Comparison of methods for the calculation of superparamagnetic relaxation times

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A general expression for the correlation time of the decay of the magnetization of an assembly of single-domain noninteracting ferromagnetic particles is given in terms of the inverse of the Fokker-Planck operator. The results of Moro and Nordio [G. Moro and P. L. Nordio, Mol. Phys. **56**, 255 (1985)], given in the context of dielectric relaxation, are recovered when the Fokker-Planck operator is axially symmetric. Their result is a particular example of Szabo's calculation of the correlation times of the autocorrelation functions of the Legendre polynomials by means of a generalization of the theory of first-passage times [A. Szabo, J. Chem. Phys. **72**, 4620 (1980)]. Likewise, the results of Garanin, Ischenko, and Panina (D. A. Garanin, V. V. Ischenko, and L. V. Panina, Teor. Mat. Fiz. **82**, 242 (1990) [Theor. Math. Phys. **82**, 169 (1990)]) for the integral relaxation time, i.e., the area under the curve of the normalized decay of the magnetization, are regained in the axially symmetric case where it is possible to integrate the Fokker-Planck equation directly. It is shown by manipulation of Kummer's functions that the exact integral expression for the correlation time for simple uniaxial anisotropy derived by Coffey *et al.* [W. T. Coffey, D. S. F. Crothers, Yu. P. Kalmykov, E. S. Massawe, and J. T. Waldron. Phys. Rev. E **49**, 1869 (1994)] by representing the Fokker-Planck equation as a differential-recurrence relation is identical to the integral relaxation time originally derived by Garanin *et al.* by direct integration of the Fokker-Planck equation. [S1063-651X(96)08811-3]

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I. INTRODUCTION

A single-domain ferromagnetic particle with uniaxial anisotropy is characterized by an internal magnetic potential that has two stable stationary points with a potential barrier between them. If the particle is sufficiently fine, the direction of the magnetization may undergo a rotation due to thermal agitation, surmounting the barrier, as first described by Néel [1].

The calculation of the relaxation behavior of an assembly of such particles is usually accomplished [1–3] by assuming that the relaxation of the magnetization is dominated by a single relaxation mode, namely, that associated with the time of reversal of the magnetization over the energy barrier between two stable orientational states. This means that in the set of eigenvalues $\{\lambda_k\}$ and corresponding amplitudes $\{A_k\}$ of the Sturm-Liouville equation (to which the Fokker-Planck equation underlying the process may be converted), $\lambda_1 \ll \lambda_k$, $k \geqslant 2$, and $A_1 \gg A_k$ since then the decay functions $A_k \exp(-\lambda t/\tau_N)$, $k \geqslant 2$, are small compared to $A_1 \exp(-\lambda_1 t/\tau_N)$, except in the very early stages of an approach to equilibrium. The diffusional relaxation time τ_N is defined as [2],

$$\tau_N = \frac{v}{2\eta kT}\left[\frac{1}{\gamma^2} + \eta^2 M_s^2\right], \tag{1}$$

where γ is the gyromagnetic ratio, M_s is the saturation magnetization, k is the Bolzmann constant, T is the absolute temperature, v is the volume of the particle, and η is the phenomenological damping constant from Gilbert's equation, namely [2],

$$\dot{\mathbf{M}} = \gamma \mathbf{M} x [\mathbf{H}_{\rm tot} - \eta \dot{\mathbf{M}}]. \tag{2}$$

In Eq. (2) $\mathbf{M}$ denotes the magnetization and

$$\mathbf{H}_{\rm tot} = \mathbf{h}_r - \partial V/\partial \mathbf{M}, \tag{3}$$

where $\mathbf{h}_r$ is the random white-noise field arising from thermal agitation and V is the barrier potential including that of the internal crystalline anisotropy and the applied external field $\mathbf{H}$.

