# Effect of a dc bias magnetic field on the magnetization relaxation of antiferromagnetic nanoparticles

Bachir Ouari and Yuri P. Kalmykov
*LAMPS, Université de Perpignan, 52, Avenue Paul Alduy, 66860 Perpignan Cedex, France*
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The relaxation of the magnetization of antiferromagnetic nanoparticles owing to thermal agitation is treated via the Fokker-Planck equation describing the evolution of the distribution function of the magnetization orientations of an individual nanoparticle. By solving this equation using matrix-continued fractions, the correlation function of the longitudinal component of the magnetization, its characteristic relaxation times, and dynamic susceptibility are calculated for arbitrary dc field orientations across wide ranges of frequencies, temperatures, and damping. Furthermore, a simple analytic equation for the dynamic susceptibility at low frequencies is also proposed. It is shown that a dc field applied at an angle to the easy axis of the particle alters essentially the magnetization dynamics of the particle owing to coupling of the magnetization reversal mode with the precessional modes.

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## I. INTRODUCTION

Fine *ferromagnetic* particles are characterized by thermal instability[1] of their magnetization resulting in spontaneous change in their orientation from one metastable state to another by surmounting energy barriers, giving rise to superparamagnetism.[2,3] For *antiferromagnetic* nanoparticles, the ensuing thermal instability of the magnetization may differ in many respects from those of ferromagnetic nanoparticles because of the intrinsic properties of antiferromagnetic materials. Moreover, the magnetic behavior of antiferromagnetic nanoparticles can be quite different from that observed in the bulk.[4,5] The basic theory of antiferromagnetic nanoparticles was developed by Néel,[6,7] who concluded 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. 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,[6,7] 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. An understanding of 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 initial analytic treatment of the thermal fluctuations of the magnetization of fine magnetic particles owing to Néel[1] based on the classical transition state theory was further developed by Brown[8,9] and is consequently known as the Néel-Brown theory. This treatment 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 derive the particular Fokker-Planck equation describing the time evolution of the probability density function of magnetization orientations. At temperatures much lower than the ordering (Néel) temperature $T_N$, this theory may be adapted to antiferromagnetic nanoparticles, as has been suggested by Raikher and Stepanov[4] 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. For an antiferromagnetic particle subjected to a dc magnetic field $\mathbf{H}$, the magnetic moments of the sublattices $\mathbf{m}_1$ and $\mathbf{m}_2$ are given by[4]

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

where $M_S$ is the sublattice magnetization in a bulk, $\chi_A$ is a parameter characterizing the induced magnetic moment of the particle, $\mathbf{u} = (\mathbf{m}_1 - \mathbf{m}_2)/2vM_S$ is the unit vector along the decompensation magnetic moment $\mu = \mathbf{u}\mu$, and $v$ is the particle volume. The free energy of the particle subjected to a dc magnetic field $\mathbf{H}$ applied at an angle to the easy axis is

$$\begin{aligned} V(\vartheta,\varphi) = \beta^{-1}\sigma[&\sin^2\vartheta - 2h(\gamma_1\sin\vartheta\cos\varphi \\ &+ \gamma_2\sin\vartheta\sin\varphi + \gamma_3\cos\vartheta) \\ &+ 2\sigma h^2\zeta(\gamma_1\sin\vartheta\cos\varphi \\ &+ \gamma_2\sin\vartheta\sin\varphi + \gamma_3\cos\vartheta)^2], \end{aligned} \quad (1)$$

where $\vartheta$ and $\varphi$ are the polar and azimuthal angles of the spherical coordinate system, $\beta = (kT)^{-1}$, $k$ is Boltzmann's constant, $T$ is the absolute temperature, $\sigma = v\beta K$ is the dimensionless anisotropy parameter, $K$ 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 $\gamma_1$, $\gamma_2$, $\gamma_3$ are the direction cosines of the vector $\mathbf{H}$. As long as the applied field $\mathbf{H}$ is much weaker than the exchange field, the only possible motion of the vector $\mu$ is rotation that 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 $W(\mu,t)$ of $\mu$, viz.,

$$\begin{aligned} \frac{\partial}{\partial t}W = L_{\mathrm{FP}}W = \frac{1}{2\tau_N}\{&\beta[\alpha^{-1}\mathbf{u}\cdot(\nabla V\times\nabla W) \\ &+ \nabla\cdot(W\nabla V)] + \nabla^2 W\}, \end{aligned} \quad (2)$$

where $L_{\mathrm{FP}}$ is the Fokker-Planck operator, $\nabla = \partial_{\mathbf{u}}$ is the gradient operator on the unit sphere, $\tau_N$ is the free diffusion time of the magnetization, and $\alpha$ is the dimensionless damping parameter.

