Reversal time of the magnetization of antiferromagnetic nanoparticles

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The reversal time of the magnetization τ of antiferromagnetic nanoparticles is evaluated for a uniform magnetic field applied at an arbitrary angle ψ to the easy axis by using the adaptation of the Kramers escape rate theory to fine ferromagnetic particles given by Coffey *et al.* [Adv. Chem. Phys. **117**, 483 (2001); Phys. Rev. E **63**, 021102 (2001)]. The resulting analytic formula yields an accurate approximation to the reversal time τ for all values of the damping and agrees favorably with the numerically exact solution of the corresponding Fokker-Planck equation for the evolution of the probability density function of magnetization orientations.

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I. INTRODUCTION

Ferromagnetic nanoparticles are characterized by an internal potential, which may have several local equilibrium states with potential barriers between them. If the particles are small (~100 Å) so that these barriers are relatively low, the magnetization may escape from one potential well to another due to thermal agitation.[1] The ensuing thermal instability of the magnetization results in the phenomenon of superparamagnetism[2,3] because each fine particle behaves like an enormous paramagnetic atom having a magnetic moment $\sim 10^4$–10^5 bohr magnetons. In the particular case of *antiferromagnetic* nanoparticles, the dynamics 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, e.g., enhanced magnetic moment and coercivity, exchange bias, increase in magnetic moment with temperature, decrease in the susceptibility with temperature below the ordering (Néel) temperature T_N and its enhancement compared to that in bulk.[4] The basic theory of antiferromagnetic nanoparticles was developed by Néel,[5,6] who concluded that the 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,[5,6] 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. Indeed measurements on ferritin and ferrihydrite showed[7] that the effective bulk susceptibility of these particles exceeds that of a macrocrystal by a factor of 2 or 3. 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]

The initial theory of thermal fluctuations of the magnetization of fine magnetic particles due to Néel[1] was further developed by Brown[8,9] using the theory of stochastic processes. At temperatures much lower than T_N, this theory may be adapted to antiferromagnetic nanoparticles.[4] In the simplest case, the magnetic moments of the sublattices $\mathbf{m}_1$ and $\mathbf{m}_2$ of an antiferromagnetic particle subjected to a dc magnetic field $\mathbf{H}$ are given by[4]

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

where M_S is the sublattice magnetization in a bulk, χ_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 $\boldsymbol{\mu}=\mathbf{u}\mu$, and v is the particle volume. As long as the applied field $\mathbf{H}$ is much weaker than the exchange field, the only possible motion of the vector $\boldsymbol{\mu}$ is rotation. Thus in the context of the Brown model,[8,9] the magnetization dynamics of uniaxial antiferromagnetic nanoparticles are similar to the rotations of Brownian particles in liquids and are governed by a Fokker-Planck equation for the probability density function W of $\mathbf{M}$, viz.,

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

where L_{FP} is the Fokker-Planck operator, the operators ∇ and Δ are the gradient and Laplacian on the surface of a sphere of unit radius,

$$\tau_N = \beta\mu(1+\alpha^2)/(2\gamma\alpha) \tag{3}$$

is the free diffusion time of the magnetization, $\beta=(kT)^{-1}$, k is Boltzmann's constant, T is the absolute temperature, and α is the dimensionless dissipation parameter. Here the normalized free energy of the particle subjected to a dc magnetic field $\mathbf{H}$ applied at an angle ψ to the easy axis of the magnetization is[4]

$$\beta V = \sigma[\sin^2\vartheta - 2h(\cos\psi\cos\vartheta + \sin\psi\sin\vartheta\cos\varphi) + 2\sigma h^2\zeta(\cos\psi\cos\vartheta + \sin\psi\sin\vartheta\cos\varphi)^2], \tag{4}$$

where $\sigma=v\beta K$ is the dimensionless anisotropy parameter, K is the anisotropy constant, $h=\xi/(2\sigma)$ is the applied field parameter, $\xi=\beta\mu H$, and $\zeta=v\chi_A/\beta\mu^2$ is the "antiferromagnetic" parameter (without loss of generality it is supposed that the field $\mathbf{H}$ is in the xz plane). Equation (4) is similar to that the equation governing the magnetization dynamics of uniaxial

single-domain ferromagnetic particles and at $\zeta=0$ reduces to it.

