Constant-magnetic-field effect in Néel relaxation of single-domain ferromagnetic particles

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The relaxation behavior of an assembly of noninteracting single-domain ferromagnetic particles in the presence of a constant magnetic field is studied by solving the corresponding Fokker-Planck equation. The analysis is performed by first converting that equation into a hierarchy of differential-recurrence relations by expanding the solution in Legendre polynomials. The spectrum of eigenvalues and their associated amplitudes is then determined by matrix methods where all the desired physical quantities such as the magnetization correlation time and complex magnetic susceptibility may be computed numerically. In order to ensure the accuracy of the results obtained this solution is compared with an exact solution derived in terms of matrix continued fractions. It is shown that the conventional assumption in the theory of superparamagnetism, that except in the very early stages of relaxation to equilibrium the only appreciable time constant is the one associated with the smallest nonvanishing eigenvalue, is no longer true when an applied constant magnetic field exceeds a certain critical value. The breakdown of this assumption manifests itself in (a) a dramatically large deviation of the magnetization correlation time (area under the curve of the decay of the magnetization) from the inverse of the lowest eigenvalue, and (b) in the presence of relatively strong high-frequency modes superimposed on the Néel one usually assigned to the lowest eigenvalue. The results are compared with available experimental data.

I. INTRODUCTION

A single-domain ferromagnetic particle with uniaxial anisotropy is characterized by an internal magnetic potential which 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 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), $\lambda_1 \ll \lambda_k$, $k \geq 2$, and $A_1 \gg A_k$ since then the decay functions $A_k \exp(-\lambda_1 t/\tau_N)$, $k \geq 2$ are small compared to $A_1 \exp(-\lambda_1 t/\tau_N)$ except in the very early stages of the approach to equilibrium. The diffusional relaxation time τ_N is defined as[2]

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

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

$$\dot{\mathbf{M}} = \gamma \mathbf{M} \times [\mathbf{H}_T - \eta \dot{\mathbf{M}}] . \tag{2}$$

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

$$\mathbf{H}_T = \mathbf{h} - \partial V/\partial \mathbf{M} , \tag{3}$$

where $\mathbf{h}$ 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

$$T \approx \frac{\tau_N}{\lambda_1} . \tag{4}$$

Very recently, the relaxation behavior has been reexamined by Coffey *et al.*[5] for the simple uniaxial potential of the crystalline anisotropy

$$V(\vartheta) = K \sin^2\vartheta . \tag{5}$$

This is an axially symmetric bistable potential with anisotropy constant K representing the free energy per unit volume of a particle. The stable configurations of the magnetization $\mathbf{M}$ are at $\vartheta = 0$ and $\vartheta = \pi$ where the orientation for $\mathbf{M}$, on a sphere of radius M_s is specified by the spherical polar coordinates ϑ and ϕ, ϑ being the polar angle. The calculation of the decay of the longitudinal component of the magnetization following the removal of a weak constant applied field $\Delta\mathbf{H}$ superimposed on the field $\mathbf{H}$ then amounts to the calculation of A_k and λ_k in the equation

$$M_z(t) = mNf_1(t) = mN\langle \cos\vartheta \rangle = \frac{m^2 N \Delta H}{kT} \sum_k A_k e^{-\lambda_k t/\tau_N} . \tag{6}$$

Here N is the number of particles per unit volume,

$$m = M_s v \tag{7}$$

is the magnetic moment of a particle. According to linear-response theory the decay function $f_1(t)$ from Eq. (6) is connected with the equilibrium longitudinal autocorrelation function of the magnetization $C_1(t)$ as follows:

$$\frac{f_1(t)}{f_1(0)} = C_1(t) = \frac{\langle \cos\vartheta(0)\cos\vartheta(t)\rangle_0 - \langle \cos\vartheta(0)\rangle_0^2}{\langle \cos^2\vartheta(0)\rangle_0 - \langle \cos\vartheta\rangle_0^2} , \tag{8}$$

where the symbol $\langle\ \rangle_0$ means the equilibrium ensemble average, so that the magnetization correlation time $T_\parallel$ according to Eq. (6) is given by

$$T_\parallel = \int_0^\infty C_1(t)dt = \tau_N \frac{\sum_k A_k \lambda_k^{-1}}{\sum_k A_k} . \tag{9}$$

It should be noted that in our notation λ_k differs by a factor of 2 from that used in Refs. 2–4. Coffey *et al.*[5] derived an exact analytic equation for the correlation time $T_\parallel$. Also, by calculating successive λ_k and A_k Coffey *et al.*[5] were readily able to show (cf. their Table I) that the relaxation process under the influence of the potential of Eq. (5) is accurately represented by the first decay mode. Thus, Eq. (4) holds accurately in this case (Table II of Ref. 5).