 

When a dc magnetic field is 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 $\varphi$. Owing 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]

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

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

The goal of the present paper is to study the longitudinal relaxation of the magnetization of antiferromagnetic nanoparticles subjected to a dc magnetic field applied at an *arbitrary* angle to the easy axis of a particle (so that the axial symmetry is broken). We present results of calculations of the longitudinal complex magnetic susceptibility $\chi(\omega)$ of an 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 that characterize, respectively, the long, short, and overall behavior of the magnetization. The calculations are mainly accomplished by using the matrix-continued fractions,[10] however, simple analytic equations for the low- and high-frequency parts of the spectrum $\chi(\omega)$ and relaxation times are also obtained. In the low-temperature limit, our matrix-continued fraction solution for the reversal time of the magnetization $\tau$ agrees with independent analytic estimates of Ouari *et al.*,[11] who have evaluated $\tau$ of antiferromagnetic nanoparticles by adapting the Kramers escape rate theory[12,13] to fine ferromagnetic particles given by Coffey *et al.*[14] We remark in passing that in the limiting case $\zeta = 0$, the free energy from Eq. (1) reduces to that of uniaxial superparamagnets; this case has been treated in Refs. 10 and 15–19.

## II. BASIC EQUATIONS

A concise theoretical description of the longitudinal relaxation of the magnetization in antiferromagnetic nanoparticles can be given by linear response theory (see Ref. 10, Chap. 2). Here it is supposed that a particle in the presence of a strong uniform magnetic field $\mathbf{H}$ is subjected in addition to a small probe field $\mathbf{H}_1$ [$\beta(\mu\cdot\mathbf{H}_1) \ll 1$] parallel to $\mathbf{H}$. Then the decay of the longitudinal component of the averaged magnetization $\langle M_{||}\rangle(t) = v^{-1}\langle\mu_{||}\rangle(t)$ of the particle, when the field $\mathbf{H}_1$ has been switched off at time $t = 0$, is[10]

$$\langle M_\parallel\rangle(t) - \langle M_\parallel\rangle_0 = \chi C_\parallel(t)H_1, \quad (4)$$

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

$$C_\parallel(t) = \frac{\langle M_\parallel(0)M_\parallel(t)\rangle_0 - \langle M_\parallel(0)\rangle_0^2}{\langle M_\parallel^2(0)\rangle_0 - \langle M_\parallel(0)\rangle_0^2} = \sum_{k=1}^{k=\infty} c_k e^{-\lambda_k t}. \quad (5)$$

$\lambda_k$ are the eigenvalues of the Fokker-Planck operator $L_{\rm FP}$ in Eq. (2), $\sum_{k=1}^{k=\infty} c_k = 1$, $\chi = v\beta[\langle M_\parallel^2(0)\rangle_0 - \langle M_\parallel(0)\rangle_0^2]$ is 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 $C_\parallel(t)$, one can calculate the longitudinal dynamic susceptibility of the particle $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$ given by[10]

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

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

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

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

$$\frac{\chi(\omega)}{\chi} \sim \begin{cases} 1 - i\omega\tau_{\rm cor} + \cdots, & \omega\to 0, \\ -i\,(\omega\tau_{\rm ef})^{-1} + \cdots, & \omega\to\infty, \end{cases} \quad (7)$$

where

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

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

$$\tau_{\rm cor} = \int_0^\infty C_\parallel(t)dt, \quad (9)$$

characterizes the overall behavior of $C_\parallel(t)$ while the effective relaxation time $\tau_{\rm ef}$ yields precise information on the initial decay of $C_\parallel(t)$, namely,