Now the reversal time of the magnetization in magnetic nanoparticles is associated with the inverse of the smallest nonvanishing eigenvalue λ_1 of the Fokker-Planck operator L_{FP} in Eq. (2) characterizing the slowest overbarrier relaxation mode and, hence, the long-time behavior of the magnetization. In order to find an asymptotic estimate for $\tau \sim \lambda_1^{-1}$, Brown[9] and Smith and De Rozario[10] adapted to magnetic relaxation an ingenious method originally proposed by Kramers[11] for thermally activated escape of Brownian particles from a potential well. Thus they approximately calculated the reversal time $\tau \sim \lambda_1^{-1}$ for single-domain ferromagnetic particles in the so-called intermediate-to-high damping (IHD) limit ($\alpha \geq 1$). Much later, Klik and Gunther[12] derived the corresponding formula in the very low damping (VLD) limit ($\alpha \ll 1$). Later still for values of damping, in the range $0.01 < \alpha < 1$ (the so-called Kramers turnover regime), Coffey *et al.*[13] have shown that the Mel'nikov-Meshkov formalism[14] for bridging the VLD and IHD Kramers escape rates as a function of the dissipation parameter for point Brownian particles, can be extended to the reversal time of the magnetization of magnetic nanoparticles. The results of Coffey *et al.*[13] agree closely with numerical solutions of the Fokker-Planck Eq. (2), e.g., Refs. 15 and 16, and Langevin dynamics simulations of the magnetization reversal, for example, Refs. 17 and 18, for a variety of magnetocrystalline anisotropy potentials (cubic, biaxial, etc.). The results also have been successfully compared with experiments[19] on the angular variation in the switching field for individual Co and BaFeCoTiO particles justifying the Néel-Brown (in effect, the Kramers) conception of the superparamagnetic relaxation process and emphasizing the vital importance of an accurate determination of the damping dependence of the reversal time of the magnetization.

In the present paper, we use the method of Coffey *et al.*[13] to evaluate the reversal time of the magnetization for *antiferromagnetic* nanoparticles subjected to a dc magnetic field applied at an arbitrary angle ψ to the easy axis of a particle. The particular case of a dc magnetic field parallel to the easy axis, i.e., $\psi=0$, has been considered recently by Raikher *et al.*[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 a high magnetic anisotropy limit. The limiting case $\zeta=0$, corresponding to uniaxial ferromagnetic particles, has been treated in Refs. 15 and 20–23.

II. ASYMPTOTIC FORMULAS FOR THE REVERSAL TIME

The free energy, Eq. (4), has a bistable structure with minima at $\mathbf{n}_1$ and $\mathbf{n}_2$ separated by a potential barrier with a saddle point at $\mathbf{n}_0$. The saddle point is generally in the equatorial region while $\mathbf{n}_1$ and $\mathbf{n}_2$ lie in the north and south polar regions, respectively (see Fig. 1). For $\psi \neq 0$, corresponding to a nonaxially symmetric energyscape, we have a strong intrinsic dependence of the reversal time and other magnetic characteristics (such as the complex magnetic susceptibility)

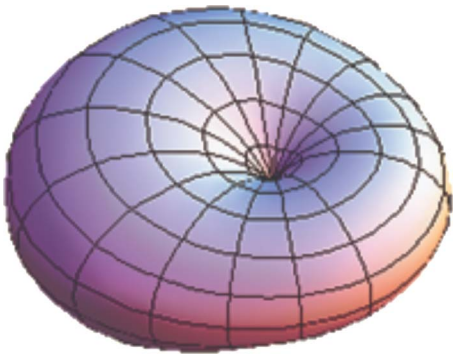

FIG. 1. (Color online) Three-dimensional plot of the potential Eq. (4) for $\sigma=10$, $h=0.15$, $\zeta=0.15$, and $\psi=\pi/3$.

on the damping α which arises from coupling of the longitudinal and transverse relaxation modes. For antiferromagnetic anisotropy, Eq. (4), where the energyscape has two nonequivalent wells with one saddle point, the reversal time τ of the magnetization can be estimated using the approach of Coffey *et al.*[13] as

$$\tau \sim \tau_{\mathrm{IHD}} \frac{A(\alpha S_1 + \alpha S_2)}{A(\alpha S_1)A(\alpha S_2)}. \tag{5}$$