In view of the above considerations the early studies [2–4] of the relaxation process were confined to the calculation of the smallest nonvanishing eigenvalue of the Sturm-Liouville equation making the assumption that the process is dominated by a single relaxation mode with the time constant

$$\tau \simeq \frac{\tau_N}{\lambda_1}. \tag{4}$$

Recently, Garanin, Ischenko, and Panina [5] and earlier Moro and Nordio [6] (in the context of the analogous problem in chemical physics) have introduced the concept of the ''integral relaxation time,'' which in the present context is proportional to the area under the curve that describes the relaxation of the magnetization after an abrupt change of a magnetic field that had been applied along the anisotropy axis. Furthermore, in linear response (that is, for an infinitesimal change in the magnetic field) they were able [5] to obtain a general expression for the integral relaxation time of

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a single-domain ferromagnetic particle for all values of the barrier height parameter. Here the integral relaxation time is identical to the correlation time of linear-response theory since the decay of the magnetization is now proportional to the magnetization autocorrelation function [7]. This formulation has been used extensively by Coffey *et al.* [7]. According to Garanin, Ischenko, and Panina [5] and Coffey *et al.* [7,8], the integral relaxation time presents a more accurate picture of the relaxation process rather than the approximation [2] of a ''long-lived'' [5] exponential decay mode that underlies Eq. (4) as the contribution of all the decay modes are included in the integral relaxation time. This is of particular importance in the problem of the response following an infinitesimal change in a strong bias field applied along the anisotropy axis where, as discovered in Ref. [8] at a value of the bias field far less than the critical value required to maintain the two-well structure of the potential, there is an abrupt departure of the decay time of the longest-lived (Néel) mode from the correlation time. Another advantage of the integral relaxation time is that it is possible to obtain exact analytic solutions for it in a number of problems [7,9,10]. Indeed, this property has very recently allowed Garanin [11] to give a physical explanation of the constant magnetic-field effect described in Ref. [8].

The analysis given by Garanin, Ischenko, and Panina [5] and Moro and Nordio [6] was carried out by converting the Fokker-Planck equation underlying the problem to a Sturm-Liouville equation by using in the case of Ref. [6] the Laplace transform and linear-response theory and in Ref. [5] assuming a solution where the time variation of the bias field is $\exp i\omega t$ and confirming the response to small impressed fields. The solution of the resulting Sturm-Liouville equation in the zero-frequency limit then yields the integral relaxation time. In the special case of an applied field parallel to the anisotropy axis so that the problem is axially symmetric, a first integral of the Sturm-Liouville equation may be immediately written down so that the complete solution is easily given by quadratures [12], so yielding an *exact* expression (in integral form) for the correlation time.

A different approach to the problem has been made in the series of papers by Coffey and co-workers [7–10]. Instead of representing the solution (of the Fokker-Planck equation) as a Sturm-Liouville problem, they have used a Floquet approach [12,13] whereby the solution is expanded in a Fourier series, in the manner often used for the solution of Hill's equation [12–14]. Thus the calculation of the decay of the magnetization invariably reduces to the solution of a set of differential-recurrence relations. On taking the Laplace transform this set may be solved numerically using the standard methods of linear algebra. In a few special cases [7,9,10] we cite in particular [7] the simple uniaxial potential of the crystalline anisotropy (after a weak collinear dc field has been removed):

$$\upsilon V(\vartheta)=K\upsilon(1-z^2)=K\upsilon\ \sin^2\vartheta, \tag{5}$$

where K is the anisotropy constant and ϑ is the polar angle specifying the direction of the magnetization; the set of algebraic recurrence relations (which now reduce to a three-term one) may be solved analytically in the zero-frequency limit so that the correlation time may be calculated. In particular for the potential of Eq. (5), the correlation time may [7] be expressed as a sum of the products of Kummer functions [15], which in turn may be expressed in integral form [Eqs. (40) and (54) of Ref. [7]].

It is not immediately obvious that the Sturm-Liouville approach of Garanin, Ischenko, and Panina [5] and Moro and Nordio [6] will yield the same results as the Floquet approach favored by Coffey and co-workers [7–10]. It is the purpose of this paper to show that the results of both approaches are identical, taking as an example the simple potential of Eq. (5) for which exact results from both methods of attack on the problem are available. In addition, we shall highlight the advantages and drawbacks associated with both methods of solution. It is first necessary to summarize the general calculation of the integral relaxation time.