$$\tau_{\rm ef} = -1/\dot{C}_\parallel(0). \quad (10)$$

The relaxation times $\tau_{\rm cor}$ and $\tau_{\rm ef}$ contain contributions from all the eigenvalues $\lambda_k$ of the Fokker-Planck operator $L_{\rm FP}$. The smallest nonvanishing eigenvalue $\lambda_1$ 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 $\lambda_k$ characterize high-frequency "intrawell" modes. The dependences of the effective relaxation time $\tau_{\rm ef}$ on the model parameters (external field and anisotropy constants) may differ considerably from those of $\tau_{\rm cor}$ and $\tau$ as $\tau_{\rm ef}$ is not governed by $\lambda_1$. The effective relaxation time $\tau_{\rm ef}$ can also be expressed in terms of equilibrium averages as[10]

$$\tau_{\rm ef} = 2\tau_N\frac{\langle u_\parallel^2\rangle_0 - \langle u_\parallel\rangle_0^2}{1 - \langle u_\parallel^2\rangle_0}, \quad (11)$$

where $u_\parallel = \cos\psi\cos\vartheta + \sin\psi\sin\vartheta\cos\varphi$. 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 $\psi$ is the angle

between **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 by 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]

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

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

## III. CALCULATION OF THE OBSERVABLES

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

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

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 the Appendix.

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 the Appendix). Having determined $\tilde{c}_{l,m}(\omega)$, we can evaluate the spectrum $C_\parallel(\omega) = \int_0^\infty e^{-i\omega t}C_\parallel(t)dt$ of the longitudinal relaxation function $C_\parallel(t)$ as

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

as well as the dynamic susceptibility $\chi(\omega)$ from Eq. (6). Moreover, by using matrix-continued fractions, we can also evaluate the integral relaxation time

$$\tau_{\rm cor} = \tilde{C}_\parallel(0), \tag{15}$$

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

Now the smallest nonvanishing eigenvalue $\lambda_1$ characterizes the slowest overbarrier relaxation mode and, hence, the long-time behavior of the magnetization. In order to find a low-temperature (high barrier) asymptotic estimate for $\lambda_1$ of the Fokker-Planck operator $L_{\rm FP}$ in Eq. (2), Brown[9] and Smith and De Rozario[21] adapted to magnetic relaxation an ingenious method originally proposed by Kramers[12,13] for thermally activated escape of point Brownian particles from a potential well. Thus they estimated the superparamagnetic relaxation time $\tau = 1/\lambda_1$ in the so-called intermediate-to-high damping (IHD) limit ($\alpha \geqslant 1$). Later, Klik and Gunther[22,23] derived the corresponding formula for $\tau$ in the very low damping limit ($\alpha \ll 1$). Finally, Coffey *et al.*[14] have obtained the asymptotic formula for $\tau$, which is valid for all values of damping. The results of Coffey *et al.*[14] agree closely with numerical solutions of the Fokker-Planck equation (2) (Refs. 18 and 24) and Langevin dynamics simulations[25–27] 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.[28] For antiferromagnetic nanoparticles with the free energy, Eq. (1), the reversal time of the magnetization $\tau$ have been estimated analytically by Ouari *et al.*[11] by using the approach of Coffey *et al.*[14] as

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

where $\tau_{\rm IHD}$ is the reversal time in the IHD limit, $\alpha \geqslant 1$, $S_{1,2}$ are the dimensionless actions, and $A(\delta)$ is the so-called depopulation factor[14] (equations for $\tau_{\rm IHD}$, $A$, and $S_{1,2}$ are given in Ref. 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.*.[11] have derived from Eq. (3) the analytic equation for the reversal time $\tau$ in the low-temperature limit, viz.,

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

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 given in 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, because the dynamics of the system are governed by a single variable $\vartheta$, the integral relaxation time $\tau_{\rm cor} = \tilde{C}_\parallel(0)$ can also be calculated from the analytic equation as (see Ref. 10, Chap. 2, Sec. 2.10 for details)

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

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

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

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

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 $\tau_{\mathrm{ef}}$ from Eq. (11) is given by