The second type of uniaxial potential which is of interest in the study of superparamagnetism is when a constant field $\mathbf{H}$ of arbitrary strength is superimposed on the anisotropy potential.[1–4] In general such a field can only be applied at some angle to the easy axis of magnetization since that axis is in a random position. However, in order to preserve the axial symmetry of the problem and its attendant mathematical simplifications we shall suppose as in Refs. 1–4 that the field is applied along the polar axis so that the potential V is of the form

$$\frac{vV(\vartheta)}{kT} = \sigma \sin^2\vartheta - \xi\cos\vartheta = \sigma(\sin^2\vartheta - 2h\cos\vartheta) , \tag{10}$$

where the barrier height parameter

$$\sigma = \frac{Kv}{kT} \tag{11}$$

and the external field parameter

$$\xi = \frac{vHM_s}{kT} \tag{12}$$

with

$$h = \frac{\xi}{2\sigma} . \tag{13}$$

This potential was originally introduced by Néel[1] who gave an expression for the time of reversal of the magnetization using the discrete orientation approximation. It was further studied by Brown[2] who obtained approximate expressions for the lowest nonvanishing eigenvalue in the limit of large and small σ using the Kramers transition state method[6] and perturbation theory, respectively. Later λ_1^{-1} was calculated numerically by Aharoni.[4] However, he did not calculate any other λ^s or the associated amplitudes including that of λ_1^{-1} thus it was not possible to ascertain the role they play in the relaxation process, nor is it possible to test the accuracy of the single-mode approximation. On the other hand the analysis presented by Garanin, Ischenko, and Panina[7] enabled them to derive an integral expression for the correlation time $T_\parallel$ from the Sturm-Liouville equation. However, rather than trying to calculate $T_\parallel$ exactly from their equation, they presented various asymptotic formulas for $T_\parallel$ and the complex susceptibility.

It is the purpose of this paper to study in detail the relaxation behavior of the system under consideration. In order to accomplish this, we shall calculate accurately the eigenvalues $\{\lambda_k\}$ and the corresponding amplitudes $\{A_k\}$, the correlation time $T_\parallel$ and the complex susceptibility for the potential of Eq. (10) using the methods we have described in Refs. 5 and 8–10. The most important results of the calculation are (a) that the correlation time has behavior dramatically different from λ_1^{-1} above certain critical values of the parameters σ and h and (b) the existence of high-frequency relaxation modes in addition to that arising from the low-frequency one associated with the reversal of $\mathbf{M}$. This is due to the fact that the high-frequency modes make a distinct contribution to the response.

II. DIFFERENTIAL-RECURRENCE RELATIONS FOR RELAXATION IN THE PRESENCE OF A CONSTANT FIELD

The Fokker-Planck equation for the probability density $W(\vartheta,\phi,t)$ of orientations of the magnetization $\mathbf{M}$ on the unit sphere for an axially symmetric potential $V(\vartheta)$ is[2]

$$2\tau_N \frac{\partial W}{\partial t} = \frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left[\sin\vartheta\left[\frac{\partial W}{\partial\vartheta}+\frac{\nu}{kT}\frac{\partial V}{\partial\vartheta}W\right]\right] + \frac{1}{\sin\vartheta}\frac{\partial}{\partial\phi}\left\{\frac{\nu}{kT}\left[\frac{\partial V}{\alpha\partial\vartheta}\right]W+\frac{1}{\sin\vartheta}\frac{\partial W}{\partial\phi}\right\}, \tag{14}$$

where

$$\alpha=\eta\gamma M_s \tag{15}$$

is a dimensionless damping parameter. In order to study the longitudinal relaxation behavior we suppose that the constant field term in the potential of Eq. (10) is altered by a small amount $\xi_1=m\Delta H/kT\ll 1$ at an initial time $t=0$ so that we determine the after-effect solution of Eq. (14). We can disregard the dependence of W on ϕ for the longitudinal relaxation; hence we may assume that the distribution function W is

$$W(\vartheta,t)=\sum_{l=0}^{\infty} a_l(t)P_l(\cos\vartheta)\ , \tag{16}$$

where the P_l are the Legendre polynomials. On substituting Eq. (16) into Eq. (15) we obtain the differential-recurrence relation

$$\frac{2\tau_N}{l(l+1)}\dot{f}_l(t)+\left[1-\frac{2\sigma}{(2l-1)(2l+3)}\right]f_l(t) = \frac{\xi}{2l+1}[f_{l-1}(t)-f_{l+1}(t)] + \frac{2\sigma(l-1)}{(2l+1)(2l-1)}f_{l-2}(t) - \frac{2\sigma(l+2)}{(2l+1)(2l+3)}f_{l+2}(t)\ , \tag{17}$$

where

$$f_l(t)=\langle P_l(\cos\vartheta)\rangle-\langle P_l(\cos\vartheta)\rangle_0=\frac{a_l(t)-a_l(\infty)}{(2l+1)a_0} \tag{18}$$

and the symbols $\langle\ \rangle$ and $\langle\ \rangle_0$ mean ensemble average and equilibrium ensemble average, respectively.