II. CALCULATION OF THE INTEGRAL RELAXATION TIME

In general, the Fokker-Planck equation for the probability density $W(\vartheta,\varphi,t)$ of orientations of the magnetization vector **M** on the unit sphere is [2] (φ is the azimuthal angle)

$$2\tau_N\frac{\partial W}{\partial t}=\Lambda^2 W+\frac{\beta}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left(\sin\vartheta\frac{\partial V}{\partial\vartheta}W-\frac{1}{a}\frac{\partial V}{\partial\phi}W\right)+\frac{\beta}{\sin\vartheta}\frac{\partial}{\partial\phi}\left(\frac{1}{\sin\vartheta}\frac{\partial V}{\partial\phi}W+\frac{1}{a}\frac{\partial V}{\partial\vartheta}W\right), \tag{6}$$

where $\beta=\upsilon/kT$, $a=\eta\gamma Ms$ is a dimensionless damping parameter, $V(\vartheta,\phi)$ is the Gibbs free-energy density, and Λ^2 denotes the angular part of the Laplacian

$$\Lambda^2=\frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left(\sin\vartheta\frac{\partial}{\partial\vartheta}\right)+\frac{1}{\sin^2\vartheta}\frac{\partial^2}{\partial\phi^2}.$$

Equation (6) has the generic form

$$\frac{\partial W}{\partial t}=LW, \tag{7}$$

where L denotes the Fokker-Planck operator in Eq. (6). The potential energy $\upsilon V(\vartheta,\varphi)$ arises from the crystalline anisotropy potential and the action of an external uniform magnetic field **H** characterized by the external field parameter

$$\xi=\upsilon\frac{M_sH}{kT}. \tag{8}$$

If we now suppose that ξ is decreased abruptly by a small amount ξ_1 such that $\xi_1\ll 1$ so that the ensuing response is linear in ξ_1, the solution of Eq. (7) at any time after the perturbation has been made will be of the form [16,17]

$$W_t=W^{(0)}+W^{(1)}+W^{(2)}+\cdots, \tag{9}$$

where $W^{(0)}$ is the new equilibrium distribution (attained after the relaxation from the former equilibrium distribution W_0, which had prevailed up to the time $t=0$ before the abrupt change in the field), $W^{(1)}$ is the portion of the response linear in ξ_1, $W^{(2)}$ that quadratic in ξ_1, and so on. Since

$$LW^{(0)}=0, \tag{10}$$

we shall have

$$\frac{\partial W^{(1)}}{\partial t} = L W^{(1)}. \tag{11}$$

The formal solution of Eq. (11) is

$$W^{(1)} = (\exp Lt) W_0^{(1)}, \tag{12}$$

where $W_0^{(1)}$ denotes the value of $W^{(1)}$ at $t=0$ that is before ξ_1 has been removed. Hence the time-dependent orientational distribution function is, in linear response,

$$W_t = W^{(0)} + (\exp Lt) W_0^{(1)}. \tag{13}$$

The ratio of potential energy to thermal energy is, for uniaxial anisotropy,

$$\frac{vV}{kT} = -\sigma(\mathbf{u}\cdot\mathbf{n})^2 - \xi(\mathbf{u}\cdot\mathbf{h}), \tag{14}$$

where

$$\sigma = \frac{Kv}{kT} \tag{15}$$

is the barrier height parameter, **u**, **h**, and **n** are unit vectors in the direction of **M**, the field axis **H**, and the anisotropy axis is denoted by **n**. If we alter ξ by the small amount ξ_1 in order to apply the perturbation we have

$$W_0 \equiv W_0(\vartheta,\varphi,o) = \frac{e^{-\beta V[1+\xi_1(\mathbf{u}\cdot\mathbf{h})]}}{\int_\Omega e^{-\beta V}[1+\xi_1(\mathbf{u}\cdot\mathbf{h})]d\Omega}, \tag{16}$$

where $d\Omega$ denotes the element of solid angle, i.e.,

$$d\Omega = \sin\vartheta \; d\vartheta \; d\phi.$$

Thus, ignoring terms $O(\xi_1^2)$, we have the linear approximation

$$W_0 \equiv W(\vartheta,\phi,o) = W^{(0)}(\vartheta,\phi)\{1+\xi_1[(\mathbf{u}\cdot\mathbf{h}) - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0]\}, \tag{17}$$

where the symbol $\langle\,\rangle_0$ denotes the equilibrium ensemble average over $W^{(0)}$. Thus Eq. (13) becomes the zero on the angular brackets indicating that the average is to be performed in the absence of the perturbation ξ_1,

$$W_t = W^{(0)}(\vartheta,\phi) + \xi_1(\exp Lt) W^{(0)}(\vartheta,\phi)[(\mathbf{u}\cdot\mathbf{h}) - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0]. \tag{18}$$