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

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

## IV. RESULTS AND DISCUSSION

The inverse of the smallest nonvanishing eigenvalue of the Fokker-Planck equation $\lambda_1^{-1}$, the integral relaxation time $\tau_{\mathrm{cor}}$ (both calculated with the matrix-continued fraction method), and the reversal time $\tau$ predicted by Eq. (16) as functions of the anisotropy (or the inverse temperature) parameter $\sigma$ 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 $\tau_{\mathrm{cor}}$ may have a behavior dramatically different from that of $\lambda_1^{-1}$ 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 $\tau$ may differ exponentially from $\tau_{\mathrm{cor}}$ owing to the so-called depletion effect.[30] This effect is qualitatively similar to that for ferromagnetic nanoparticles.[10,30] The integral relaxation time $\tau_{\mathrm{cor}}$ as functions of the antiferromagnetic parameter $\zeta$ calculated numerically by the matrix-continued fraction method are shown in Fig. 2 for various values of the oblique angle $\psi$. This figure demonstrates that the variations in the antiferromagnetic parameter $\varsigma$ have a very pronounced effect on the relaxation process.

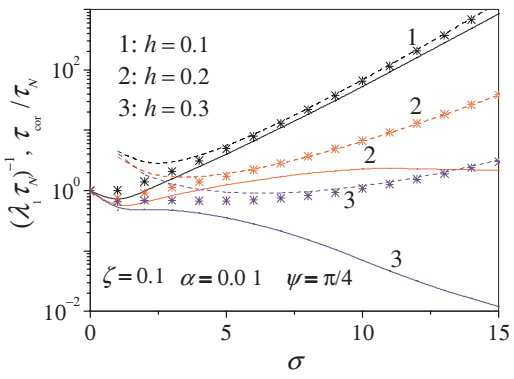


FIG. 1. (Color online) $\tau_{\mathrm{cor}}/\tau_N$ and $(\lambda_1\tau_N)^{-1}$ vs the anisotropy (inverse temperature) parameter $\sigma$ for $\zeta = 0.1$, $\alpha = 0.01$, $\psi = \pi/4$, and various values of the field parameter $h$. Solid lines 1–3: Matrix-continued fraction solution for $\tau_{\mathrm{cor}}/\tau_N$, Eqs. (9) and (15). Dashed lines: Eq. (16). Symbols: Eq. (A3).

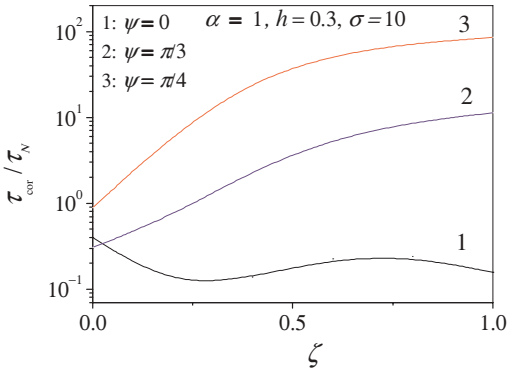


FIG. 2. (Color online) $\tau_{\mathrm{cor}}/\tau_N$ vs the antiferromagnetic parameter $\zeta$ for $\sigma = 10$, $\alpha = 1$, $h = 0.3$, and various values of the angle $\psi$.

Figures 3 and 4 illustrate the results of the calculation of the imaginary part of the susceptibility $\chi''(\omega) = \chi\omega\,\mathrm{Re}[\tilde{C}_{\parallel}(\omega)]$ for $\alpha = 1$ (moderate damping) and various values of the model parameters $\sigma$, $\psi$, and $h$, using the matrix-continued fraction solution and the approximate Eq. (12). These figures indicate that at $\alpha \geqslant 1$ 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 $\omega_1$ and half-width $\Delta\omega_1$ of this band are completely determined by the smallest nonvanishing eigenvalue $\lambda_1$. In addition, a far weaker second relaxation band appears at high frequencies. This relaxation band is owing to the individual near degenerate high-frequency "intrawell" modes corresponding to the eigenvalues $\lambda_k \gg \lambda_1$. 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. 4(a)].