Here

$$\tau_{\mathrm{IHD}} \sim (\Gamma_{12}^{\mathrm{IHD}} + \Gamma_{21}^{\mathrm{IHD}})^{-1} \tag{6}$$

is the reversal time in the IHD limit, $\Gamma_{ij}^{\mathrm{IHD}}$ is the escape rate from the well i to the well j given by Brown's IHD formula[9]

$$\Gamma_{ij}^{\mathrm{IHD}} \sim \frac{\Omega_0 \omega_i}{2\pi\omega_0} e^{-\beta(V_0 - V_i)}, \tag{7}$$

where

$$\omega_i = \frac{\gamma}{\mu}\sqrt{c_1^{(i)} c_2^{(i)}}, \quad \omega_0 = \frac{\gamma}{\mu}\sqrt{-c_1^{(0)} c_2^{(0)}} \tag{8}$$

are the well and saddle angular frequencies, respectively,

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

is the (over) damped saddle angular frequency,

$$c_1^{(k)} = \frac{\partial^2 V}{\partial u_1^{(k)2}}, \quad c_2^{(k)} = \frac{\partial^2 V}{\partial u_2^{(k)2}} \quad (k = 0,1,2)$$

$u_1^{(k)} = \sin\vartheta_k \cos\varphi_k$, $u_2^{(k)} = \sin\vartheta_k \sin\varphi_k$, $u_3^{(k)} = \cos\vartheta_k$ are the direction cosines, $A(\alpha S_i)$ is the depopulation factor defined as

$$A(\alpha S_i) = \exp\left[\frac{1}{\pi}\int_0^\infty \frac{\ln[1 - \exp\{-\alpha S_i (x^2 + 1/4)\}]}{x^2 + 1/4} dx\right], \tag{9}$$

and S_i is the dimensionless action at the saddle-point energy V_0 defined as

$$S_i = \beta \oint_{V(\vartheta,\varphi)=V_0} \left[\sin^2 \vartheta \frac{\partial V}{\partial \cos \vartheta} d\varphi - \frac{1}{\sin^2 \vartheta} \frac{\partial V}{\partial \varphi} d \cos \vartheta \right]. \tag{10}$$

The contour integral in Eq. (10) is taken along the critical energy trajectory (the separatrix) $\vartheta(\varphi)|_{V=V_0}$ on which the magnetization may reverse by passing through the saddle point of the energy V_0. This critical energy trajectory is determined from the equation $V(\vartheta,\varphi)=V_0$. When the energy attains the value V_0, the magnetization may escape the well.

The quantities $c_1^{(k)}$, $c_2^{(k)}$, and V_k are evaluated as follows. First we note the relationship between the direction cosines $u_n^{(k)}$ and $u_m'^{(k)}$ in the basic polar coordinate system P and a polar coordinate system P' with the origin at the stationary point $\mathbf{n}_k$ is

$$u_n^{(k)} = \sum_{m=1}^{3} R_{mn}^{(k)} u_m'^{(k)} \quad (n = 1,2,3), \tag{11}$$

where $R_{mn}^{(k)}$ are the elements of the transformation (rotation) matrix $\mathbf{R}^{(k)}$ relating P and P' which is defined as[20]

$$\mathbf{R}^{(k)} = \begin{pmatrix} \cos \varphi_k \cos \vartheta_k & \sin \varphi_k \cos \vartheta_k & -\sin \vartheta_k \\ -\sin \varphi_k & \cos \varphi_k & 0 \\ \cos \varphi_k \sin \vartheta_k & \sin \varphi_k \sin \vartheta_k & \cos \vartheta_k \end{pmatrix}.$$

$c_1^{(k)}$, $c_2^{(k)}$, and V_k can now be evaluated and are given by

$$\beta c_1^{(k)} = 2\sigma[\cos 2\vartheta_k + h \cos(\vartheta_k - \psi) - 2h^2 \zeta \sigma \cos 2(\vartheta_k - \psi)],$$

$$\beta c_2^{(k)} = 2\sigma[\cos^2 \vartheta_k + h \cos(\vartheta_k - \psi) - 2h^2 \zeta \sigma \cos(\vartheta_k - \psi)^2],$$

$$\beta V_k = \sigma[\sin^2 \vartheta_k - 2h \cos(\vartheta_k - \psi) + 2h^2 \zeta \sigma \cos(\vartheta_k - \psi)^2], \tag{12}$$

where ϑ_k are solutions of equation $\partial_\vartheta V|_{\varphi=0}=0$ listed in Appendix. Here we have assumed without loss of the generality that the external field is in the xz plane. Next, in order to evaluate the actions S_1 and S_2 defined by the contour integrals given by Eq. (10), one must determine the critical trajectories $\vartheta_1(\varphi)|_{V=V_0}$ and $\vartheta_2(\varphi)|_{V=V_0}$. Equations for them can be obtained by solving the trigonometric equation,

$$\sin^2 \vartheta - 2h(\cos \psi \cos \vartheta + \sin \psi \sin \vartheta \cos \varphi) + 2\sigma h^2 \zeta (\cos \psi \cos \vartheta + \sin \psi \sin \vartheta \cos \varphi)^2 = \beta V_0 / \sigma.$$

Hence that S_1 and S_2 can be evaluated. Explicit equations for $c_1^{(k)}$, $c_2^{(k)}$, $V_0 - V_{1,2}$, and $S_{1,2}$ are presented in Appendix.