We are interested in the decay of the magnetization $M_z(t)$ which in this case is

$$M_z(t)=mN[\langle\cos\vartheta\rangle-\langle\cos\vartheta\rangle_0] =mN\int_0^{\pi}(\cos\vartheta-\langle\cos\vartheta\rangle_0)W(\vartheta,t)\sin\vartheta d\vartheta =mNf_1(t)\ , \tag{19}$$

so that we are required to calculate $f_1(t)$. This is considerably more involved than the case $\xi=0$ because Eq. (17) does not decouple into separate sets for even and odd $f_1(t)$. Furthermore, it is a five-term recurrence relation so that it is not obvious how its solution may be found in the form of a scalar continued fraction. It may however, be cast into the form of a three-term matrix recurrence relation as in Ref. 10.

The initial conditions for $f_l(0)$ may be determined as follows:

$$f_l(0)=\frac{\int_{-1}^{+1}e^{\sigma x^2+(\xi+\xi_1)x}P_l(x)dx}{\int_{-1}^{+1}e^{\sigma x^2+(\xi+\xi_1)x}dx}-\langle P_l(x)\rangle_0\ , \tag{20}$$

which in the linear approximation in the perturbation ξ_1 reduces to

$$f_l(0)=\xi_1[\langle xP_l(x)\rangle_0-\langle x\rangle_0\langle P_l(x)\rangle_0] \tag{21}$$

or

$$f_l(0)=\xi_1\left[\frac{l+1}{(2l+1)}\langle P_{l+1}\rangle_0+\frac{l}{2l+1}\langle P_{l-1}\rangle_0 -\langle P_1\rangle_0\langle P_l\rangle_0\right] \tag{22}$$

or

$$f_l(0)=\xi_1\left[\frac{l+1}{2l+1}f^0_{l+1}+\frac{l}{2l+1}f^0_{l-1}-f^0_1f^0_l\right]\ , \tag{23}$$

where

$$f^0_l(0)=\langle P_l\rangle_0\ .$$

The equilibrium quantities f^0_l satisfy the set of equations

$$\left[1-\frac{2\sigma}{(2l-1)(2l+3)}\right]f^0_l=\frac{\xi}{2l+1}[f^0_{l-1}-f^0_{l+1}] +\frac{2\sigma(l-1)}{(2l+1)(2l-1)}f^0_{l-2} -\frac{2\sigma(l+2)}{(2l+1)(2l+3)}f^0_{l+2}\ , \tag{24}$$

which is the set of Eq. (17) with $\dot{f}_l=0$.

The recurrence relation of Eq. (24) can be evaluated for any l if we have a knowledge of $\langle P_0\rangle_0$, $\langle P_1\rangle_0$, and $\langle P_2\rangle_0$. These may be given just as in Ref. 10 by using the results of p. 369 of Ref. 11. We have

$$\langle P_0\rangle_0=1\ , \tag{25}$$

$$\langle P_1\rangle_0=\frac{\xi}{T(\xi,\sigma)}-\frac{\xi}{2\sigma}\ , \tag{26}$$

$$\langle P_2\rangle_0=\frac{3}{2}\left[\frac{\xi L(\xi)+1-\xi^2/2\sigma}{T(\xi,\sigma)}+\frac{\xi^2/2\sigma-1}{2\sigma}\right]-\frac{1}{2}\ , \tag{27}$$

where

$$L(\xi)=\coth\xi-\frac{1}{\xi} \tag{28}$$

is the Langevin function and the function $T(\xi,\sigma)$ is given in terms of Dawson's integral[11,12]

$$D(x)=e^{-x^2}\int_0^x e^{t^2}dt$$

of given argument as

$$T(\xi,\sigma)=\sqrt{\sigma}\left[[\xi L(\xi)+1+\xi]D\left[\sqrt{\sigma}+\frac{\xi}{2\sqrt{\sigma}}\right]+[\xi L(\xi)+1-\xi]D\left[\sqrt{\sigma}-\frac{\xi}{2\sqrt{2\sigma}}\right]\right] . \tag{29}$$

The set of equations (17) may be solved to yield the relaxation behavior of $f_1(t)$ in either of two ways. The first is to arrange them in the form

$$\dot{\mathbf{X}}(t)=\mathbf{A}\mathbf{X}(t) , \tag{30}$$

and the second is to arrange the differential-recurrence relation as a three-term matrix one, the Laplace transform of which may then be obtained analytically in terms of a matrix continued fraction as described for the corresponding two-dimensional problem in Ref. 10.

