos\vartheta\rangle_0)e^{-\beta V(z')}dz'\right]^2\frac{e^{\beta V(z)}}{1-z^2}dz, \qquad (18)$$

where $\beta V(z) = -\sigma' z^2 - \xi z$, $z = \cos\vartheta$, $\xi = \beta\mu H$ is the external field parameter,

$$\langle\cos\vartheta\rangle_0 = \frac{1}{Z}\int_{-1}^{1}xe^{-\beta V(x)}dx = \frac{e^{\sigma'}\sinh(2\sigma' h')}{\sigma' Z} - h', \qquad (19)$$

$$\langle\cos^2\vartheta\rangle_0 = \frac{1}{Z}\int_{-1}^{1}x^2e^{-\beta V(x)}dx = \frac{e^{\sigma'}[\cosh(2\sigma' h') - h\sinh(2\sigma' h')]}{\sigma' Z} + h'^2 - \frac{1}{2\sigma'}, \qquad (20)$$

$$Z=\int_{-1}^{1}e^{-\beta V(z)}dz=\frac{1}{2}\sqrt{\frac{\pi}{\sigma'}}e^{-\sigma' h'^2}\left\{\mathrm{erfi}[(1+h')\sqrt{\sigma'}]+\mathrm{erfi}[(1-h')\sqrt{\sigma'}]\right\},$$

is the partition function, $h'=\xi/(2\sigma')$, and $\mathrm{erfi}(z)=(2/\sqrt{\pi})\int_0^z e^{t^2}dt$ is the error function of imaginary argument. Finally, the effective relaxation time τ_{ef} from Eq. (11)is given by

$$\tau_{ef}=2\tau_N\frac{\langle\cos^2\vartheta\rangle_0-\langle\cos^2\vartheta\rangle_0^2}{1-\langle\cos^2\vartheta\rangle_0}, \qquad (21)$$

where $\langle\cos\vartheta\rangle_0$ and $\langle\cos^2\vartheta\rangle_0$ are defined by Eqs. (19) and (20), respectively.

Results and Discussion

The inverse of the smallest nonvanishing eigenvalue of the Fokker Planck equation λ_1^{-1}, the integral relaxation time τ_{cor} (both calculated with the matrix continued fraction method), and the reversal time τ predicted by Eq. (16) as functions of the anisotropy (or the inverse temperature) parameter σ are shown in Fig. 1 for various values of the external field parameter parameters h. As is apparent from Fig. 1, with increasing h, the integral relaxation time τ_{cor} may have a behavior dramatically different from that ofλ_1^{-1} above certain critical values of the parameters h_c. In particular, if the dc field parameter h exceeds h_c nevertheless well below that destroying the bistable potential structure of the potential, then τ may differ exponentially from τ_{cor}due to the so-called depletion effect [30]. This effect is qualitatively similar to that for ferromagnetic nanoparticles [10, 30]. The integral relaxation time τ_{cor} as functions of the antiferromagnetic parameter ζ calculated numerically by the matrix continued fraction method are shown in Fig. 2 for various values of the oblique angle ψ. This figure demonstrates that the variations in the antiferromagnetic parameter ς have a very pronounced effect on the relaxation process.