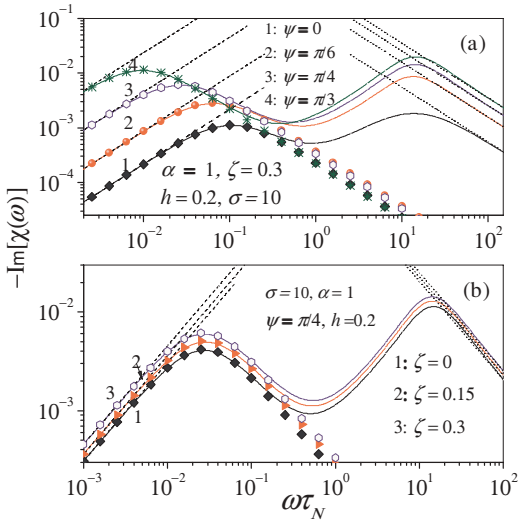


FIG. 3. (Color online) $-\mathrm{Im}[\chi(\omega)]$ vs $\omega\tau_N$ (a) for $\alpha = 1$, $h = 0.2$, $\zeta = 0.3$, $\sigma = 10$, and various values of $\psi$ and (b) for $h = 0.2$, $\alpha = 1$, $\sigma = 10$, $\psi = \pi/4$ and various values of $\zeta$. Solid lines: Matrix-continued fraction solution, Eqs. (6) and (14). Symbols: Eq. (12); dotted and dashed lines: Eqs. (7), (9), and (11).

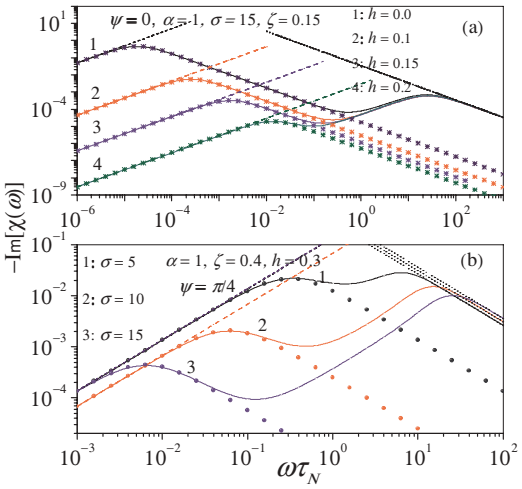


FIG. 4. (Color online) $-\mathrm{Im}[\chi(\omega)]$ vs $\omega\tau_N$ (a) for $\sigma = 15$, $\alpha = 1$ (moderate damping), $\zeta = 0.15$, $\psi = 0$, and various values of the field parameter $h$ and (b) for $h = 0.3$, $\alpha = 1$, $\zeta = 0.15$, $\psi = \pi/4$, and various values of the anisotropy parameter $\sigma$. Solid lines: Matrix-continued fraction solution, Eqs. (6) and (14). Symbols: Eq. (12); dotted and dashed lines: Eqs. (7), (9), and (11).

At low damping $\alpha \ll 1$, there is an inherent geometric dependence of $\chi''(\omega)$ on the value of $\alpha$ 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 $\chi''(\omega)$ owing 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 ($\alpha \ll 1$) and strongly manifests itself at high frequencies. As $\alpha$ decreases, the peak shifts to higher frequencies and its half-width decreases (in our normalized units, see Fig. 5). Clearly, in 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 overbarrier relaxation mode.

Our results demonstrate that variations in the bias field parameter $h$ and antiferromagnetic parameter $\zeta$ significantly

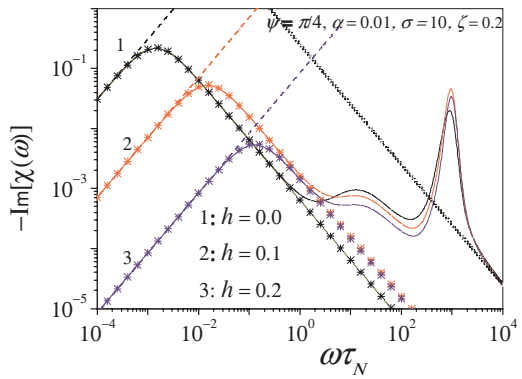


FIG. 5. (Color online) $-\mathrm{Im}[\chi(\omega)]$ vs $\omega\tau_N$ for $\alpha = 0.01$, $\sigma = 10$, $\zeta = 0.2$, $\psi = \pi/4$, and various values of $h$. Solid lines: Matrix-continued fraction solution, Eqs. (6) and (14). Symbols: Eq. (12); dotted and dashed lines: Eqs. (7), (9), and (11).

