Damping dependence of the reversal time of the magnetization of single-domain ferromagnetic particles for the Néel-Brown model: Langevin dynamics simulations versus analytic results

Yuri P. Kalmykov,[1] William T. Coffey,[2] Unai Atxitia,[3] Oksana Chubykalo-Fesenko,[3] Pierre-Michel Déjardin,[1] and Roy W. Chantrell[4]

[1]*Laboratoire de Mathématiques, Physique et Systèmes, Université de Perpignan Via Domitia, 52, Avenue Paul Alduy, 66860 Perpignan Cedex, France*

[2]*Department of Electronic and Electrical Engineering, Trinity College, Dublin 2, Ireland*

[3]*Instituto de Ciencia de Materiales de Madrid, CSIC, c.Sor Juana Inés de la Cruz, 3, 28049 Madrid, Spain*

[4]*Department of Physics, The Computational Magnetism Group, University of York, York YO10 5DD, United Kingdom*

(Received 4 March 2010; revised manuscript received 11 May 2010; published 13 July 2010)

The damping dependence of the thermally activated reversal time of the magnetization of noninteracting uniaxial single-domain ferromagnetic particles is determined using Langevin dynamics simulations and the analytic Néel-Brown theory with the latter given both in the form of the exact matrix-continued fraction solution of the governing Fokker-Planck equation and its accompanying asymptotes for the escape rate. The reversal time from Langevin dynamics simulations is extremely sensitive to the initial and switching conditions used. Thus if the latter are chosen inappropriately the simulation result may markedly disagree with the analytic one particularly for low damping, where the precessional effects dominate, so that complete agreement can only be obtained by correctly choosing these conditions.

DOI: 10.1103/PhysRevB.82.024412 PACS number(s): 75.60.Jk, 75.50.Tt, 76.20.+q, 05.40.−a

I. INTRODUCTION

Fine single-domain ferromagnetic particles are characterized by thermal instability of the magnetization $\mathbf{M}(t)$ resulting in superparamagnetism because each behaves in a magnetic sense as a giant Langevin paramagnet. The initial analytic treatment of the thermal fluctuations due to Néel[1] based on classical transition-state theory (TST) was further developed by Brown[2] and is consequently known as the Néel-Brown theory. This treatment utilizes the classical theory of Brownian motion (which unlike TST accounts for the departure from thermal equilibrium due to the energy interchange between a particle and its heat bath) with the Landau-Lifshitz-Gilbert (LLG) equation augmented by white-noise fields as Langevin equation. This equation is then used to derive the particular Fokker-Planck equation (FPE) governing the time evolution of the probability density function W of magnetization orientations on a sphere of radius M_s. Here M_s is the saturation magnetization assumed to be constant so that the only variable is the orientation of $\mathbf{M}$ and the relevant FPE is[2,3]

$$2\tau_N \frac{\partial W}{\partial t} = \frac{\beta}{\alpha}\mathbf{n}\cdot(\nabla V \times \nabla W) + \nabla\cdot(\nabla W + \beta W \nabla V). \quad (1)$$

In Eq. (1), $\nabla = \partial/\partial\mathbf{n}$ is the gradient operator on the unit sphere, $\mathbf{n}$ is a unit vector along $\mathbf{M}$, V is the free-energy density comprising the *nonseparable* Hamiltonian of the anisotropy and Zeeman energy densities, $\beta = v/(kT)$, v is the volume of the single-domain ferromagnetic particle, k is Boltzmann's constant, T is the absolute temperature, α is the dimensionless damping constant, $\tau_N = \tau_0(\alpha + \alpha^{-1})$, $\tau_0 = \beta M_s/(2\gamma)$ is the characteristic free diffusion time of $\mathbf{M}(t)$, and γ is the gyromagnetic ratio.