The mean change in the magnetic moment in the direction of **h** following the perturbation is then

$$\begin{aligned}
\langle \Delta m\rangle_{\xi 1}(t) &= M_s[\langle \mathbf{u}\cdot\mathbf{h}\rangle - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0]_{\xi_1} \\
&= \xi_1 M_s \int_\Omega (\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0) e^{Lt} W^{(0)} \\
&\quad \times (\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0) d\Omega \\
&= \xi_1 M_s \langle (\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0)(0)(\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0)(t)\rangle_0 \\
&= \frac{H_1}{kT} \langle \Delta\mathbf{m}(0)\cdot\Delta\mathbf{m}(t)\rangle_0 \\
&= \frac{H_1}{kT}[\langle \mathbf{m}(0)\cdot\mathbf{m}(t)\rangle_0 - \langle m\rangle_0^2] = \frac{H_1}{kT} b_m(t),
\end{aligned} \tag{19}$$

and we have noted that $\langle m\rangle_0 = \langle m(0)\rangle_0 = \langle m(\infty)\rangle_0$. Thus the time-dependent magnetic moment due to the small change in the external field has been expressed in terms of the *equilibrium correlation function* $b_m(t)$. The integral relaxation time T is [5] the area under the slope of the normalized decay function so that

$$T = \frac{\int_0^\infty \langle \Delta m\rangle_{\xi_1}(t)dt}{\langle \Delta m\rangle_{\xi_1}(0)}. \tag{20}$$

Equation (20) may be written in terms of the zero-frequency limit of its Laplace transform as

$$\begin{aligned}
T &= \lim_{s\to 0}\left\{ \int_0^\infty \frac{\langle \Delta m\rangle_{\xi_1}(t) e^{-st} dt}{\langle \Delta m\rangle_{\xi_1}(0)} \right\} \\
&= \lim_{s\to 0} \frac{\langle \Delta\widetilde{m}\rangle_{\xi_1}(s)}{\langle \Delta m\rangle_{\xi}(0)} = \frac{\langle \Delta\widetilde{m}\rangle_{\xi_1}(0)}{\langle \Delta m\rangle_{\xi_1}(0)}.
\end{aligned} \tag{21}$$

Equation (21) is a general expression and does not rely on the assumption of linear response *per se*. It may be related, however, to the response in the presence of an alternating field only when linearity of the response is assumed, in which case Eq. (21) may be related to the magnetization correlation function $b_m(t)$ by means of Eq. (19) as follows. We have $\mathcal{L}$ denoting the Laplace transformation

$$T = \lim_{s\to 0} \mathcal{L}\left\{ \frac{\langle \mathbf{m}(0)\cdot\mathbf{m}(t)\rangle_0 - \langle m\rangle_0^2}{\langle m^2\rangle_0 - \langle m\rangle_0^2} \right\} = \frac{\widetilde{b}_m(0)}{b_m(0)} = \widetilde{c}_m(0), \tag{22}$$

or in view of Eq. (19)

$$T = \lim_{s\to 0}\left\{ \frac{\int_\Omega (\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0)(s-L)^{-1} W^{(0)}[\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0] d\Omega}{\int_\Omega [(\mathbf{u}\cdot\mathbf{h}) - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0]^2 d\Omega} \right\} \tag{23}$$

$$= \int_\Omega \frac{[(\mathbf{u}\cdot\mathbf{h}) - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0](-L)^{-1} W^{(0)}[\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0]}{\langle [\mathbf{u}\cdot\mathbf{h} - \langle \mathbf{u}\cdot\mathbf{h}\rangle_0]^2\rangle_0} \, d\Omega. \tag{24}$$

The formal solution for T, Eq. (24), although derived by referring to the uniaxial potential of Eq. (14), is a general formula that applies *regardless* of the precise form of the potential $\upsilon V(\vartheta,\phi)$ as long as the perturbation ξ_1 is applied parallel to the field axis. The task of calculating the integral relaxation time from Eq. (24) always reduces to the determination of

$$(-L)^{-1}\frac{e^{-\beta V}}{Z}[\mathbf{u}\cdot\mathbf{h}-\langle\mathbf{u}\cdot\mathbf{h}\rangle_0], \tag{25}$$

where the partition function is

$$Z=\int_{-1}^{+1}e^{-\beta V(z)}dz.$$

Analytic solutions of this problem may be determined in only a few specialized cases, in particular those where a first integral [5,6] may be determined when the problem is couched in Sturm-Liouville form or when the zero-frequency limit of the continued-fraction solution [4] in the Floquet representation [7,9] may be recognized as a known function.