III. SOLUTION OF THE SET OF RECURRENCE RELATIONS (17), FORMULATION AS A MATRIX DIFFERENTIAL EQUATION

The first method proceeds as follows. In Eq. (30) the column vector $\mathbf{X}(t)$ is

$$\mathbf{X}(t)=\begin{bmatrix} f_1(t) \\ f_2(t) \\ \cdot \\ \cdot \\ f_l(t) \\ \cdot \\ \cdot \end{bmatrix} . \tag{31}$$

The system matrix $\mathbf{A}$ is determined by Eq. (17) and is given by

$$\mathbf{A}=-\frac{1}{\tau_N}\begin{bmatrix} 1-\frac{2}{5}\sigma & \frac{\xi}{3} & \frac{2}{5}\sigma & 0 & 0 & 0 & \cdots \\ -\frac{3}{5}\xi & 3-\frac{2}{7}\sigma & \frac{3}{5}\xi & \frac{24}{35}\sigma & 0 & 0 & \cdots \\ -\frac{24}{35}\sigma & -\frac{6}{7}\xi & 6-\frac{4}{15}\sigma & \frac{6}{7}\xi & \frac{20}{21}\sigma & 0 & \cdots \\ 0 & -\frac{20}{21}\sigma & -\frac{10}{9}\xi & 10-\frac{20}{77}\sigma & \frac{10}{9}\xi & \frac{40}{33}\sigma & \cdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \end{bmatrix} . \tag{32}$$

In Eq. (17), l is taken large enough (equal to L say) to ensure convergence of the set of Eqs. (30). The lowest nonvanishing eigenvalue which corresponds to the reciprocal of the greatest relaxation time, is then the smallest root of the characteristic equation

$$\det(\lambda\mathbf{I}-\mathbf{A})=0 . \tag{33}$$

The relaxation modes of $f_1(t)$ may be found from Eq. (30) by assuming that[13] $\mathbf{A}$ has a linearly independent set of L eigenvectors $(\mathbf{R}_1,\ldots,\mathbf{R}_L)$, so that[13,14]

$$\mathbf{X}(t)=b_1e^{\lambda_1 t}\mathbf{R}_1+b_2e^{\lambda_2 t}\mathbf{R}_2\cdots+b_Le^{\lambda_L t}\mathbf{R}_L , \tag{34}$$

where the b_i are to be determined from the initial conditions (23). Equations (30) may now be solved to any desired degree of accuracy to yield the decay of the longitudinal component of the magnetization according to Eq. (6), namely

$$mNf_1(t)=mN[\langle\cos\vartheta\rangle-\langle\cos\vartheta\rangle_0]=\frac{m^2\Delta HN}{kT}\sum_{k=1}^{\infty}A_ke^{-\lambda_k t/\tau_N} . \tag{35}$$

The corresponding normalized complex susceptibility

$$\chi_\parallel(\omega)=\chi_\parallel'(\omega)-i\chi_\parallel''(\omega)=G\alpha_\parallel(\omega) , \tag{36}$$

where

$$G=\frac{m^2N}{3kT} , \tag{37}$$

is then given by

$$\alpha_\parallel(\omega)=\sum_{k=1}^{\infty}\frac{3A_k}{1+i\omega\tau_N/\lambda_k} . \tag{38}$$

Here we have made use of the linear-response theory formula[15]

$$\alpha_\parallel(\omega)=\alpha_\parallel'(\omega)-i\alpha_\parallel''(\omega)=\alpha_\parallel'(0)\left[1-i\omega\int_0^{\infty}\frac{f_1(t)}{f_1(0)}e^{-i\omega t}dt\right] . \tag{39}$$

The correlation time $T_\parallel$ in terms of the A_k and λ_k is given by Eq. (9).

We note that the correlation time $T_\parallel$ itself may be evaluated more simply by noting that as in Ref. 5

$$T_\parallel=\lim_{s\to 0}\int_0^{\infty}C_1(t)e^{-st}dt=\tilde{C}_1(0)=\frac{\tilde{f}_1(0)}{f_1(0)} \tag{40}$$

with the tilde denoting the Laplace transform where $\tilde{f}_1(0)$ is determined from the matrix equation[5]

$$\tilde{\mathbf{X}}(0)=-\mathbf{A}^{-1}\mathbf{X}(0)\ , \tag{41}$$

where $\mathbf{X}(0)$ is the initial value vector that is

$$\mathbf{X}(0)=\begin{bmatrix} f_1(0) \\ f_2(0) \\ f_3(0) \\ \vdots \end{bmatrix} .$$

Equation (41) is obtained from the Laplace transform of Eq. (30) for $s=0$.

We have formulated the problem of determining the initial values $f_1(0)$ from the recurrence relation with the first three f_l^0 given as in Eqs. (25)–(29). The f_l^0 may however, be evaluated numerically by simply calculating $\mathbf{A}^{-1}$. First we note that the set of Eq. (24) constitutes the *inhomogeneous set*

$$\mathbf{A}\mathbf{F}^0=\mathbf{B}\ , \tag{42}$$

with

$$\mathbf{F}^0=\begin{bmatrix} f_1^0 \\ f_2^0 \\ \vdots \\ f_l^0 \\ \vdots \end{bmatrix} \tag{43}$$

and

$$\mathbf{B}=\begin{bmatrix} \frac{\xi}{3} \\ \frac{2\sigma}{15} \\ 0 \\ 0 \\ \vdots \end{bmatrix} , \tag{44}$$

where we have taken into account the fact that $f_0^0=1$. Thus we can calculate numerically all f_l^0 needed from the equation

$$\mathbf{F}^0=\mathbf{A}^{-1}\mathbf{B}\ . \tag{45}$$

This is the formulation of the solution of Eq. (17) as the solution of the matrix differential equation (30).