For the axially symmetric case, $\psi=0$, where the above equations are no longer valid, the reversal time τ may be evaluated using the mean first passage time (MFPT) approach.[24] Here the normalized free energy of the particle, Eq. (4), may be presented in the form of that for a uniaxial particle, viz.,

$$\beta V_{ef}(z) = -\sigma' z^2 - \xi z, \tag{13}$$

where $\sigma' = \sigma - \xi^2 \zeta / 2$ is an effective anisotropy constant and $z=\cos \vartheta$. Thus Eq. (2) becomes a single variable Fokker-Planck equation[26]

$$2\tau_N \frac{\partial W}{\partial t} = \frac{\partial}{\partial z}\left[(1-z^2)\left(\frac{\partial W}{\partial z} + \beta W \frac{\partial V_{ef}}{\partial z} \right) \right].$$

The asymmetric bistable potential V_{ef} has a maximum at $z_{\max} = -\xi / 2\sigma'$ and two nonequivalent minima separated by the barriers $\Delta V_{\pm} = \beta^{-1} \sigma' (1 \pm \xi / 2\sigma')^2$. The reversal time can then be estimated via the MFPT as $\tau^{-1} = (\tau_+^{-1} + \tau_-^{-1})/2$, where τ_+ and τ_- are the MFPTs for transitions from the point domains $z=1$ and $z=-1$, respectively. Thus we have

$$\tau^{-1} = \frac{1}{4\tau_N}\left[\left(\int_{-1}^{z_{\max}} \frac{e^{\beta V_{ef}(z)}}{1-z^2} \int_{-1}^{z} e^{-\beta V_{ef}(z')} dz' dz \right)^{-1} + \left(\int_{z_{\max}}^{1} \frac{e^{\beta V_{ef}(z)}}{1-z^2} \int_{z}^{1} e^{-\beta V_{ef}(z')} dz' dz \right)^{-1} \right]. \tag{14}$$

In the high-barrier approximation, $\sigma' \gg 1$, estimation of Eq. (14) using the steepest descent method yields Brown's high-energy barrier asymptotic formula[8,9] for uniaxial particles in the presence of a dc field an effective anisotropy parameter σ' and the usual field parameter ξ,

$$\tau \sim \frac{\tau_N \sqrt{\pi} e^{\sigma'(1-\xi/2\sigma')^2}}{\sigma'^{3/2}[1-(\xi/2\sigma')^2][1-\xi/2\sigma' + (1+\xi/2\sigma')e^{-2\xi}]}. \tag{15}$$

If the departures from axial symmetry are small, $h \sin \psi < 0.05$, the nonaxially symmetric asymptotic Eq. (5) for the reversal time may be smoothly connected to the axially symmetric results, Eq. (15), by means of suitable bridging integrals. This procedure is described in Ref. 25 for the uniform field transversally applied to the easy axis of the magnetization for a particle with uniaxial anisotropy.

III. RESULTS AND DISCUSSION

In order to assess the asymptotic formulas so obtained, we compare them with numerical solutions of Eq. (2) using matrix continued fractions.[16,26] In this context, the solution of the Fokker-Planck Eq. (2) is first reduced to an infinite hierarchy of differential-recurrence equations for the statistical moments $c_{l,m}(t) = \langle Y_{l,m}[\vartheta(t), \varphi(t)] \rangle$ governing the magnetization relaxation,[26] namely,

$$\frac{d}{dt} c_{l,m}(t) = \sum_{l',m'} d_{l',m',l,m} c_{l,m}(t), \tag{16}$$

where $d_{l',m',l,m}$ are the matrix elements of the Fokker-Planck operator in Eq. (2) and the angular brackets denote the statistical average. A method of derivation of Eq. (16) for arbitrary anisotropy potential is given in Refs. 26 and 27. Equation (16) can be transformed into the tridiagonal vector recurrence equation,[26]

$$\frac{d}{dt} \mathbf{C}_n(t) = \mathbf{Q}_n^- \mathbf{C}_{n-1}(t) + \mathbf{Q}_n \mathbf{C}_n(t) + \mathbf{Q}_n^+ \mathbf{C}_{n+1}(t), \tag{17}$$

where $\mathbf{C}_n(t)$ are the column vectors arranged in an appropriate way from $c_{l,m}(t)$ and $\mathbf{Q}_n^-, \mathbf{Q}_n, \mathbf{Q}_n^+$ are the matrices with elements $d_{l',m',l,m}$. Equation (17) can now be solved using