One of the most important physical parameters is the magnetization switching (or reversal) time τ due to thermal agitation over the internal magnetocrystalline energy barrier of the particle. In the Néel-Brown model,[2,3] the reversal time may be calculated numerically for a given anisotropy potential as the inverse of the smallest nonvanishing eigenvalue λ_1 of the Fokker-Planck operator in Eq. (1). However, a practical disadvantage of the λ_1^{-1} method is that it is, in principle, impossible to write λ_1 in closed form since it is the smallest nonvanishing root of the secular equation of the system of differential recurrence relations for the statistical moments resulting from separation of the variables. Accordingly much effort has been expended in finding analytic approximations for λ_1. Thus the high barrier (low-temperatures) asymptotes, which is the case of greatest interest, are obtained by extending the Kramers theory[4,5] of thermally activated escape of particles over a potential barrier to the magnetization reversal. The Kramers theory[4] was originally given for point particles of one degree of freedom coupled to a heat bath governed by a FPE, where the position and the momentum constitute the canonical variables and known as Klein-Kramers equation. However, the magnetization reversal problem is, in general, characterized by the nonseparable Hamiltonian[3,5] of the anisotropy and Zeeman energies so that two degrees of freedom are involved, namely, the polar and azimuthal angles (ϑ, φ). Hence modifications to the original Kramers treatment are necessary. A particular simple case of the magnetization Kramers problem first noted by Brown[2] is axial symmetry. Then the Hamiltonian is simply a function of ϑ hence it is possible[2] to find a high barrier asymptotic formula for λ_1 which is valid for all values of the coupling to the heat bath α all that is necessary being a knowledge of the stationary points of the potential. In addition, for the simplest uniaxial anisotropy potential of the form $\sin^2\vartheta$ it is also possible to write down a formula valid for all barrier heights.[22] The situation for nonaxially symmetric potentials is, however, much more complicated. The Kramers analysis for such potentials having been initiated by Smith and de Rozario[6] was continued by Brown[3] who formally extended

the so-called Kramers intermediate-to-high damping (IHD) escape rate[4,5] to the magnetization by evaluating λ_1 in terms of the escape rate $\Gamma_{ij}^{\mathrm{IHD}}$ from well i to well j as

$$\lambda_1 \sim \Gamma_{ij}^{\mathrm{IHD}} = \Gamma_{ij}^{\mathrm{TST}} \Omega_0(\alpha)/\omega_0 \tag{2}$$

with $\Gamma_{ij}^{\mathrm{TST}}$ as the escape rate for TST as applied to the magnetization, namely,

$$\Gamma_{ij}^{\mathrm{TST}} = (\omega_i/2\pi)e^{-\Delta V}, \tag{3}$$

where $\Delta V = \beta(V_0 - V_i)$ is the dimensionless barrier height, $\omega_i = \gamma\sqrt{c_1^{(i)}c_2^{(i)}}/M_s$ and $\omega_0 = \gamma\sqrt{-c_1^{(0)}c_2^{(0)}}/M_s$ are the well and saddle angular frequencies, respectively, and

$$\Omega_0(\alpha) = \frac{\beta}{4\tau_0(\alpha+\alpha^{-1})}\left[\sqrt{(c_2^{(0)}-c_1^{(0)})^2 - 4\alpha^{-2}c_1^{(0)}c_2^{(0)}} - c_1^{(0)} - c_2^{(0)}\right]$$

is the damped saddle angular frequency. We emphasize that Eq. (2) is simply a special case of Langer's extension[7] of the Kramers IHD escape rate to many degrees of freedom and nonseparable Hamiltonians so yielding the upper bound of the escape rate. Clearly for vanishing damping, $\alpha \to 0$, the IHD escape rate $\Gamma_{ij}^{\mathrm{IHD}}$ from Eq. (2) reduces to the TST escape rate $\Gamma_{ij}^{\mathrm{TST}}$, which is obviously independent of α. However this is not the *true* VLD limit or *energy-controlled diffusion*, where the energy loss per cycle of the almost periodic motion of the magnetization on the saddle-point energy (escape) trajectory is much less than the thermal energy, as noted by Klik and Gunther.[8] Rather, it comprises the *intermediate damping limit* corresponding to Néel's TST result.[1] Recognizing this Klik and Gunther[8] derived the *correct* VLD magnetization Kramers escape rate $\Gamma_{ij}^{\mathrm{VLD}}$, viz.,

$$\Gamma_{ij}^{\mathrm{VLD}} \sim \alpha S_i \Gamma_{ij}^{\mathrm{TST}}, \tag{4}$$

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

$$S_i = \beta \oint_{V=V_0} (1-z^2)\frac{\partial V}{\partial z}d\varphi - \frac{1}{1-z^2}\frac{\partial V}{\partial \varphi}dz, \tag{5}$$

and $z = \cos\vartheta$. The conditions of applicability of these IHD and VLD solutions for superparamagnets are defined by $\alpha \geq 1$ and $\alpha \ll 1$, respectively. However, experimental values of α usually lie in the Kramers turnover region characterized by $10^{-2} \leq \alpha < 1$. Hence, Coffey *et al.*[5,9] have extended the Mel'nikov-Meshkov formalism[10] connecting VLD and IHD escape rates for point particles, to describe the relaxation time of the magnetization. Thus they obtained for the escape rate Γ_{ij} from a single well over one saddle point

$$\Gamma_{ij} = A(\alpha S_i)\Gamma_{ij}^{\mathrm{IHD}} = A(\alpha S_i)\frac{\omega_i \Omega_0(\alpha)}{2\pi\omega_0}e^{-\Delta V}, \tag{6}$$

where the depopulation factor A is

$$A(\delta) = \exp\left[\frac{1}{\pi}\int_0^\infty \frac{\ln\{1-\exp[-\delta(\lambda^2+1/4)]\}}{\lambda^2+1/4}d\lambda\right].$$