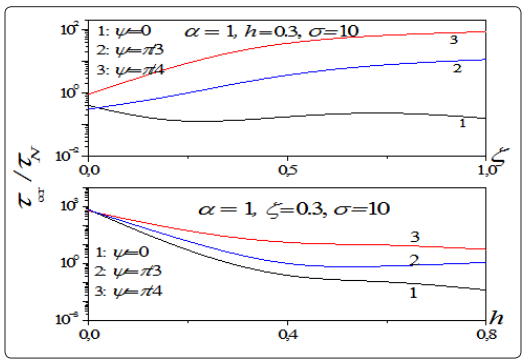

Figure 1

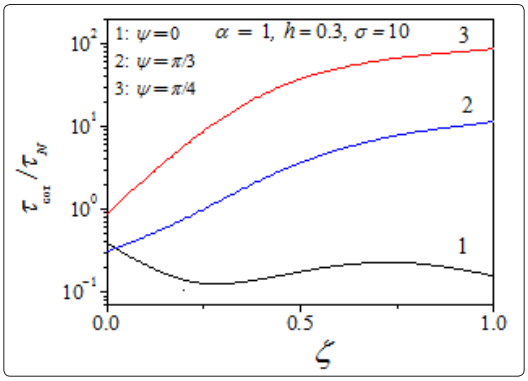

Figure 2

Figures 3 and 4 illustrate the results of the calculation of the imaginary part of the susceptibility $\chi''(\omega)=\chi\omega\,\mathrm{Re}[\tilde{G}(\omega)]$ for α=1 (moderate damping) and various values of the model parameters σ, ψ and h, using the matrix continued fraction solution and the approximate Eq. (12). These figures indicate that at α ≥1 only two distinct dispersion bands appear in the spectrum of $\chi''(\omega)$. The low frequency relaxation band of $\chi''(\omega)$ is dominated by the barrier crossing mode so that the characteristic frequency ω_1 and half-width $\Delta\omega_1$ of this band are completely determined by the smallest nonvanishing eigenvalue. In addition, a far weaker second relaxation band appears at high frequencies. This relaxation band is due to the individual near degenerate high-frequency "intrawell" modes corresponding to the eigenvalues $\lambda_k >> \lambda_1$. At low fields, the amplitude of this band is far weaker than that of the first band. However, in a strong magnetic field, this band can dominate in the spectrum $\chi''(\omega)$ (Fig. 4a).

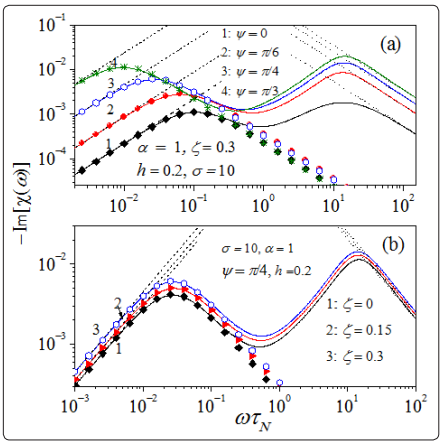

Figure 3

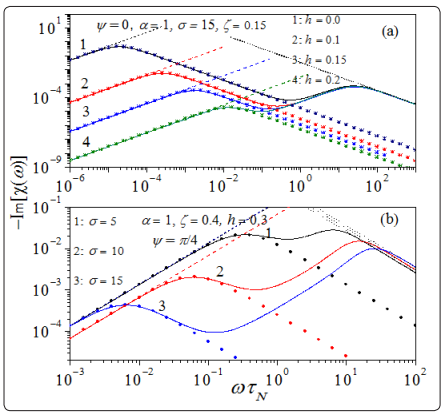

Figure 4

At low damping α<<1, there is an inherent geometric dependence of $\chi''(\omega)$ on the value of α arising from the *coupling of the longitudinal and transverse relaxation modes*. This coupling appears in the dynamical equation of motion and results in the appearance of the third antiferromagnetic resonance peak in the spectrum of $\chi''(\omega)$ due to excitation of transverse (precessional) modes with characteristic frequencies close to the precession frequency of the magnetization (see Figs. 5-7). This peak appears only at low damping (α<<1) and strongly manifests itself at high frequencies. As α decreases, the peak shifts to higher frequencies and its half-width decreases (in our normalized units, see Fig. 5). Clearly, Figs. 5-7, the agreement between the numerical calculation and Eq. (12) is very good at low-frequencies because the low-frequency response is mainly determined by the over barrier relaxation mode.