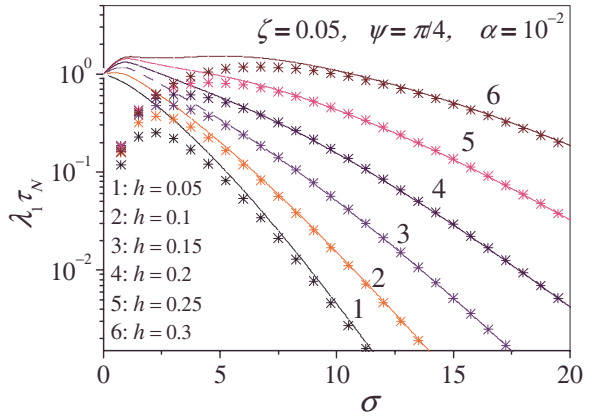

FIG. 2. (Color online) $\lambda_1\tau_N$ vs the anisotropy (or the inverse temperature) parameter σ for α=0.01, ζ=0.05, $\psi=\pi/4$, and various values of the field parameter h. Solid lines: matrix continued fraction solution. Symbols: the universal Eq. (5).

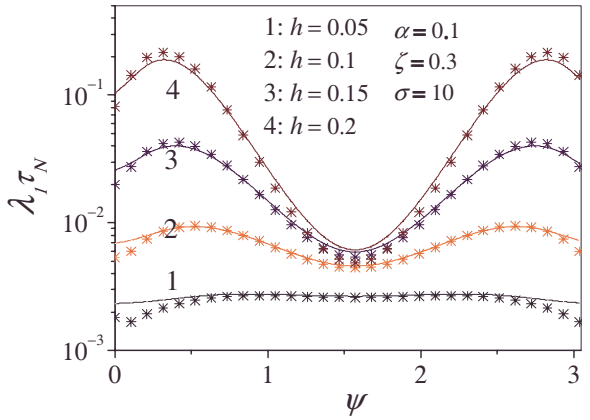

FIG. 4. (Color online) $\lambda_1\tau_N$ vs the oblique angle ψ for ζ=0.3, σ=10, α=0.1, and various values of h. Solid lines: matrix continued fraction solution. Symbols: the universal Eq. (5).

matrix continued fractions (the details of solution for the problem under consideration will be published elsewhere). By solving Eq. (17) we have, in particular, the smallest nonvanishing eigenvalue λ_1 of the Fokker-Planck operator which is associated with the slowest relaxation mode and for sufficiently high potential barriers the asymptotic behavior of λ_1 must correspond to the Kramers escape rate[6,23] and its extension to include all values of the damping so providing a numerical check to the asymptotic Eq. (5) for the reversal time.

By way of illustration, λ_1 calculated with the matrix continued fraction method and τ^{-1} predicted by the universal Eq. (5) as functions of the anisotropy (or the inverse temperature) parameter σ are shown in Figs. 2 and 3 for various values of the model parameters h (external field parameter) and ζ (antiferromagnetic parameter). In Figs. 4 and 5, λ_1 and τ^{-1} are plotted as a function of ψ and ζ, respectively. Figures 3 and 5 demonstrate that changes in the antiferromagnetic parameter ζ significantly affect the relaxation process. Namely, as ζ increases the reversal time decreases, which may be attributed to decrease in the effective anisotropy constant $\sigma'=\sigma-\xi^2\zeta/2$. Thus, the variation in the model parameters affects strongly the reversal time of the magnetization leading to changes in the reversal time τ of several orders of magnitude. Figures 6 and 7 illustrate the damping dependence of λ_1 and τ^{-1} for various values of the external field (h) and antiferromagnetic (ζ) parameters. Here τ_N/τ calculated from the IHD, Eq. (6), is shown for comparison; the dependence of τ_N on α being given by Eq. (3). As seen in Figs. 2–7, in the high barrier limit, $\sigma>5$, and with the nonaxially symmetric condition, $h\sin\psi\geq 0.05$, the analytic Eq. (5) provides a good approximation to the reversal time for a wide range of the model parameters. On the other hand, these figures show that Eq. (5) deviates from the numerical results at high temperatures, $0<\sigma\leq 5$, where the barriers are low and so that as expected our asymptotic solution is no longer valid.