The contour integral in Eq. (5) is taken along the critical-energy trajectory or separatrix $\vartheta(\varphi)|_{V=V_0}$ on which the magnetization may reverse by passing through the saddle point(s) of the energy density V_0. Equation (6) agrees closely with the numerical solution for the reversal time obtained via the FPE [Eq. (1)] for all α, e.g.[11,12] It also agrees with a number of computer simulations[13–15] and with experiments[16] emphasizing the vital importance of an accurate determination of the damping dependence of the escape-rate prefactor $A(\alpha S_i)\Omega_0(\alpha)\omega_i/(2\pi\omega_0)$ in Eq. (6).

The above considerations concerning the damping dependence of the escape rate are of the upmost importance in both Monte Carlo (MC) and Langevin dynamics simulations of the reversal time of the magnetization of fine particles.[13–15,17–21] In analyzing the results of such simulations, the value of the analytical solutions of the Néel-Brown theory for λ_1^{-1} provide rigorous benchmark solutions with which they must comply. However, certain simulations[17–21] pertaining to that theory seem to predict results for the reversal time at variance with it, a question which requires detailed examination, the explanation of which is the prime objective of this paper. Indeed, simulated and analytical estimations of the relaxation time for uniaxial particles sometimes differ by more than one order of magnitude[17] and the reason for such a pronounced difference has hitherto remained profoundly unclear. Furthermore, in Ref. 21, numerical estimates of the switching time were obtained by using the FPE to link the MC and the Langevin micromagnetic schemes, both for noninteracting as well as interacting arrays of fine particles. Moreover, close numerical convergence (hitherto not obtained[19,20]) is claimed between the MC method and Langevin dynamics simulation results. Authors of Ref. 21 also claimed that their Metropolis MC method is accurate for a large range of damping factors α, unlike previous time-quantified MC methods[19,20] which fail for small α, where the precessional motion dominates. However, these simulated results[21] do not reproduce the known Kramers-Brown *asymptotic* solutions for the reversal time at low damping. In view of the foregoing discrepancy, we summarize the conditions allowing one to derive asymptotic formulas for the escape rate from the FPE via the Kramers method and we demonstrate that if these conditions are *systematically* applied in the computer simulations then they too can accurately reproduce the analytic asymptotes. The comparison between analytical and simulation approaches will be illustrated via a single-domain ferromagnetic particle possessing uniaxial anisotropy with a uniform field applied at an angle to the anisotropy axis.

II. LANGEVIN DYNAMICS SIMULATIONS

Langevin dynamics simulations in micromagnetics having been originally introduced by Lyberatos and Chantrell[23] were subsequently further developed by many authors[17,18,24–30] (for a review see Ref. 26 and references therein). This development followed the seminal work of Brown.[2,3] He, as mentioned above, included thermal fluctuations in the dynamics of an ensemble of noninteracting macrospins in order to describe the deviations from the average trajectory and so formally introduced random fields in the LLG for the time evolution of $\mathbf{M}(t)$ which then becomes the Langevin equation of

the process. These thermal fields were supposed uncorrelated both in space and time, and so were represented by Gaussian white noise allowing one to construct a FPE. Brown also showed how to evaluate the spectral density of the thermal fields following Einstein's method[22] by using the fluctuation-dissipation theorem and requiring the equilibrium distribution function of the orientations of the magnetization $\mathbf{M}(t)$ to coincide with the Boltzmann distribution. The concept of a fluctuating thermal field was also generalized to interacting particles,[24,27,31] hastening the development of thermal micromagnetics.