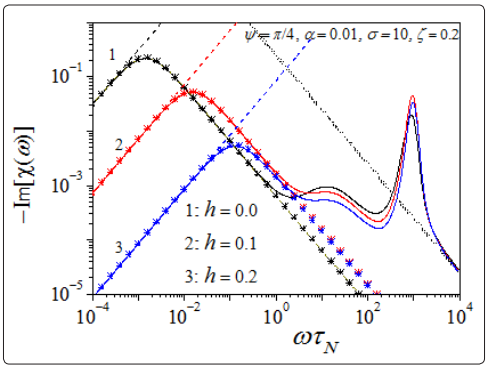

Figure 5

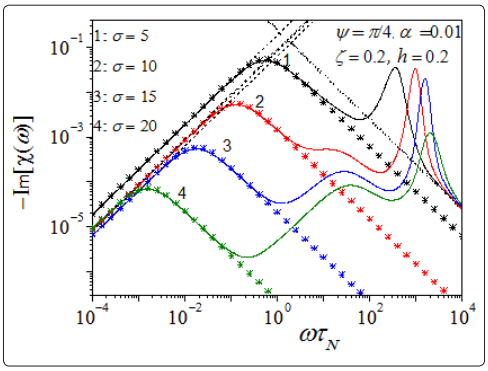

Figure 6

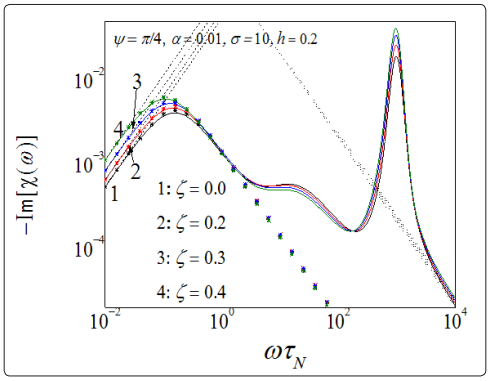

Figure 7

Our results demonstrate that variations in the bias field parameter h and antiferromagnetic parameter ζ significantly affect the magnetization relaxation process. These parameters are controlled, respectively, by the decompensation magnetic moment μ and χ_A parameter characterizing the induced magnetic moment of the particle. In our calculations, μ and χ_A were considered as model parameters. Conversely, μ can be estimated for randomly oriented spins as $\mu \sim z\,\mu_B N^{1/2}$, where z is the number of uncompensated spins per atoms, μ_B is the Bohr magneton, and N is the number of magnetic atoms [4]. Simple estimations show that the effective spontaneous magnetization of antiferromagnetic nanoparticles ranges from several tenths to several units of gauss, i.e., it is of the same order of magnitude as the magnetization of weak ferromagnets [4]. Furthermore, μ and χ_A as well as their temperature dependence can also be obtained experimentally from static magnetic measurements [31].

In conclusion, we have treated the longitudinal relaxation of the magnetization of antiferromagnetic particles subjected to a uniform external field **H** applied at an arbitrary angle to the easy axis of the particle (so that the axial symmetry is broken) using the kinetic model suggested by Raikher and Stepanov [4]. Numerically exact calculations of the observables (dynamic magnetic susceptibility, relaxation times of the magnetization, etc.) have been accomplished using an effective matrix continued fraction method. The main advantage of this method is that it allows us to evaluate the quantity of interest ($\chi(\omega)$, etc.) in wide ranges of damping and temperatures including relatively high temperatures, where asymptotic approaches (like that due to Kramers) are no longer applicable. We have shown that the magnetization dynamics in the presence of thermal agitation are very sensitive to both the dc field strength and orientation and damping owing to the coupling between the precession of the magnetization and its thermally activated reversal over the saddle point. In particular, the pronounced damping and dc field dependence of can be used to determine the damping coefficient α just as for uniaxial superparamagnets [19, 28]. Furthermore, we have shown that the simple analytic Eq. (12) provides an accurate description of the dynamic susceptibility ($\chi(\omega)$ of antiferromagnetic nanoparticles at low frequencies ($\omega\tau \leq 1$). This implies that the longtime behavior ($\tau << t$) of the longitudinal component of the magnetization $\langle M_{\square}\rangle(t)$ may be accurately approximated by a single exponential, viz., $\langle M_{\square}\rangle(t) - \langle M_{\square}\rangle_0 \sim \exp(-t/\tau)$

with the relaxation time τ from Eq. (16).

Here we have restricted ourselves to the study of a single particle. For practical applications, in order to account for the polydispersity of the particles of a real sample and the fact the particles are randomly oriented in space, one must also average the reversal time, dynamic susceptibility, etc., over appropriate distribution functions (averaging over particle volumes and orientations can be readily accomplished numerically using Gaussian quadratures). Our approach can also be used to estimate other physical parameters such as angular and temperature variations in the switching field of an individual nanoparticle and nonlinear dynamic susceptibilities. Furthermore, our results can be used to study stochastic resonance32 and dynamic hysteresis33 in antiferromagnetic nanoparticles which may differ essentially from those in fine ferromagnetic particles [32-37].