III. INTEGRABLE CASES OF EQ. (25)

Equation (25) and so T may be calculated exactly for the case of axial symmetry. Here Eq. (6), with $z=\cos\vartheta$, becomes

$$2\tau_N\frac{\partial W}{\partial t}(z,t)=\frac{\partial}{\partial z}\left[(1-z^2)\left(\frac{\partial W(z,t)}{\partial z}+\beta V'(z)W(z,t)\right)\right], \tag{26}$$

and taking the Laplace transform

$$2\tau_N[s\widetilde{W}(z,s)-W(z,0)]=\frac{\partial}{\partial z}\left[(1-z^2)\left(\frac{\partial\widetilde{W}(z,s)}{\partial z}+\beta V'(z)\widetilde{W}(z,s)\right)\right] \tag{27}$$

and in the limit $s\to 0$, using the final value theorem of Laplace transformation,

$$2\tau_N[W(z,\infty)-W(z,0)]=\frac{\partial}{\partial z}\left[(1-z^2)\left(\frac{\partial\widetilde{W}(z,0)}{\partial z}+\beta V'(z)\widetilde{W}(z,0)\right)\right]. \tag{28}$$

The unknown functions W on the left-hand side may now be determined explicitly, allowing us to integrate Eq. (28). We have

$$\begin{aligned}2\tau_N[W(z,\infty)-W(z,0)]&=2\tau_N\left[\frac{e^{-\beta V}}{Z}-\frac{e^{-\beta V}[1+\xi_1 z]}{Z[1+\xi_1\langle z\rangle_0]}\right]\\&=-2\xi_1\tau_N\frac{e^{-\beta V}}{Z}[z-\langle z\rangle_0]+O(\xi_1^2);\end{aligned} \tag{29}$$

thus Eq. (28) becomes in the linear approximation in ξ_1 (all sub- and superscripts are dropped from W as the meaning of W is now obvious)

$$-2\tau_N\xi_1\frac{e^{-\beta V}}{Z}[z-\langle z\rangle_0]=\frac{d}{dz}\left[(1-z^2)\left(\frac{d\widetilde{W}}{dz}+\beta V'(z)\widetilde{W}\right)\right]=2\tau_N L\widetilde{W}(z,0). \tag{30}$$

The particular integral of Eq. (30), and so Eq. (25), is best calculated by introducing the variable

$$F(z)=e^{\beta V(z)}\widetilde{W}(z,0), \tag{31}$$

so that Eq. (30) now becomes

$$-2\tau_N\xi_1\frac{e^{-\beta V}}{Z}[z-\langle z\rangle_0]=\frac{d}{dz}\left[(1-z^2)e^{-\beta V}\frac{dF(z)}{dz}\right], \tag{32}$$

which, mindful of the interval of the solution being $[-1,1]$ becomes

$$(1-z^2)e^{-\beta V}\frac{dF(z)}{dz}+c_1=\frac{-2\tau_N\xi_1}{Z}\int_{-1}^{z}e^{-\beta V(z_1)}\times[z_1-\langle z_1\rangle_0]dz_1,$$

with, since the probability current must vanish on the boundaries,

$$c_1=(1-z^2)e^{-\beta V}\frac{dF(z)}{dz}\bigg|_{z=-1}=0.$$

Note that this condition also implies that $F'(z)$ is *finite* on the boundaries, that is, at the poles of the sphere. Thus

$$\frac{dF(z)}{dz}=-\frac{2\tau_N\xi_1}{1-z^2}\frac{e^{\beta V(z)}}{Z}\int_{-1}^{z}e^{-\beta V(z_1)}[z_1-\langle z_1\rangle_0]dz_1, \tag{33}$$