Following the standard approach of micromagnetics, we write (utilizing the LLG) the Langevin equation for the dynamics of the magnetization vector $\mathbf{M}$ for the particular case of a uniaxial single-domain particle in the presence of a dc external magnetic field $\mathbf{H}$ applied at an arbitrary angle ψ to the easy axis as

$$\frac{d\mathbf{M}}{dt} = \frac{\gamma}{1+\alpha^2}[\mathbf{M}\times(\mathbf{H}_{eff}+\mathbf{h})] + \frac{\alpha\gamma M_s^{-1}}{1+\alpha^2}\{[\mathbf{M}\times(\mathbf{H}_{eff}+\mathbf{h})]\times\mathbf{M}\}, \tag{7}$$

where $\mathbf{H}_{eff}=-\partial_{\mathbf{M}}V=\mathbf{H}+H_K\cos\vartheta\mathbf{e}_z$, $H_K=2K/M_s$, K is the anisotropy constant, $\mathbf{e}_z$ is a unit vector along the z axis, $\mathbf{u}=\mathbf{M}/M_s$, and the free-energy density V is

$$V(\vartheta,\varphi) = \sigma\beta^{-1}[\sin^2\vartheta - 2h(\cos\psi\cos\vartheta + \sin\psi\sin\vartheta\cos\varphi)]. \tag{8}$$

Here $\sigma=\beta K$ is the dimensionless barrier height parameter, $h=\xi/(2\sigma)$ is the applied field parameter, and $\xi=\beta M_s H$. The thermal field $\mathbf{h}$ has the white-noise properties

$$\langle h_i(t)\rangle = 0, \quad \langle h_i(t)h_j(t)\rangle = \frac{2\alpha}{\gamma\beta M_s}\delta_{ij}\delta(t-t'), \quad i=x,y,z$$

meaning that the $h_i(t)$ components are statistically independent and that $h_i(t)$ and $h_j(t)$ are uncorrelated at very short times, i.e., much shorter than the time of a single precession (δ_{ij} is Kronecker's symbol). Now in order to yield the Boltzmann equilibrium distribution, the Langevin Eq. (7) should be interpreted as a Stratonovitch vector stochastic differential equation.[24] This is accomplished by a suitable choice of the numerical integration scheme here that of Heun.[24] The Heun scheme is stable and is in accordance with the Stratonovich stochastic calculus.[26] We remark that several authors[29,30] have argued that even simpler integration schemes, e.g., the Runge-Kutta method, would reproduce the correct Boltzmann equilibrium distribution, if the magnetization vector is renormalized at each time step.

Now the standard method of simulating the reversal time τ using Langevin dynamics is simply to average it over many trajectory realizations. However, this method contains several arbitrary assumptions. First of all in choosing the initial conditions, it is customary to take them as the *same* for all trajectories, for example, starting with all trajectories in an equilibrium nonthermal magnetization minimum. Second, in choosing the switching condition, it is supposed that the particle magnetization has switched if $m_z<m_0$, where m_0 is a characteristic value. However, if m_0 is taken as the exact transition (saddle) point, then the possibility that the magnetization may *revert* to the original minimum should be taken into account. In the symmetric case, imposition of this switching condition consequently yields a switching time approximately two times smaller than if one imposed the condition close to a minimum. In the general case, using different values of m_0 can yield results deviating from each other by a factor between 1 and 2 *in the* IHD *regime*.[32] However, for low damping, the deviation can be much more pronounced due to *precessional* effects. The axially symmetric case, when the applied field is parallel to the anisotropy axis is, however, insensitive to the conditions discussed above.

The reversal time τ can also be calculated by solving the Langevin Eq. (7) (or its accompanying FPE) analytically for λ_1 using matrix-continued fractions (MCF) (Refs. 22, 33, and 34) (see appendix for details). Here we shall also use this independent method for comparison.

III. COMPARISON WITH ANALYTIC RESULTS FOR A UNIAXIAL PARTICLE

The free-energy per unit volume V, Eq. (8), has a bistable structure with two 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 north and south polar regions, respectively. For some critical applied field value $h_c(\psi)=(\cos^{2/3}\psi+\sin^{2/3}\psi)^{-3/2}$ the potential [Eq. (8)] loses its bistable character so that the second minimum becomes a point of inflexion. The corresponding universal (Mel'nikov) formula for the switching time τ is given by[5]

$$\tau \approx \tau_{\mathrm{IHD}}\frac{A(\alpha S_1+\alpha S_2)}{A(\alpha S_1)A(\alpha S_2)}, \tag{9}$$

where the magnetization reversal time in the IHD limit is given by

$$\tau_{\mathrm{IHD}} = (\Gamma_{12}^{\mathrm{IHD}}+\Gamma_{21}^{\mathrm{IHD}})^{-1} \tag{10}$$

and the actions S_1 and S_2 are given explicitly in Ref. 11. The particular form of the depopulation factor in Eq. (9) arises because both wells are involved in the relaxation process. We emphasize that in the derivation of Eq. (9) it is assumed that the potential is nonaxially symmetric. If the departures from axial symmetry become small the nonaxially symmetric asymptotic formulas for the escape rate obtained by the method of steepest descents may be smoothly connected to the axially symmetric results by means of suitable bridging integrals. Such a procedure is described, e.g., in Refs. 5 and 35 for the particular case of a uniform field transversally applied to the easy axis of the magnetization for a particle with uniaxial anisotropy. We remark that for the axially symmetric case $\psi=0$, i.e., the dc field is parallel to the easy axis of the particle, so that τ is then given by Brown's asymptotic formula[1]