In conclusion, we have evaluated the reversal time of the magnetization τ for antiferromagnetic nanoparticles subjected to a uniform external field $\mathbf{H}_0$ applied at an arbitrary angle ψ to the easy axis of the particle using the model suggested by Raikher *et al.*[4] Moreover, we have shown that the simple universal Eq. (5) provides an accurate description of τ for antiferromagnetic nanoparticles in the low-temperature limit for all values of damping including the

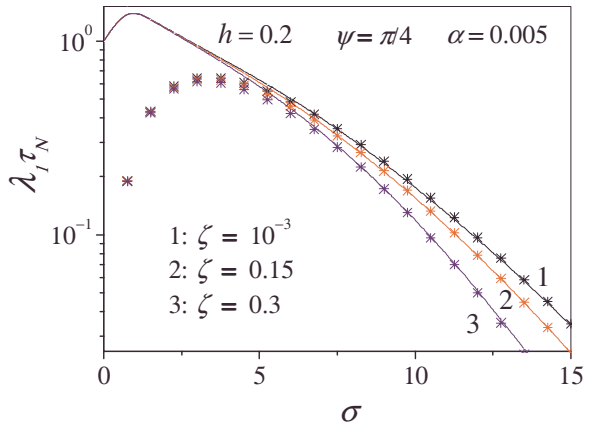

FIG. 3. (Color online) $\lambda_1\tau_N$ vs σ for h=0.2, $\psi=\pi/4$, α=0.005, and various values of the antiferromagnetic parameter ζ. Solid lines: matrix continued fraction solution. Symbols: the universal Eq. (5).

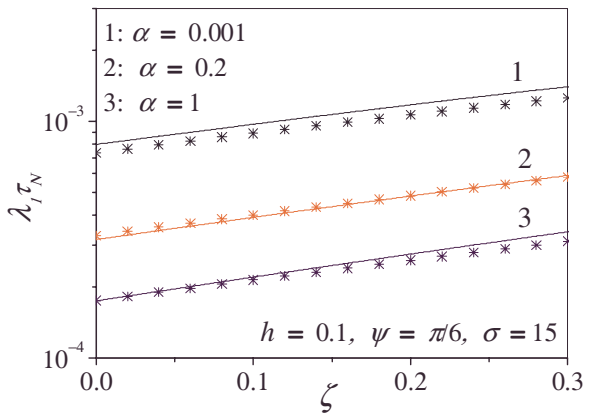

FIG. 5. (Color online) $\lambda_1\tau_N$ vs the antiferromagnetic parameter ζ for h=0.1, $\psi=\pi/6$, σ=15, and various values of α. Solid lines: matrix continued fraction solution. Symbols: the universal Eq. (5).

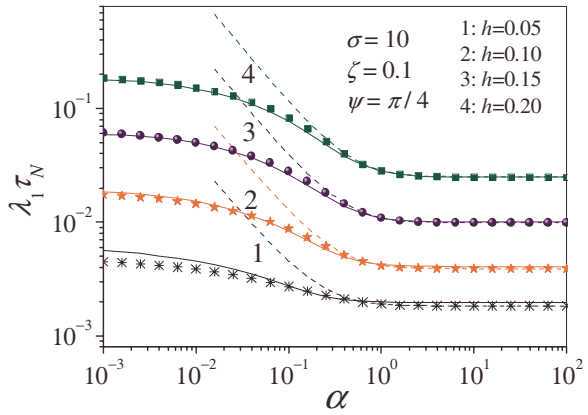

FIG. 6. (Color online) $\lambda_1\tau_N$ vs the damping parameter α for $\zeta=0.1$, $\psi=\pi/4$, $\sigma=10$, and various values of the field parameter $h=0.05$, 0.10, 0.15, and 0.20. Solid lines: matrix continued fraction solution. Symbols: the universal Eq. (5). Dashed lines: the IHD Eq. (6).