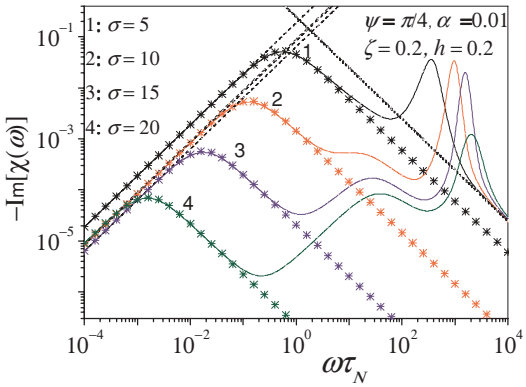


FIG. 6. (Color online) $-\mathrm{Im}[\chi(\omega)]$ vs $\omega\tau_N$ for $\alpha = 0.01$, $h = 0.2$, $\zeta = 0.2$, $\psi = \pi/4$, and various values of $\sigma$. Solid lines: Matrix-continued fraction solution, Eqs. (6) and (14). Symbols: Eq. (12); dotted and dashed lines: Eqs. (7), (9), and (11).

affect the magnetization relaxation process. These parameters are controlled, respectively, by the decompensation magnetic moment $\mu$ and parameter $\chi_A$, characterizing the induced magnetic moment of the particle. In our calculations, $\mu$ and $\chi_A$ were considered as model parameters. Conversely, $\mu$ can be estimated for randomly oriented spins as[4] $\mu \sim z\mu_B N^{1/2}$, where $z$ is the number of uncompensated spins per atoms, $\mu_B$ is the Bohr magneton, and $N$ is the number of magnetic atoms. Simple estimations show[4] 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. Furthermore, $\mu$ and $\chi_A$ as well as their temperature dependence also can 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) by using the kinetic model suggested by Raikher and Stepanov.[4] Numerically exact calculations of the observables (dynamic magnetic susceptibility, relaxation times of

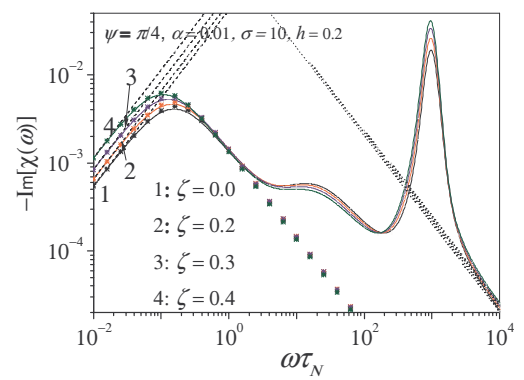


FIG. 7. (Color online) $-\mathrm{Im}[\chi(\omega)]$ vs $\omega\tau_N$ for $\alpha = 0.01$, $h = 0.2$, $\sigma = 10$, $\psi = \pi/4$, and various values of the antiferromagnetic parameter $\zeta$. Solid lines: Matrix-continued fraction solution, Eqs. (6) and (14). Symbols: Eq. (12); dotted and dashed lines: Eqs. (7), (9), and (11).

the magnetization, etc.) have been accomplished by 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 (such as that owing 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 $\chi(\omega)$ can be used to determine the damping coefficient $\alpha$ 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 ($\omega\tau \leqslant 1$). This implies that the long-time behavior ($\tau \ll t$) of the longitudinal component of the magnetization $\langle M_\parallel\rangle(t)$ may be accurately approximated by a *single* exponential, viz., $\langle M_\parallel\rangle(t) - \langle M_\parallel\rangle_0 \sim \exp(-t/\tau)$ with the relaxation time $\tau$ 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 by using Gaussian quadratures). Our approach also can 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 resonance[32] and dynamic hysteresis[33] in antiferromagnetic nanoparticles, which may differ essentially from those in fine ferromagnetic particles.[34–37]

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## APPENDIX: MATRIX-CONTINUED FRACTION SOLUTION

Equation (13) can be transformed into the tree-term vector recurrence equation

$$\tau_N \frac{d}{dt}\mathbf{C}_n(t) = \mathbf{Q}_n^- \mathbf{C}_{n-1}(t) + \mathbf{Q}_n \mathbf{C}_n(t) + \mathbf{Q}_n^+ \mathbf{C}_{n+1}(t). \quad \text{(A1)}$$