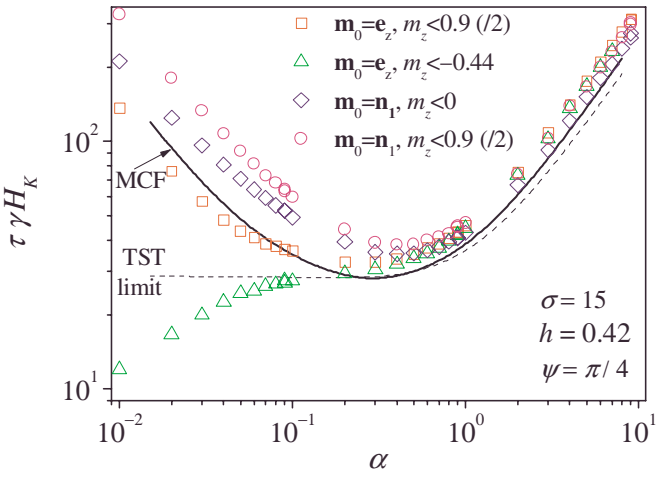

FIG. 1. (Color online) $\tau\gamma H_K$ vs α for $\sigma=15$, $h=0.42$, and $\psi=\pi/4$. Solid line: MCF solution of the Landau-Lifshitz-Gilbert equation. Symbols: simulation results using different initial and switching conditions indicated in the figure. Dashed line: τ_{IHD} given by Eq. (10). The switching time is divided by two for the switching condition $m_z<-0.9$.

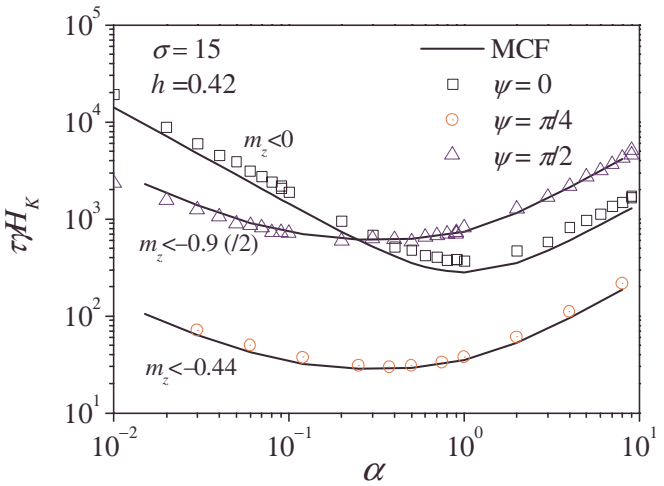

FIG. 2. (Color online) $\tau\gamma H_K$ vs α for $\sigma=15$, $h=0.42$, and various values of $\psi=0$, $\pi/4$, and $\pi/2$, Solid lines: MCF solution. Symbols: simulation results starting with initial conditions distributed according to the Boltzmann distribution around the magnetization minimum. The switching condition is indicated for each curve and the switching time is divided by two for the switching condition $m_z<-0.9$.

$$\tau \sim \tau_0(\alpha+\alpha^{-1})\frac{\sigma^{-3/2}\sqrt{\pi}}{1-h^2}[(1-h)e^{-\sigma(1-h)^2} + (1+h)e^{-\sigma(1+h)^2}]^{-1}. \quad (11)$$

We also emphasize the difference between the (overall) reversal time of the magnetization τ and the inverse individual escape rates Γ_{ij}. In general, both depend on the energyscape as well as the damping regime, however, they can differ considerably from each other. For example, (i) for a potential with two equivalent wells 1 and 2 and one saddle point, $\tau \approx (\Gamma_{12}^{\mathrm{VLD}})^{-1}$ for $\alpha \ll 1$ and $\tau \approx (2\Gamma_{12}^{\mathrm{IHD}})^{-1}$ for $\alpha \geq 1$; (ii) for a potential with two strongly nonequivalent wells ($\Gamma_{12} \gg \Gamma_{21}$) and one saddle point, $\tau \approx (\Gamma_{12}^{\mathrm{VLD}})^{-1}$ for $\alpha \ll 1$ and $\tau \approx (\Gamma_{12}^{\mathrm{IHD}})^{-1}$ for $\alpha \geq 1$, where $\Gamma_{12}^{\mathrm{IHD}}$ is the escape rate from the shallow well 1.