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Appendix A: Matrix Continued Fraction Solution
Equation (13) can be transformed into the tree-term vector recurrence equation

$$\tau_N \frac{d}{dt}\mathbf{C}_n(t) = \mathbf{Q}_n^{-}\mathbf{C}_{n-1}(t) + \mathbf{Q}_n\mathbf{C}_n(t) + \mathbf{Q}_n^{+}\mathbf{C}_{n+1}(t)\,. \qquad (A.1)$$

Here the column vectors $\mathbf{C}_n(t)$ are arranged from $C_{n,m}(t)$, viz.,

$$\mathbf{C}_0(t) = \mathbf{0}\,,\ \mathbf{C}_n(t) = \begin{pmatrix} c_{2n,-2n}(t) \\ c_{2n,-2n+1}(t) \\ \vdots \\ c_{2n,2n}(t) \\ c_{2n-1,-2n+1}(t) \\ c_{2n-1,-2n+2}(t) \\ \vdots \\ c_{2n-1,2n-1}(t) \end{pmatrix},$$

while the matrices Q_n, Q^+_n, Q^-_n are defined as

$$\mathbf{Q}_n = \begin{pmatrix} \mathbf{X}_{2n} & \mathbf{W}_{2n} \\ \mathbf{Y}_{2n-1} & \mathbf{X}_{2n-1} \end{pmatrix}, \ \mathbf{Q}^+_n = \begin{pmatrix} \mathbf{Z}_{2n} & \mathbf{Y}_{2n} \\ \mathbf{0} & \mathbf{Z}_{2n-1} \end{pmatrix}, \ \mathbf{Q}^-_n = \begin{pmatrix} \mathbf{V}_{2n} & \mathbf{0} \\ \mathbf{W}_{2n-1} & \mathbf{V}_{2n-1} \end{pmatrix} \quad \text{(A.2)}$$

In turn, the matrices Q_n, Q^+_n, Q^-_n consist of submatrices V_l, W_l, X_l Y_l, and Z_l which have the dimensions (2l+1) X (2l-3), (2l+1) X (2l-1), (2l+1) X (2l+1), (2l+1) X (2l+3), and (2l+1) X (2l+5), respectively. The elements of these submatrices are expresseed in terms of the matrix elements of the Fokker-Planck operator $d_{l,m,l',m'}$ and are given by

$$(\mathbf{V}_l)_{n,m} = \delta_{n-4,m} v^{--}_{l,-l+m+3} + \delta_{n-3,m} v^{-}_{l,-l+m+2} + \delta_{n-2,m} v_{l,-l+m+1} + \delta_{n-1,m} v^{+}_{l,-l+m} + \delta_{n,m} v^{++}_{l,-l+m-1},$$

$$(\mathbf{W}_l)_{n,m} = \delta_{n-3,m} w^{--}_{l,-l+m+2} + \delta_{n-2,m} w^{-}_{l,-l+m+1} + \delta_{n-1,m} w_{l,-l+m} + \delta_{n,m} w^{+}_{l,-l+m-1} + \delta_{n+1,m} w^{++}_{l,-l+m-2},$$

$$(\mathbf{X}_l)_{n,m} = \delta_{n-2,m} x^{--}_{l,-l+m+1} + \delta_{n-1,m} x^{-}_{l,-l+m} + \delta_{n,m} x_{l,-l+m-1} + \delta_{n+1,m} x^{+}_{l,-l+m-2} + \delta_{n+2,m} x^{++}_{l,-l+m-3},$$

$$(\mathbf{Y}_l)_{n,m} = \delta_{n-1,m} y^{--}_{l,-l+m} + \delta_{n,m} y^{-}_{l,-l+m-1} + \delta_{n+1,m} y_{l,-l+m-2} + \delta_{n+2,m} y^{+}_{l,-l+m-3} + \delta_{n+3,m} y^{++}_{l,-l+m-4},$$

$$(\mathbf{Z}_l)_{n,m} = \delta_{n,m} z^{--}_{l,-l+m-1} + \delta_{n+1,m} z^{-}_{l,-l+m-2} + \delta_{n+2,m} z_{l,-l+m-3} + \delta_{n+3,m} z^{+}_{l,-l+m-4} + \delta_{n+4,m} z^{++}_{l,-l+m-5},$$

where

$$x_{n,m} = -\frac{n(n+1)}{2} - i\frac{hm\sigma\gamma_3}{\alpha} + \sigma\left[h^2\zeta\sigma\left(1-3\gamma_3^2\right)+1\right]\frac{n(n+1)-3m^2}{(2n-1)(2n+3)},$$

$$\begin{matrix} x^{-}_{n,m} \\ x^{+}_{n,m} \end{matrix} = \pm(\gamma_1 \pm i\gamma_2)h\sigma\left[\frac{3h\zeta\gamma_3\sigma(2m\mp 1)}{(2n-1)(2n+3)} \mp \frac{i}{2\alpha}\right]\sqrt{(n+1\mp m)(n\pm m)}$$