IV. SOLUTION OF EQ. (17) IN TERMS OF MATRIX CONTINUED FRACTIONS

The advantage of posing the problem in such a manner is that an exact formula in terms of matrix continued fractions may be written for the Laplace transform of the after-effect function. Furthermore, the correlation time and the complex susceptibility may also be written as matrix continued fractions. The starting point of the calculation is the matrix differential-recurrence relation[13]

$$\dot{\mathbf{C}}_l(t)=\mathbf{Q}_l^-\mathbf{C}_{l-1}(t)+\mathbf{Q}_l\mathbf{C}_l(t)+\mathbf{Q}_l^+\mathbf{C}_{l+1}(t)\ , \tag{46}$$

where the $\mathbf{Q}_l^{\pm}$, $\mathbf{Q}_l$ are time independent $l\times l$ matrices and the $\mathbf{C}_l(t)$ are time-dependent column vectors. Equation (17) takes the form of the matrix three-term differential-recurrence relation Eq. (46) if we rearrange it as follows:

$$\begin{bmatrix} \dot{f}_{2l-1}(t) \\ \dot{f}_{2l}(t) \end{bmatrix}=\frac{1}{\tau_N}\begin{bmatrix} \frac{4\sigma(l-1)(2l-1)l}{(4l-1)(4l-3)} & \frac{\xi(2l-1)l}{(4l-1)} \\ 0 & \frac{2\sigma(2l-1)(2l+1)l}{(4l+1)(4l-1)} \end{bmatrix}\begin{bmatrix} f_{2l-3}(t) \\ f_{2l-2}(t) \end{bmatrix}$$
$$+\frac{1}{\tau_N}\begin{bmatrix} l(2l-1)\left[\frac{2\sigma}{(4l-3)(4l+1)}-1\right] & \frac{-\xi l(2l-1)}{(4l-1)} \\ \frac{\xi(2l+1)l}{(4l+1)} & l(2l+1)\left[\frac{2\sigma}{(4l-1)(4l+3)}-1\right] \end{bmatrix}\begin{bmatrix} f_{2l-1}(t) \\ f_{2l}(t) \end{bmatrix}$$
$$+\frac{1}{\tau_N}\begin{bmatrix} \frac{-2\sigma(2l+1)(2l-1)l}{(4l-1)(4l+1)} & 0 \\ \frac{-\xi(2l+1)l}{(4l+1)} & \frac{-4\sigma(l+1)(2l+1)l}{(4l+1)(4l+3)} \end{bmatrix}\begin{bmatrix} f_{2l+1}(t) \\ f_{2l+2}(t) \end{bmatrix} . \tag{47}$$

A general method of solution of Eq. (46) in terms of matrix continued fractions was given by Risken.[13] Another approach which has the merit of being considerably more simple than the previously available algorithm has been described in Ref. 10. Thus Eq. (47) may be solved for the Laplace transform

$$\tilde{\mathbf{C}}_1(s)=\begin{bmatrix} \tilde{f}_1(s) \\ \tilde{f}_2(s) \end{bmatrix}$$

using the method of Ref. 10 to yield

$$\tilde{\mathbf{C}}_1(s)=[s\mathbf{I}-\mathbf{Q}_1^--\mathbf{Q}_1^+\mathbf{S}_2(s)]^{-1}\left\{\mathbf{C}_1(0)+\sum_{n=2}^{\infty}\prod_{k=2}^{n}\mathbf{Q}_{k-1}^+\tilde{\mathbf{S}}_2(s)(\mathbf{Q}_k^-)^{-1}\mathbf{C}_n(0)\right\} , \tag{48}$$

where the 2×2 matrices $\mathbf{Q}_n^{\pm}$, $\mathbf{Q}_n$ are as shown in Eq. (47), $\tilde{\mathbf{S}}_n(s)$ is the 2×2 matrix continued fraction defined as

$$\tilde{\mathbf{S}}_n(s)=(s\mathbf{I}-\mathbf{Q}_n-\mathbf{Q}_n^{+}\tilde{\mathbf{S}}_{n+1}(s))^{-1}\mathbf{Q}_n^{-} . \tag{49}$$

The initial value vectors

$$\mathbf{C}_n(0)=\begin{bmatrix} f_{2n-1}(0) \\ f_{2n}(0) \end{bmatrix}$$

are most conveniently determined from Eqs. (23) and (24) in this case. The after-effect function $\tilde{f}_1(s)$ is simply calculated from the scalar product of $\tilde{\mathbf{C}}_1(s)$ with the unit vector $\mathbf{e}=(1,0)$ namely

$$\tilde{f}_1(s)=[\tilde{\mathbf{C}}_1(s)\cdot\mathbf{e}] , \tag{50}$$

while the correlation time $T_{\parallel}$ and the complex susceptibility $\chi_{\parallel}(\omega)$ are given as before by Eqs. (40) and (39), respectively. Equations (48)–(50) constitute the exact solution of the problem in terms of matrix continued fractions.