Comparison of the results of calculation of the switching time from the Coffey *et al.* universal asymptotic Eq. (9), the exact matrix-continued fraction solution[22,33] (both based on the Néel-Brown theory), and numerical Langevin simulations are shown in Fig. 1 for various initial and switching conditions. Clearly for IHD damping, $\alpha > 1$, both analytic and numerical simulation methods yield very similar results. However, for *low damping*, the switching time τ predicted by the numerical Langevin simulations can deviate substantially from the universal turnover formula in Eq. (9) and may even *lie under* the lower bound for τ predicted by the TST theory. Indeed, the results for low damping differ not only quantitatively but also qualitatively. Clearly, the particular choice of initial conditions changes completely the low-damping behavior. For example, the switching time for low damping is even smaller than the TST limit, if one starts with the initial condition leading to a strong precession, where switching can occur dynamically without crossing the saddle point. The particular choice of the switching condition also plays an important role. All the data in Fig. 1 represent significant deviations from the expected analytical asymptote.

The contradictions which are simply artifacts of the initial and switching conditions used in the simulations may be explained (cf. our introduction) as follows. In IHD damping the distribution function is almost everywhere the Boltzmann distribution which holds in the depths of the well and only very near the barrier does the distribution function slightly deviate from the equilibrium distribution due to the slow draining of particles across the barrier. However for $\alpha \ll 1$, the damping is so small *that it renders the assumption that the magnetization approaching the saddle region from the depths of the well has the Boltzmann distribution invalid.*[5,35] *Hence, under these damping conditions, extreme care must be taken in simulations*, particularly in the choice of conditions in the well and at the saddle point. In order to illustrate this, we present amended results in Fig. 2, this time *drawing the initial conditions from the correct distribution which in this instance is the Boltzmann distribution about the minimum position*. The foregoing amendment yields full agreement with the expected theoretical value in contrast to the results in Fig. 1. Interestingly enough, the MC scheme presented in Ref. 19 ignores precession hence, precessional switching responsible for the decrease in the switching time at low damping is impossible in this instance (see Fig. 1). The so-called "corrected MC scheme," taking into account the precessional effects and reported in Ref. 21 produced results in agreement with the Langevin dynamics, however, in disagreement with the theoretical asymptotic values only because of the incorrect choice of initial conditions.

In Figs. 1 and 2 we also present the switching time obtained by the MCF method. The actual Boltzmann distribution at equilibrium is implicit in the derivation of the MCF method based on the separation of the variables in the FPE, consequently, the results of that method are in perfect agreement with the improved Langevin dynamics simulations.

IV. CONCLUSION

In the Kramers escape rate picture, the behavior of the switching time τ can be explained as follows. That time as a function of the barrier height parameter σ for large σ is approximately Arrhenius type and arises from an *equilibrium* property of the system (namely, the Boltzmann distribution at the bottom of the well). On the other hand, the damping dependence of τ is due to *nonequilibrium* (dynamical) properties of the system and so is contained in the prefactor in Eq. (6), the *detailed* nature of which depends on the behavior of the energy distribution function at the saddle points of the magnetocrystalline anisotropy. For example, in the IHD regime the distribution function at the saddle point is almost the Boltzmann distribution, while in the VLD regime, the region of nonequilibrium runs deep into the well so that the Boltzmann distribution holds only very near the minimum. The generalization of the Mel'nikov-Meshkov approach[10] to the magnetization escape rate given by Coffey *et al.*[5,9] correctly accounts for the behavior of the distribution function at the saddle point for all values of the damping allowing one to evaluate the damping dependence of the switching time providing a rigorous benchmark solution with which the computer simulation must comply. One may conclude that the numerical values of the switching time in the low-damping regime crucially depend on the initial and switching conditions so that drawing the initial conditions from the Boltzmann distribution near the bottom of the well is absolutely essential for the simulations to be consistent with the Néel-Brown theory. We believe that most of the numerical simulations in which large deviations from the analytical asymptote have been reported were performed under conditions incompatible with the assumptions underlying the Néel-Brown theory. In this paper, we have presented a rigorous procedure allowing one to comply with these conditions leading to agreement between the results yielded by both methods.

ACKNOWLEDGMENTS

The work of U.A. and O.C.-F. was supported by the Spanish grants MAT2007-66719-C03-01 and CS2008-023.