IHD ($\alpha \geq 1$), turnover ($\alpha \sim 1$), and VLD ($\alpha \ll 1$) damping regimes. Equation (5) can also be used to estimate others physical parameters such as angular and temperature variations in the switching field of an individual particle and the linear and nonlinear dynamic susceptibilities. Furthermore, our results can be used to study stochastic resonance[28] and dynamic hysteresis[29] in antiferromagnetic nanoparticles which may differ essentially from those in fine ferromagnetic particles.[30–33] For the particular case, $\psi=0$, our results entirely agree with those reported by Raikher *et al.*[4] We remark that throughout the calculations it was supposed that all particles are identical. In order to account for polydispersity of the particles in a real sample, one must also average τ over appropriate distribution functions (e.g., over that of particle volumes). Furthermore, the surface effects and interactions between particles have been ignored in the analysis. These effects present formidable mathematical difficulties in the formulation of a realistic theoretical model. However, they can, in principle, be controlled in the interpretation of experimental data by varying the particles' volumes and their concentration.

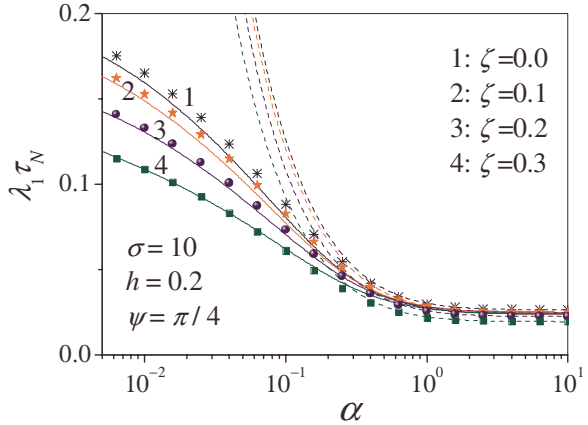

FIG. 7. (Color online) $\lambda_1\tau_N$ vs the damping parameter α for $h=0.2$, $\psi=\pi/4$, $\sigma=10$, and various values of the antiferromagnetic parameter $\zeta=0$, 0.1, 0.2, and 0.3. Solid lines: matrix continued fraction solution. Symbols: the universal Eq. (5). Dashed lines: the IHD Eq. (6).

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APPENDIX: EVALUATION OF THE PARAMETERS IN EQ. (5)

If $x=\cos\vartheta$, the equation $\partial_\vartheta V=0\big|_{\varphi=0}$ determining the minima and the saddle point takes the following form,

$$2x\sqrt{1-x^2}-2h(\cos\psi\sqrt{1-x^2}-x\sin\psi)\times[1-2h\zeta\sigma(x\cos\psi+\sin\psi\sqrt{1-x^2})]=0. \quad \text{(A1)}$$

The roots of this equation, namely, $x_0=\cos\vartheta_0$ and $x_{1,2}=\cos\vartheta_{1,2}$, corresponding to the saddle point and minima of the free energy density, respectively, can be written as converging Taylor series for any desired order of the field parameter h, viz.,

$$\cos\vartheta_0=-\cos\psi h-\frac{1}{2}(1-2\zeta\sigma)\sin 2\psi h^2+\cos\psi[\zeta\sigma(1-3\cos 2\psi)-\sin^2\psi]h^3-\frac{1}{8}[3+2\zeta\sigma-(1-26\zeta\sigma+16\zeta^2\sigma^2)\cos 2\psi]\times\sin 2\psi h^4+\cdots, \quad \text{(A2)}$$

$$\cos\vartheta_{1,2}=\pm 1\mp\frac{1}{2}h^2\sin^2\psi+h^3(1+2\zeta\sigma)\sin^2\psi\cos\psi\mp\frac{1}{16}h^4\sin^2\psi\{13+16\zeta\sigma(2+\zeta\sigma)+[11+16\zeta\sigma(4+\zeta\sigma)]\cos 2\psi\}+\cdots. \quad \text{(A3)}$$

The corresponding Taylor series for $\beta(V_0-V_{1,2})$, $\beta c_1^{(0)}$, $\beta c_2^{(0)}$, $\omega_{1,2}$, and ω_0 can be readily obtained from Eqs. (8), (12), (A2), and (A3) and are given by

$$\beta(V_0-V_{1,2})=\sigma\Big\{1\pm 2h(\cos\psi\mp\sin\psi)+(1-2\zeta\sigma\cos 2\psi)h^2+\cos\psi\sin\psi\times[(1-4\zeta\sigma)\cos\psi\mp(4\zeta\sigma+1)\sin\psi]h^3+\frac{1}{2}[4\zeta\sigma\cos 2\psi+(1+4\zeta^2\sigma^2)\sin^2 2\psi]h^4+\cdots\Big\}, \quad \text{(A4)}$$