V. RESULTS AND COMPARISON WITH EXPERIMENTAL DATA

In Fig. 1 and Table I we show the behavior of the lowest eigenvalue λ_1 calculated from the characteristic Eq. (33) as a function of σ for various values of h showing that our calculation agrees with the corresponding Fig. 1 of Aharoni[4] in all respects. A 45×45 matrix $\mathbf{A}$ was sufficient to obtain a value of λ_1^{-1} accurate to three significant digits for the range of parameters chosen. In Fig. 2 and Table II we show the behavior of the correlation time $T_{\parallel}$ computed from the matrix formula Eq. (41) for the same values of h. The most astonishing result of this calculation is that $T_{\parallel}^{-1}$ differs greatly from λ_1^{-1} above a certain critical value of h. Again a 45×45 matrix yielded satisfactory results. For example, at $h=0.2$ and $\sigma=20$, $T_{\parallel}=13.6$ and $\lambda_1^{-1}=1.01\times10^4$. This behavior is further emphasized in Table III where one can easily see that λ_1^{-1} differs dramatically from $T_{\parallel}$. Thus the correlation time is far more sensitive to any alteration of the two potential well structure of Eq. (5) caused by the imposition of the field ξ than is λ_1^{-1}. We were first alerted to this behavior by calculating $T_{\parallel}$ using the matrix formula of Eq. (41) which merely requires a knowledge of the initial value vector $\mathbf{X}(0)$ and $\mathbf{A}^{-1}$. More insight into the disparity between λ_1^{-1} and $T_{\parallel}$ may be gained by calculating $T_{\parallel}$ using Eq. (9). This requires a knowledge of a sufficiently large set of the eigenvalues $\{\lambda_k\}$ and their corresponding amplitudes $\{A_k\}$ and is shown in Table IV. The reason for the disparity between $T_{\parallel}$ and λ_1^{-1} now becomes obvious. It is due to the fact that at short to intermediate times the high-frequency decay modes cannot be neglected as they contribute significantly to the correlation time. Indeed it is apparent by using the values of Table IV that Eq. (9) for the correlation time $T_{\parallel}$ cannot be reduced to Eq. (4) in contrast to the case $\xi=0$ where the A_k are negligible for all $k>1$ (see Table I of Ref. 5) so that

$$\frac{\sum A_k\lambda_k^{-1}}{\sum A_k}\approx\frac{1}{\lambda_1} . \tag{51}$$

The results are corroborated by the matrix continued fraction method of Sec. IV.

By way of further illustration of our results we show in Table V the behavior of the first ten decay modes $A_k\exp(-\lambda_k t/\tau_N)$ for various values of the time t ranging from 0.01 to 10. The general structure of the decay

TABLE I. The lowest nonvanishing eigenvalue λ_1 for various values of the barrier height (σ) and field (h) parameters.

	$2\lambda_1$						
h	$\sigma=0.2$	$\sigma=0.5$	$\sigma=1$	$\sigma=2$	$\sigma=5$	$\sigma=10$	$\sigma=20$
0.01	1.84	1.63	1.31	0.808	0.136	0.002 93	4.24×10^{-7}
0.1	1.84	1.63	1.31	0.832	0.179	0.008 6	7.79×10^{-6}
0.2	1.85	1.63	1.34	0.906	0.313	0.038 3	0.000 198
0.4	1.85	1.66	1.43	1.2	0.91	0.385	0.032 7
0.5	1.85	1.68	1.49	1.41	1.4	0.883	0.206
0.7	1.86	1.72	1.67	1.97	2.78	2.93	2.27
0.8	1.86	1.75	1.78	2.32	3.67	4.52	4.78
1	1.88	1.82	2.05	3.12	5.82	8.78	12.9

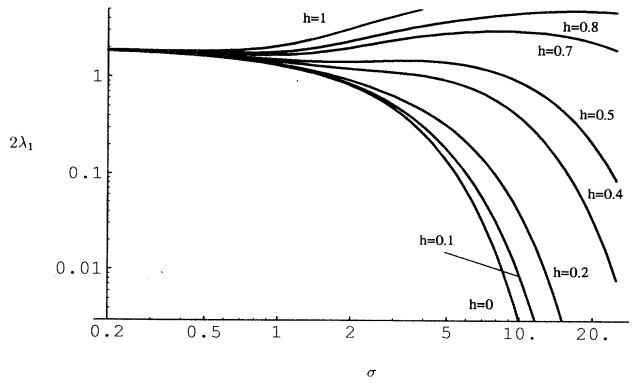

FIG. 1. The lowest nonvanishing eigenvalue λ_1 as a function of the barrier height parameter σ for various values of the field parameter h.