APPENDIX: MATRIX-CONTINUED FRACTION SOLUTION

In order to calculate the reversal time τ, one can use the matrix-continued fraction approach developed in Ref. 34. The solution of the stochastic Langevin Eq. (7) or the corresponding Fokker-Planck Eq. (1) reduces to the solution of an infinite hierarchy of differential-recurrence equations for the statistical moments $c_{l,m}(t)=\langle Y_{l,m}\rangle(t)$ [averaged spherical harmonics $Y_{l,m}(\vartheta,\varphi)$] governing the magnetization decay

$$\frac{d}{dt}c_{l,m}(t)=\sum_{l',m'} d_{l',m',l,m}c_{l',m'}(t), \tag{A1}$$

where $d_{l',m',l,m}$ are the matrix elements of the Fokker-Planck operator in Eq. (1). A method of derivation of the statistical moment system Eq. (A1) for an arbitrary free energy is given in Refs. 22 and 33. The solution of Eq. (A1) can always be obtained by matrix-continued fractions.[22] In essence, we transform the moment system Eq. (A1) into a tridiagonal vector recurrence equation

$$\tau_N\frac{d}{dt}\mathbf{C}_n(t)=\mathbf{Q}_n^-\mathbf{C}_{n-1}(t)+\mathbf{Q}_n\mathbf{C}_n(t)+\mathbf{Q}_n^+\mathbf{C}_{n+1}(t), \tag{A2}$$

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 whose elements are $d_{l',m',l,m}$ (for the problem in question they are given explicitly in Refs. 22 and 34). The *exact solution* of Eq. (A2) for the Laplace transform of $\mathbf{C}_1(t)$ is given by[22]

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

where the infinite matrix-continued fraction $\boldsymbol{\Delta}_n(s)$ is defined by the recurrence equation

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

and $\mathbf{I}$ are the unit matrices. Furthermore, in terms of the matrix-continued fractions $\boldsymbol{\Delta}_n(0)$, one can also estimate the smallest nonvanishing eigenvalue λ_1 given by the smallest root of the secular equation[22,34]

$$\det(\lambda_1\mathbf{I}-\mathbf{S})=0, \tag{A3}$$

where the matrix $\mathbf{S}$ is defined as

$$\mathbf{S}=-\tau_N^{-1}[\mathbf{Q}_1+\mathbf{Q}_1^+\boldsymbol{\Delta}_2(0)\mathbf{Q}_2^-]\times\left[\mathbf{I}+\sum_{n=2}^{\infty}\prod_{m=1}^{n-1}\mathbf{Q}_m^+\prod_{k=1}^{n-1}\boldsymbol{\Delta}_{n-k+1}^2(0)\mathbf{Q}_{n-k+1}^-\right]^{-1},$$

i.e., λ_1 is the smallest nonvanishing eigenvalue of the matrix $\mathbf{S}$. The inverse of λ_1 corresponds to the reversal time of the magnetization τ.

[1] L. Néel, Ann. Géophys. (C.N.R.S.) **5**, 99 (1949).
[2] W. F. Brown, Jr., Phys. Rev. **130**, 1677 (1963).
[3] W. F. Brown, Jr., IEEE Trans. Magn. **15**, 1196 (1979).
[4] H. A. Kramers, Physica (Amsterdam) **7**, 284 (1940).
[5] W. T. Coffey, D. A. Garanin, and D. J. McCarthy, Adv. Chem. Phys. **117**, 483 (2001).
[6] D. A. Smith and F. A. de Rozario, J. Magn. Magn. Mater. **3**, 219 (1976).
[7] J. S. Langer, Ann. Phys. (N.Y.) **54**, 258 (1969).
[8] I. Klik and L. Gunther, J. Appl. Phys. **67**, 4505 (1990); J. Stat. Phys. **60**, 473 (1990).
[9] P. M. Déjardin, D. S. F. Crothers, W. T. Coffey, and D. J.