and so integrating once more

$$F(z)=-2\tau_N\xi_1\int_{-1}^{z}\frac{dz_2}{(1-z_2^2)}\frac{e^{\beta V}}{Z}\int_{-1}^{z_2}e^{-\beta V(z_1)}\times[z_1-\langle z_1\rangle_0]dz_1+c_2, \tag{34}$$

where

$$c_2=F(z)|_{z=-1}. \tag{35}$$

Thus the formal expression (25) reduces in this instance to

$$\frac{e^{\beta V(z)}}{2\tau_N\xi_1}F(z)=-e^{-\beta V(z)}\int_{-1}^{z}\frac{dz_2}{(1-z_2^2)}\frac{e^{\beta V}}{Z}\int_{-1}^{z_2}e^{-\beta V(z_1)}\times[z_1-\langle z_1\rangle_0]dz_1+\frac{c_2}{\xi_1}e^{-\beta V(z)} \tag{36}$$

and so the exact integral relaxation time (correlation time) is from Eq. (24)

$$\frac{T}{2\tau_N}=\frac{-\int_{-1}^{+1}dz\ e^{-\beta V(z)}[z-\langle z\rangle_0]\int_{-1}^{z}\frac{dz_2}{(1-z_2^2)}\frac{e^{\beta V}}{Z}\int_{-1}^{z_2}e^{-\beta V(z_1)}[z_1-\langle z_1\rangle_0]dz_1}{\langle[z-\langle z\rangle_0]^2\rangle_0} \tag{37}$$

since the second term in Eq. (36) vanishes in the integral in Eq. (37). We further note on integration by parts, that Eq. (37) may be written as

$$\frac{T}{2\tau_N}=\int_{-1}^{+1}\frac{\frac{dz}{1-z^2}\frac{e^{\beta V(z)}}{Z}\{\int_{-1}^{z}e^{-\beta V(z_1)}[z_1-\langle z_1\rangle_0]dz_1\}^2}{\langle z^2\rangle_0-\langle z\rangle_0^2}. \tag{38}$$

Equation (38) is the exact solution for the integral relaxation time for the axially symmetric Fokker-Planck operator of Eq. (26). It was essentially derived in the chemical physics context using a different formulation by Moro and Nordio [6] and was later rederived in the context of magnetic relaxation by Garanin, Ischenko, and Panina [5], who approached the problem by calculating the response due to a weak alternating field. We shall now evaluate Eq. (38) for the particular case of a weak dc field reduced to zero at time $t=0$ so that [8]

$$\beta V(\vartheta)=\sigma\ \sin^2\vartheta=\sigma(1-z^2) \tag{39}$$

and we shall prove that the solution yielded by Eq. (38) is identical in all respects to the integral form of the solution found by summing the series of products of Kummer [15] functions yielded by the continued-fraction solution in Coffey *et al.* [7].

IV. COMPARISON OF THE INTEGRAL RELAXATION TIME AND KUMMER FUNCTION SOLUTIONS

In order to evaluate T using Eq. (38) for the potential of Eq. (39) and to show that it is identical to Eq. (55) of Coffey *et al.* [7], namely, [$M(a,b,z)$ denotes Kummer's function [15]; the subscript $\|$ denotes the longitudinal correlation time],

$$\frac{T_\|}{\tau_N}=M\left(1,\frac{5}{2},\sigma\right)-\frac{3e^\sigma\sigma^{-2}}{4M(\frac{3}{2},\frac{5}{2},\sigma)}\int_0^{\pi/2}d\theta\,\frac{\sqrt{1+\cos\theta}}{\cos\theta}\times\left[\frac{1}{2}(e^{\sigma\cos\theta}+e^{-\sigma\cos\theta})+2-\frac{3}{2}\left(\frac{e^{\sigma\cos\theta}-e^{-\sigma\cos\theta}}{\sigma\cos\theta}\right)\right]. \tag{40}$$