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

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

while the matrices $\mathbf{Q}_n$, $\mathbf{Q}_n^+$, $\mathbf{Q}_n^-$ are defined as

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

In turn, the matrices $\mathbf{Q}_n$, $\mathbf{Q}_n^+$, $\mathbf{Q}_n^-$ consist of submatrices $\mathbf{V}_l$, $\mathbf{W}_l$, $\mathbf{X}_l$ $\mathbf{Y}_l$, and $\mathbf{Z}_l$, which have the dimensions $(2l+1)\times(2l-3)$, $(2l+1)\times(2l-1)$, $(2l+1)\times(2l+1)$, $(2l+1)\times(2l+3)$, and $(2l+1)\times(2l+5)$, respectively. The elements of these submatrices are expressed in terms of the matrix elements of the Fokker-Planck operator $d_{l,m,l',m'}$ and are given by

$$\begin{aligned}
(\mathbf{V}_l)_{n,m} &= \delta_{n-4,m} v^{--}_{l,-l+m+3} + \delta_{n-3,m} v^{-}_{l,-l+m+2} \\
&\quad + \delta_{n-2,m} v_{l,-l+m+1} + \delta_{n-1,m} v^{+}_{l,-l+m} + \delta_{n,m} v^{++}_{l,-l+m-1}, \\
(\mathbf{W}_l)_{n,m} &= \delta_{n-3,m} w^{--}_{l,-l+m+2} + \delta_{n-2,m} w^{-}_{l,-l+m+1} + \delta_{n-1,m} w_{l,-l+m} \\
&\quad + \delta_{n,m} w^{+}_{l,-l+m-1} + \delta_{n+1,m} w^{++}_{l,-l+m-2}, \\
(\mathbf{X}_l)_{n,m} &= \delta_{n-2,m} x^{--}_{l,-l+m+1} + \delta_{n-1,m} x^{-}_{l,-l+m} + \delta_{n,m} x_{l,-l+m-1} \\
&\quad + \delta_{n+1,m} x^{+}_{l,-l+m-2} + \delta_{n+2,m} x^{++}_{l,-l+m-3}, \\
(\mathbf{Y}_l)_{n,m} &= \delta_{n-1,m} y^{--}_{l,-l+m} + \delta_{n,m} y^{-}_{l,-l+m-1} + \delta_{n+1,m} y_{l,-l+m-2} \\
&\quad + \delta_{n+2,m} y^{+}_{l,-l+m-3} + \delta_{n+3,m} y^{++}_{l,-l+m-4}, \\
(\mathbf{Z}_l)_{n,m} &= \delta_{n,m} z^{--}_{l,-l+m-1} + \delta_{n+1,m} z^{-}_{l,-l+m-2} + \delta_{n+2,m} z_{l,-l+m-3} \\
&\quad + \delta_{n+3,m} z^{+}_{l,-l+m-4} + \delta_{n+4,m} z^{++}_{l,-l+m-5},
\end{aligned}$$

where

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

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

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

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

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

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

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

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

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

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

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

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

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

The exact solution of Eq. (A1) 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]

$$\tilde{\mathbf{C}}_1(s) = \tau_N\Delta_1(s)\left\{\mathbf{C}_1(0) + \sum_{n=2}^{\infty}\left[\prod_{k=2}^{n}\mathbf{Q}^+_{k-1}\Delta_k(s)\right]\mathbf{C}_n(0)\right\}; \tag{A2}$$

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

$$\Delta_n(s) = [\tau_N s\mathbf{I} - \mathbf{Q}_n - \mathbf{Q}^+_n\Delta_{n+1}(s)\mathbf{Q}^-_{n+1}]^{-1}.$$

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

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

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

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

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

$$\widehat{\mathbf{K}}_1 = \begin{pmatrix}\mathbf{0} \\ \mathbf{D}_1\end{pmatrix}.$$

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

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

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

with

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

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

The smallest nonvanishing eigenvalue $\lambda_1$ of the Fokker-Planck operator can also be estimated by using matrix-continued fractions from the secular equation as[10,11]

$$\det[\lambda_1 \tau_N \mathbf{I} + \mathbf{Q}_1 + \mathbf{Q}_1^+ \Delta_2(-\lambda_1)\mathbf{Q}_2^-] = 0. \qquad \text{(A3)}$$

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


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