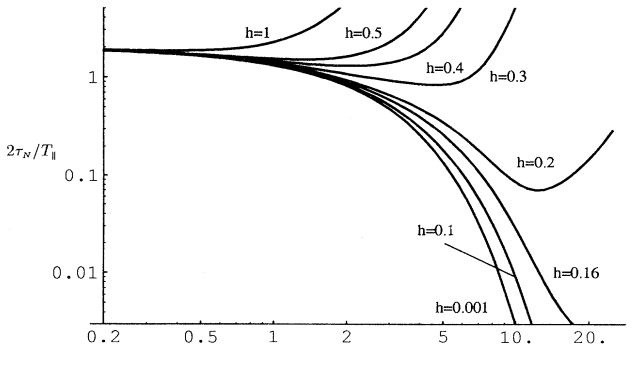

FIG. 2. Inverse of the correlation time $T_{\parallel}$ as a function of the barrier height parameter σ for various values of the field parameter h.

TABLE II. Inverse of the correlation time $T_{\parallel}$ for various values of the barrier height (σ) and field (h) parameters.

	$2/T_{\parallel}$						
h	$\sigma=0.2$	$\sigma=0.5$	$\sigma=1$	$\sigma=2$	$\sigma=5$	$\sigma=10$	$\sigma=20$
0.01	1.84	1.63	1.31	0.813	0.138	0.002 94	4.24×10^{-7}
0.1	1.84	1.63	1.32	0.84	0.183	0.008 92	0.000 010 7
0.2	1.85	1.64	1.34	0.925	0.35	0.084 2	0.147
0.4	1.85	1.66	1.45	1.29	2.74	43.3	104
0.5	1.85	1.68	1.53	1.61	9.16	51.8	112
0.7	1.86	1.74	1.74	2.65	24.6	60.8	129
0.8	1.87	1.77	1.89	3.45	28	65.1	137
1	1.88	1.85	2.24	5.73	33.2	73.7	154

modes now becomes apparent. In this case ($\sigma=10$) for small values of $t<0.1$ the high-frequency mode characterized by λ_5 makes a *greater* contribution to the decay than that of λ_1 however, as time progresses (see Fig. 3) this and all other modes vanish so that only the λ_1 mode—which is, as usual, that associated with the stability of a given distribution of magnetization—Néel relaxation remains. It is also obvious from Fig. 3 that the general behavior of the correlation function $C_1(t)$ differs markedly from that of the first mode decay function $\exp(-\lambda_1 t/\tau_N)$. The dominance of the high-frequency modes at short and intermediate times has important consequences in the frequency domain, where a high-frequency absorption peak will appear in the imaginary part $\chi_{\parallel}''(\omega)$ of the complex susceptibility along with the usual low-frequency absorption maximum associated with λ_1. This behavior is illustrated in Figs. 4 and 5 for $\sigma=5$ and 10 for various values of h. One can see from these figures that the high-frequency mode exists even at $h=0.01$. It should be noted that the existence of a weak high-frequency mode in the case of the potential (5) was first noted by Martin, Meier, and Saupe[16] for the similar problem of dielectric relaxation of nematic liquid crystals.

The discussion so far has centered on the numerical solution of the problem. We shall now describe how this solution may be related to previous analytical results.

TABLE III. Product of the lowest eigenvalue λ_1 and the correlation time $T_{\parallel}$ for various values of the barrier height (σ) and field (h) parameters.

	$T_{\parallel}\lambda_1$						
h	$\sigma=0.2$	$\sigma=0.5$	$\sigma=1$	$\sigma=2$	$\sigma=5$	$\sigma=10$	$\sigma=20$
0.01	1	0.999	0.998	0.994	0.988	0.996	0.999
0.1	1	0.999	0.997	0.99	0.974	0.964	0.725
0.2	1	0.999	0.995	0.979	0.896	0.455	0.001 34
0.4	1	0.997	0.986	0.926	0.332	0.008 87	0.000 315
0.5	0.999	0.996	0.979	0.878	0.153	0.017 1	0.001 83
0.7	0.999	0.992	0.959	0.746	0.113	0.048 2	0.017 6
0.8	0.999	0.99	0.945	0.672	0.131	0.069 5	0.034 8
1	0.998	0.984	0.913	0.544	0.176	0.119	0.084

Thus Aharoni[4] has shown that for $h\leq0.4$ and $\sigma\geq2$ the lowest eigenvalue λ_1 can be approximated by Brown's formula[2]

$$\lambda_1=\pi^{-1/2}\sigma^{3/2}(1-h^2)\{(1+h)\exp[-\sigma(1+h)^2]+(1-h)\exp[-\sigma(1-h)^2]\} \quad (52)$$

or by noting that $\xi=2\sigma h$ (Ref. 17):

$$\lambda_1=2\pi^{-1/2}\sigma^{3/2}[1-\xi^2/4\sigma^2]\{\cosh\xi-(\xi/2\sigma)\sinh\xi\}\times\exp(-\sigma-\xi^2/4\sigma^2)\ . \quad (53)$$