$$\begin{matrix} x^{--}_{n,m} \\ x^{++}_{n,m} \end{matrix} = \frac{3\zeta h^2(\gamma_1 \pm i\gamma_2)^2\sigma^2}{2(2n-1)(2n+3)}\sqrt{(n-1\pm m)(n\pm m)(n+1\mp m)(n+2\mp m)},$$

$$y_{n,m} = -\sigma h\left(n\gamma_3 + m\frac{\zeta h\sigma\left(3\gamma_3^2-1\right)-1}{i\alpha}\right)\sqrt{\frac{(n+1)^2-m^2}{(2n+1)(2n+3)}}$$

$$\begin{matrix} y^{-}_{n,m} \\ y^{+}_{n,m} \end{matrix} = h\sigma(\gamma_1 \pm i\gamma_2)\left[\frac{ih\zeta\gamma_3\sigma}{\alpha}(n\pm 2m) \mp \frac{n}{2}\right]\sqrt{\frac{(n+1\mp m)(n+2\mp m)}{(2n+1)(2n+3)}},$$

$$\begin{matrix} y^{--}_{n,m} \\ y^{++}_{n,m} \end{matrix} = \pm\frac{ih^2\zeta\sigma^2(\gamma_1 \pm i\gamma_2)^2}{2\alpha}\sqrt{\frac{(n\pm m)(n+1\mp m)(n+2\mp m)(n+3\mp m)}{(2n+1)(2n+3)}},$$

$$w_{n,m} = \frac{\sigma\left[\gamma_3 h\alpha(n+1) + im\left[h^2\zeta\left(3\gamma_3^2-1\right)\sigma-1\right]\right]}{2}\sqrt{\frac{n^2-m^2}{4n^2-1}},$$

$$\begin{matrix} w^{-}_{n,m} \\ w^{+}_{n,m} \end{matrix} = \mp(\gamma_1 \pm i\gamma_2)h\sigma\left[\frac{n+1}{2} \mp \frac{ih\sigma\zeta\gamma_3}{\alpha}(n+1\mp 2m)\right]\sqrt{\frac{(n\pm m)(n-1\pm m)}{4n^2-1}},$$

$$\begin{matrix} w^{--}_{n,m} \\ w^{++}_{n,m} \end{matrix} = \mp\frac{i(\gamma_1 \pm i\gamma_2)^2\zeta h^2\sigma^2}{2\alpha}\sqrt{\frac{(n-2\pm m)(n-1\pm m)(n+1\mp m)(n\pm m)}{4n^2-1}},$$

$$z_{n,m} = \frac{n\sigma\left[\zeta h^2\sigma\left(3\gamma_3^2-1\right)-1\right]}{2n+3}\sqrt{\frac{\left[(n+1)^2-m^2\right]\left[(n+2)^2-m^2\right]}{(2n+1)(2n+5)}},$$

$$\begin{matrix} z^{-}_{n,m} \\ z^{+}_{n,m} \end{matrix} = \pm\frac{2h^2\zeta(\gamma_1 \pm i\gamma_2)\gamma_3\sigma^2 n}{2n+3}\sqrt{\frac{\left[(n+1)^2-m^2\right](n+2\mp m)(n+3\mp m)}{(2n+1)(2n+5)}},$$

$$\begin{matrix} z^{--}_{n,m} \\ z^{++}_{n,m} \end{matrix} = \frac{h^2\zeta(\gamma_1 \pm i\gamma_2)^2\sigma^2 n}{2(2n+3)}\sqrt{\frac{(n+1\mp m)(n+2\mp m)(n+3\mp m)(n+4\mp m)}{(2n+1)(2n+5)}},$$

$$v_{n,m} = -\frac{\sigma\left[\zeta h^2\sigma\left(3\gamma_3^2-1\right)-1\right](n+1)}{(2n-1)}\sqrt{\frac{\left[(n-1)^2-m^2\right](n^2-m^2)}{(2n+1)(2n-3)}},$$

$$\begin{matrix} v^{-}_{n,m} \\ v^{+}_{n,m} \end{matrix} = \pm\frac{2\zeta h^2(\gamma_1 \pm i\gamma_2)\gamma_3\sigma^2(n+1)}{(2n-1)}\sqrt{\frac{(n-2\pm m)(n-1\pm m)(n^2-m^2)}{(2n+1)(2n-3)}},$$

$$\begin{matrix} v^{--}_{n,m} \\ v^{++}_{n,m} \end{matrix} = -\frac{\zeta h^2(\gamma_1 \pm i\gamma_2)^2\sigma^2(n+1)}{2(2n-1)}\sqrt{\frac{(n-3\pm m)(n-2\pm m)(n-1\pm m)(n\pm m)}{(2n+1)(2n-3)}}.$$