McCarthy, Phys. Rev. E **63**, 021102 (2001).
[10] V. I. Mel'nikov and S. V. Meshkov, J. Chem. Phys. **85**, 1018 (1986); V. I. Mel'nikov, Phys. Rep. **209**, 1 (1991).
[11] Yu. P. Kalmykov, J. Appl. Phys. **96**, 1138 (2004).
[12] Yu. P. Kalmykov, W. T. Coffey, B. Ouari, and S. V. Titov, J. Magn. Magn. Mater. **292**, 372 (2005); Yu. P. Kalmykov, W. T. Coffey, and S. V. Titov, Phys. Solid State **47**, 272 (2005); Yu. P. Kalmykov and B. Ouari, Phys. Rev. B **71**, 094410 (2005); B. Ouari and Yu. P. Kalmykov, J. Appl. Phys. **100**, 123912 (2006); Yu. P. Kalmykov, *ibid.* **101**, 093909 (2007); W. T. Coffey, P. M. Déjardin, and Yu. P. Kalmykov, Phys. Rev. B **79**, 054401 (2009); B. Ouari, S. Aktaou, and Yu. P. Kalmykov, *ibid.* **81**, 024412 (2010).
[13] C. Vouille, A. Thiaville, and J. Miltat, J. Magn. Magn. Mater. **272-276**, E1237 (2004).
[14] H. J. Suh, C. Heo, C. Y. You, W. Kim, T. D. Lee, and K. J. Lee, Phys. Rev. B **78**, 064430 (2008); H. J. Suh and K. J. Lee, Curr. Appl. Phys. **9**, 985 (2009).
[15] N. A. Usov and Yu. B. Grebenshchikov, in *Magnetic Nanoparticles*, edited by S. P. Gubin (Wiley, New York, 2009), p. 303; J. Appl. Phys. **105**, 043904 (2009).
[16] W. T. Coffey, D. S. F. Crothers, J. L. Dormann, Yu. P. Kalmykov, E. C. Kennedy, and W. Wernsdorfer, Phys. Rev. Lett. **80**, 5655 (1998).
[17] Y. Nakatani, Y. Uesaka, N. Hayashi, and H. Fukushima, J. Magn. Magn. Mater. **168**, 347 (1997).
[18] W. Scholz, T. Schrefl, and J. Fidler, J. Magn. Magn. Mater. **233**, 296 (2001).
[19] O. Chubykalo, U. Nowak, R. Smirnov-Rueda, M. A. Wongsam, R. W. Chantrell, and J. M. Gonzalez, Phys. Rev. B **67**, 064422 (2003).
[20] X. Z. Cheng, M. B. A. Jalil, H. K. Lee, and Y. Okabe, Phys. Rev. B **72**, 094420 (2005); J. Appl. Phys. **99**, 08B901 (2006).
[21] X. Z. Cheng, M. B. A. Jalil, H. K. Lee, and Y. Okabe, Phys. Rev. Lett. **96**, 067208 (2006).
[22] W. T. Coffey, Yu. P. Kalmykov, and J. T. Waldron, *The Langevin Equation*, 2nd ed. (World Scientific, Singapore, 2004).
[23] A. Lyberatos and R. W. Chantrell, J. Appl. Phys. **73**, 6501 (1993).
[24] J. L. García-Palacios and F. J. Lázaro, Phys. Rev. B **58**, 14937 (1998); J. L. García-Palacios, Adv. Chem. Phys. **112**, 1 (2000).
[25] D. V. Berkov, IEEE Trans. Magn. **38**, 2489 (2002).
[26] D. V. Berkov and N. L. Gorn, in *Handbook of Advanced Magnetic Materials: Characterization and Simulation*, edited by Y. Liu, D. J. Sellmyer, and D. Shindo (Springer, New York, 2006), Vol. II, p. 422.
[27] O. Chubykalo, R. Smirnov-Rueda, J. M. Gonzalez, M. A. Wongsam, R. W. Chantrell, and U. Nowak, J. Magn. Magn. Mater. **266**, 28 (2003).
[28] O. Chubykalo, J. D. Hannay, M. A. Wongsam, R. W. Chantrell, and J. M. Gonzalez, Phys. Rev. B **65**, 184428 (2002).
[29] D. V. Berkov and N. L. Gorn, J. Phys.: Condens. Matter **14**, 281 (2002).
[30] E. Martínez, L. López-Díaz, L. Torres, and O. Alejos, Physica B **343**, 252 (2004).
[31] N. Smith, Phys. Rev. B **74**, 026401 (2006).
[32] D. A. Garanin and O. Chubykalo-Fesenko, Phys. Rev. B **70**, 212409 (2004).
[33] Yu. P. Kalmykov and S. V. Titov, Phys. Rev. Lett. **82**, 2967 (1999); J. Magn. Magn. Mater. **210**, 233 (2000).
[34] Yu. P. Kalmykov and S. V. Titov, Fiz. Tverd. Tela (St. Petersburg) **40**, 1642 (1998) [Phys. Solid State **40**, 1492 (1998)]; Yu. P. Kalmykov, Phys. Rev. E **62**, 227 (2000); W. T. Coffey, D. S. F. Crothers, Yu. P. Kalmykov, and S. V. Titov, Phys. Rev. B **64**, 012411 (2001).
[35] D. A. Garanin, E. C. Kennedy, D. S. F. Crothers, and W. T. Coffey, Phys. Rev. E **60**, 6499 (1999).