We note that Eq. (38) reduces for the potential of Eq. (39) to

$$\frac{T}{2\tau_N}=\int_{-1}^{+1}\frac{dz}{1-z^2}\frac{e^{\sigma(1-z^2)}}{Z}\frac{1}{4\sigma^2}\frac{[1-e^{-\sigma(1-z^2)}]^2}{\langle z^2\rangle_0}=\int_{-1}^{+1}\frac{dz}{1-z^2}\frac{e^{-\sigma(1-z^2)}}{Z}\frac{1}{4\sigma^2}\frac{[1-e^{\sigma(1-z^2)}]^2}{\langle z^2\rangle_0}, \tag{41}$$

that is,

$$\frac{T}{2\tau_N}=\frac{1}{4\sigma^2}\frac{\langle(1-z^2)^{-1}[1-e^{\sigma(1-z^2)}]^2\rangle_0}{\langle z^2\rangle_0} \tag{42}$$

or

$$\frac{T}{\tau_N}=\frac{3e^\sigma}{\sigma^2}\frac{\int_0^1\frac{dz}{1-z^2}\{\cosh[\sigma(z^2-1)]-1\}}{M(\frac{3}{2},\frac{5}{2},\sigma)}, \tag{43}$$

where we have noted that [15]

$$\int_0^1 e^{\sigma z^2}dz=M(\tfrac{1}{2},\tfrac{3}{2};\sigma)=2Ze^\sigma, \tag{44}$$

and since the partition function $Z=\int_{-1}^{1}e^{\sigma(z^2-1)}dz$,

$$\frac{dM(a,b,z)}{dz}=\frac{a}{b}M(a+1,b+1,z). \tag{45}$$

We shall now demonstrate that Eqs. (40) and (43) are identical. Our task is to prove that (taking a common denominator)

$$4\sigma^2M(1,\tfrac{5}{2},\sigma)M(\tfrac{3}{2},\tfrac{5}{2},\sigma)-3e^\sigma\int_0^{\pi/2}d\theta\times\left\{\frac{\sqrt{1+\cos\theta}}{\cos\theta}\left[\cosh(\sigma\cos\theta)-3\frac{\sinh(\sigma\cos\theta)}{\sigma\cos\theta}+2\right]\right\}=12e^\sigma\int_0^1\frac{dz}{1-z^2}\{\cosh[\sigma(1-z^2)]-1\}. \tag{46}$$

We first remark that the leading term on the left-hand side may be written in integral form as follows. We have

$$M(1,\tfrac{5}{2},\sigma)M(\tfrac{3}{2},\tfrac{5}{2},\sigma)=e^\sigma M(1,\tfrac{5}{2},\sigma)M(1,\tfrac{5}{2},-\sigma). \tag{47}$$

Also [7], $I_v(x)$ is the modified Bessel function of the first kind,

$$M(1,\tfrac{5}{2},\sigma)M(1,\tfrac{5}{2},-\sigma)$$
$$=\frac{[\Gamma(\tfrac{5}{2})]^2\sigma^{-3/2}}{\Gamma(1)\Gamma(\tfrac{3}{2})}\int_{-\infty}^{\infty}dt\ e^{t/2}\mathrm{sech}t\ I_{3/2}(\sigma\ \mathrm{sech}t)$$
$$=2(\tfrac{3}{2})^2\tfrac{1}{2}\sqrt{\pi}\sigma^{-3/2}\int_0^{\infty}\mathrm{sech}t\ I_{3/2}(\sigma\ \mathrm{sech}t)\cosh\frac{t}{2}\,dt$$
$$=-\tfrac{9}{4}\sqrt{\pi}\sigma^{-3/2}\int_0^{\pi/2}d\theta\,\frac{dt}{d\theta}\sin\theta\ I_{3/2}(\sigma\ \sin\theta)\cosh\frac{t}{2}, \tag{48}$$

where $\mathrm{sech}t=\sin\theta$.

Whence we have

$$M(1,\tfrac{5}{2},\sigma)M(1,\tfrac{5}{2},-\sigma)$$
$$=\frac{9}{4\sigma^2}\int_0^{\pi/2}d\theta\left\{\frac{\sqrt{1+\sin\theta}}{\sin\theta}\times\left[\cosh(\sigma\ \sin\theta)-\frac{\sinh(\sigma\ \sin\theta)}{\sigma\ \sin\theta}\right]\right\}. \tag{49}$$