$$\omega_{1,2} = \frac{2\sigma\gamma}{M_S}\left\{1 \pm h\cos\psi - \frac{1}{4}[1 + 2\sigma\zeta(6\sigma\zeta - 1)\cos 2\psi]h^2 \pm \frac{1}{2}(3 - 4\sigma\zeta)\cos\psi\sin^2\psi h^3 + \frac{1}{16}[(52\sigma^2\zeta^2 - 44\sigma\zeta - 19)\cos 2\psi + 4\sigma\zeta(11\sigma\zeta - 1) - 21]\sin^2\psi h^4 + \cdots\right\}, \tag{A5}$$

$$\omega_0 = \frac{2\sigma\gamma}{M_S}\left\{1 - \frac{1}{2}h\sin\psi + \frac{1}{16}[8\zeta\sigma - 3 - (1 + 8\zeta\sigma)\cos 2\psi]h^2 - \frac{1}{32}\{[19 + 40\zeta\sigma + 32\zeta^2\sigma^2 + (17 + 56\zeta\sigma + 32\zeta^2\sigma^2)\cos 2\psi]\sin\psi\}h^3 + \frac{1}{1024}[-351 - 336\zeta\sigma + 832\zeta^2\sigma^2 - 4(7 + 176\zeta\sigma - 64\zeta^2\sigma^2)\cos 2\psi + (283 + 16\zeta\sigma - 1088\zeta^2\sigma^2)\cos 4\psi]h^4 + \cdots\right\}, \tag{A6}$$

$$\beta c_1^{(0)}/\sigma = -2 + 2h\sin\psi + [1 + (1 + 4\zeta\sigma)\cos 2\psi]h^2 + (5 + 4\zeta\sigma)\cos^2\psi\sin\psi h^3 + 4\cos^2\psi(1 - \zeta\sigma - 2\zeta^2\sigma^2)(1 - \cos 2\psi)h^4 + \cdots, \tag{A7}$$

$$\beta c_2^{(0)}/\sigma = 2h\sin\psi + (1 + 4\zeta\sigma)\cos^2\psi\sin\psi h^3 - (2\zeta^2\sigma^2 + \zeta\sigma - 1/2)\sin^2 2\psi h^4 + \cdots. \tag{A8}$$

The critical trajectories $\vartheta_1(\varphi)|_{V=V_0}$ and $\vartheta_2(\varphi)|_{V=V_0}$ can also be written as Taylor series in h, viz.,

$$\cos\vartheta_{1,2}(\varphi)|_{V=V_0} = -h\cos\psi\{1 + h(1 - 2\zeta\sigma)\sin\psi\cos\varphi + h^2[2\zeta\sigma\cos^2\psi + (1 + 2\zeta\sigma - 6\zeta\sigma\cos\varphi)\cos\varphi\sin^2\psi]\} \mp 2\sin\left(\frac{\varphi}{2}\right)\sqrt{h\sin\psi}\left\{1 - \frac{h}{2}[\zeta\sigma + (\zeta\sigma - 1)\cos\varphi]\sin\psi + \frac{h^2}{16}[4(8\zeta\sigma - 1)\cos^2\psi + \{1 - \zeta\sigma(10 + 3\zeta\sigma) + 4[1 - \zeta\sigma(1 + \zeta\sigma)]\cos\varphi + [1 - \zeta\sigma(10 + \zeta\sigma)]\cos 2\varphi\}\sin^2\psi]\right\} + \cdots \tag{A9}$$

so that one can evaluate analytically the actions S_1 and S_2 as

$$S_{1,2} = 16\sigma\sqrt{h\sin\psi}\left\{1 - \frac{1}{6}(13 + 2\zeta\sigma)h\sin\psi + \frac{h^2}{240}\{15 + 4\zeta\sigma(61 - 3\zeta\sigma) + [4\zeta\sigma(3\zeta\sigma - 61) - 315]\cos 2\psi\} + \frac{h^3}{840}\{1185 + 4\zeta\sigma(47\zeta\sigma - 735) + [1335 + 4\zeta\sigma(93\zeta\sigma - 805)]\cos 2\psi\}\sin\psi\right\} \pm 2\pi\sigma h^2\sin 2\psi\{-4 + (3 + 2\zeta\sigma)h\sin\psi + [1 - 2\zeta\sigma + (1 - 14\zeta\sigma)\cos 2\psi]h^2\} + \cdots. \tag{A10}$$

Having derived explicit equations for $\beta(V_0 - V_{1,2})$, $\beta c_1^{(0)}$, $\beta c_2^{(0)}$, $\omega_{1,2}$, ω_0, S_1, and S_2, one can estimate the reversal time τ from Eq. (5).

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