The exact solution of Eq. (A.1) for the Laplace transform $\tilde{\mathbf{C}}_1(s) = \int_0^\infty \mathbf{C}_1(t)e^{-st}dt$ can be given in terms of matrix continued fractions [10].

$$\tilde{\mathbf{C}}_1(s) = \tau_N\mathbf{\Delta}_1(s)\left\{\mathbf{C}_1(0) + \sum_{n=2}^{\infty}\left[\prod_{k=2}^{n}\mathbf{Q}^+_{k-1}\mathbf{\Delta}_k(s)\right]\mathbf{C}_n(0)\right\}, \quad \text{(A.3)}$$

the infinite matrix continued fraction $\Delta_n(S)$ is defined by the recurrence equation

$$\mathbf{\Delta}_n(s) = \left[\tau_N s\mathbf{I} - \mathbf{Q}_n - \mathbf{Q}^+_n\,\mathbf{\Delta}_{n+1}(s)\mathbf{Q}^-_{n+1}\right]^{-1}.$$

The initial value vectors $\mathbf{C}_n(0)$ can be evaluated in term of $\Delta_n(0)$. Here we may apply with small modifications the algorithm developed for uniaxial anisotropy [10]. As shown in Ref. 10, Sect. 9.2.2, the initial vectors $\mathbf{C}_n(0)$ are given by

$$\mathbf{C}_n(0) = \frac{\xi_1}{\sqrt{4\pi}}\left[\bar{\mathbf{K}}_n + \left[\mathbf{K}_n + \bar{\mathbf{K}}^H_{n+1}\mathbf{S}_{n+1}\right]\mathbf{S}_n\right]\mathbf{S}_{n-1}..\mathbf{S}_1,$$

where $\xi_1 = \beta\mu H_1$, $\mathbf{S}_n = \Delta_n(0)\ \mathbf{Q}^-_n$, the superscript H designed the Hermitian (i.e., transposition and complex) conjugate, and

$$\mathbf{K}_n = \begin{bmatrix} \mathbf{F}_{2n} & \mathbf{D}_{2n} \\ \mathbf{D}^H_{2n} & \mathbf{F}_{2n-1} \end{bmatrix},$$

$$\bar{\mathbf{K}}_n = \begin{bmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{D}_{2n-1} & \mathbf{0} \end{bmatrix}, \ \bar{\mathbf{K}}_1 = \begin{pmatrix} \mathbf{0} \\ \mathbf{D}_1 \end{pmatrix}.$$

The matrix $\mathbf{K}_n$ and $\bar{\mathbf{K}}_n$ are constituted from the diagonal submatrix and the tridiagonal submatrix $\mathbf{F}_l$ with the matrix elements defined as

$$(\mathbf{F}_l)_{n,m} = -\sqrt{\frac{4\pi}{3}}\,\mathrm{Re}\left[\gamma_3\langle Y_{1,0}\rangle_0 - \sqrt{2}(\gamma_1 - i\gamma_2)\langle Y_{1,1}\rangle_0\right]\delta_{n,m},$$

$$(\mathbf{D}_l)_{n,m} = \delta_{n-2,m} d^{-}_{l,-l+m+1} + \delta_{n-1,m} d_{l,-l+m} + \delta_{n,m} d^{+}_{l,-l+m-1}$$

with

$$d_{n,m} = \gamma_3\sqrt{\frac{n^2-m^2}{4n^2-1}},$$

$$d^{-}_{n,m} = -\left(d^{+}_{n,-m}\right)^* = -\frac{(\gamma_1 + i\gamma_2)}{2}\sqrt{\frac{(n+m-1)(n+m)}{4n^2-1}}.$$

The smallest nonvanishing eigenvalue λ_1 of the Fokker-Planck operator can also be estimated by using matrix continued fractions from the secular equation as [10, 11].

$$\det\left[\lambda_1\tau_N\mathbf{I} + \mathbf{Q}_1 + \mathbf{Q}^+_1\mathbf{\Delta}_2(-\lambda_1)\mathbf{Q}^-_2\right] = 0. \quad \text{(A.4)}$$

In the low temperature limit, the behavior of λ_1 must correspond to the Kramers escape rate so providing a numerical check on the asymptotic Eq. (16) for the reversal time $\tau \approx 1/\lambda_1$ [6, 23].

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