On substituting Eq. (49) into Eq. (46) and using Eq. (47) we find after some algebra that the left-hand side of Eq. (46) is (cancelling the common factor e^{σ} on both sides)

$$9\int_0^{\pi/2}d\theta\,\frac{\sqrt{1+\sin\theta}}{\sin\theta}\left[\cosh(\sigma\ \sin\theta)-\frac{\sinh(\sigma\ \sin\theta)}{\sigma\ \sin\theta}\right]$$
$$-3\int_0^{\pi/2}d\theta\,\frac{\sqrt{1+\cos\theta}}{\cos\theta}$$
$$\times\left[\cosh(\sigma\ \cos\theta)-3\,\frac{\sinh(\sigma\ \cos\theta)}{\sigma\ \cos\theta}+2\right],$$

which duly reduces to

$$6\int_0^{\pi/2}d\theta\,\frac{\sqrt{1+\cos\theta}}{\cos\theta}\,[\cosh(\sigma\ \cos\theta)-1]. \tag{50}$$

On writing $(1-x^2)=\cos\theta$, Eq. (50) becomes

$$12\int_0^1\frac{dx}{1-x^2}\,\{\cosh[\sigma(1-x^2)]-1\}, \tag{51}$$

so proving that Eqs. (40) and (43) are identical.

V. CONCLUSIONS

In this paper we have given a general expression, viz., Eq. (24), for the integral relaxation time T based on linear-response theory so that the integral relaxation time is identical to the correlation time of the autocorrelation function of the change in the magnetization. The result agrees in all respects with those of Garanin, Ischenko, and Punina [5] and Moro and Nordio [6]. T may be evaluated explicitly when the inverse of the Fokker-Planck operator L may be evaluated. In practice, it appears that this may be accomplished analytically only for problems with axial symmetry when the Sturm-Liouville equation may be integrated exactly, leading to Eq. (38), which now constitutes the general solution of the problem.

We have evaluated Eq. (38) for the simple uniaxial potential of the crystalline anisotropy and we have demonstrated that the result is identical to the (Floquet) method based on differential-recurrence relations of Coffey *et al.* [7]. The Sturm-Liouville method has the great merit, in the uniaxial case, that it is relatively simple to calculate T without the complicated mathematical manipulations associated with the calculation of T from the differential-recurrence relations of Coffey *et al.* [7]. In addition, it is possible to exactly evaluate T analytically for a small change in a strong dc bias field applied along the anisotropy axis. An important consequence of this is that by evaluating the integral for values of the parameter

$$h=\frac{\xi}{2\sigma}$$

in the range 0.1–0.2, Garanin [11] has been able to give a clear physical explanation of the numerical results of Coffey *et al.* [8]. He has accomplished this by demonstrating that in the above range of h the relaxation switches from being dominated by the behavior of the smallest nonvanishing eigenvalue, i.e., the Kramers escape rate [7], to being dominated by the behavior in the wells of the potential. Such an effect appears to be a general feature of relaxation in a bistable potential in the presence of a uniform field. Explicit evaluation of T from the five-term differential-recurrence relation associated with the problem would be very difficult due to the task of identifying the set of hypergeometric functions associated with the solution of these recurrence relations in the zero-frequency limit. Thus the Sturm-Liouville equation constitutes a powerful method of finding analytic solutions in the case of axial symmetry. A drawback of this method, however, is that if it is generally very difficult to evaluate the complex susceptibility $\chi(\omega)$ and to extend the solution for T to nonaxially symmetric problems as in both cases, it is impossible to integrate the equations by quadratures in the manner that leads to Eq. (38). The method based on the solution of differential-recurrence relations used by Coffey *et al.* [7–10] has the advantage that the solution for $\chi(\omega)$ may often be obtained exactly either in continued fraction [18] or matrix continued-fraction form and in cases where this is not feasible [19], as in many nonaxially symmetric problems, by computerized matrix inversion. Accurate solution of nonaxially symmetric problems is of particular importance in the constant magnetic-field effect considered by Coffey *et al.* [8] and Garanin [11] because application of the bias field at an oblique angle will in general cause this effect to manifest itself at smaller-h values than those when the field is collinear with the anisotropy axis.

We finally remark that our formula [Eq. (38)] for the correlation time may also be derived using the method of Szabo

[20] for the calculation of correlation times of autocorrelation functions of the Legendre polynomials, which is based on a generalization of the theory of first-passage times [21,22]. Furthermore, an equation similar to Eq. (38) but confined to one-dimensional translational Brownian motion has been given by Risken [22].

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