Thus we can also use the analytic equation (53) for evaluation of the correlation time $T_{\parallel}$ from Eq. (4). For larger h and smaller σ this formula is inadequate (see Table III). However in this case we can use another approximate equation, which has been already derived in Ref. 11, viz., the inverse of the effective eigenvalue.[17] The effective eigenvalue λ_{ef} is determined by evaluating Eq. (17) for $l=1$ at $t=0$:

$$\lambda_{ef}=-\frac{\dot{f}_1(0)}{f_1(0)}\ . \quad (54)$$

Thus on using Eqs. (17), (23), and (25)–(27) we can obtain an analytic equation for the effective correlation time $T_{\parallel}^{ef}=\lambda_{ef}^{-1}$ in terms of the Langevin and Dawson functions, namely[11]

$$T_{\parallel}^{ef}=2\tau_N\frac{2\sigma/T(\xi,\sigma)(\xi L(\xi)+1+\xi^2/2\sigma)-1-2\sigma\xi^2/T^2(\xi,\sigma)}{2\sigma-2\sigma/T(\xi,\sigma)[\xi L(\xi)+1-\xi^2/2\sigma]-\xi^2/2\sigma)+1}\ , \quad (55)$$

TABLE IV. Amplitudes A_k and eigenvalues λ_k of the first ten modes of the decay of the longitudinal magnetization for $h=0.01$, 0.2, and 0.4 and $\sigma=10$.

	$h=0.01$		$h=0.2$		$h=0.4$	
k	A_k	λ_k	A_k	λ_k	A_k	λ_k
1	0.858	0.001 47	0.0019	0.0192	1.11×10^{-6}	0.19
2	9.05×10^{-8}	8.06	2.01×10^{-6}	7.32	6.68×10^{-8}	7.12
3	0.002 83	12.3	0.000 063 2	12.7	7.86×10^{-7}	14.4
4	0.000 049	14.8	0.001 4	17.6	0.000 41	22.6
5	0.000 807	19.2	0.000 668	20.3	0.001 06	23.6
6	2.18×10^{-6}	25.2	0.000 101	26.6	0.000 029	31.2
7	0.000 036 1	32.1	0.000 029 8	33.4	0.000 022 7	37.7
8	8.78×10^{-8}	40	5.84×10^{-6}	41.2	6.14×10^{-6}	44.8
9	9.45×10^{-7}	49	1.02×10^{-6}	50.1	1.18×10^{-6}	53.6
10	1.92×10^{-9}	58.9	1.5×10^{-7}	60	2.06×10^{-7}	63.5

TABLE V. Effect of the first ten modes on the decay of the magnetization for $t/\tau_N=0.01$, 0.1, and 10 for $h=0.2$ and $\sigma=20$.

t	0.01	0.1	1	10
$A_1e^{-\lambda_1 t}$	6.55×10^{-7}	6.55×10^{-7}	6.55×10^{-7}	6.54×10^{-7}
$A_2e^{-\lambda_2 t}$	9.66×10^{-15}	2.13×10^{-15}	5.81×10^{-22}	1.32×10^{-87}
$A_3e^{-\lambda_3 t}$	2.46×10^{-10}	2.82×10^{-11}	1.11×10^{-20}	1.03×10^{-114}
$A_4e^{-\lambda_4 t}$	2.04×10^{-10}	1.1×10^{-11}	2.27×10^{-24}	3.18×10^{-151}
$A_5e^{-\lambda_5 t}$	3.14×10^{-4}	6.45×10^{-6}	8.6×10^{-23}	1.53×10^{-191}
$A_6e^{-\lambda_6 t}$	2.22×10^{-7}	4.22×10^{-9}	2.55×10^{-26}	1.66×10^{-198}
$A_7e^{-\lambda_7 t}$	9.08×10^{-9}	5.62×10^{-11}	4.61×10^{-33}	6.36×10^{-254}
$A_8e^{-\lambda_8 t}$	8.79×10^{-8}	1.71×10^{-10}	$1.29\times1-^{-37}$	8.04×10^{-309}
$A_9e^{-\lambda_9 t}$	1.29×10^{-6}	1.45×10^{-9}	4.52×10^{-39}	3.99×10^{-334}
$A_{10}e^{-\lambda_{10} t}$	1.23×10^{-7}	6.94×10^{-11}	2.24×10^{-43}	2.77×10^{-368}

where the L and T functions are given by Eqs. (28) and (29), respectively. We compare $T_{\|}^{ef}$ and $T_{\|}$ in Fig. 6 for different values of the field parameters h. One can see from this figure that Eq. (55) provides a good approximation to the magnetization correlation time $T_{\|}$ for $h>0.5$ and all ranges of σ. This is not surprising because we have already shown[9] that the effective eigenvalue method provides in general an acceptable approximation to the correlation time $T_{\|}$ for $\sigma=0$ in contrast to the case $h=0$ when the effective eigenvalue approach is applicable only for low potential barriers ($\sigma<1.5$) and there is exponentially large divergence from the exact solution in the limit of high barriers.[5]

The results obtained for the relaxation time are in agreement with experimental data of Barbara *et al.*,[18] where the field and temperature dependence of the relaxation time were measured for $Tb_{0.5}Ce_{0.5}Fe_2$ particles. The particles had a size distribution about a mean value 150 Å and cubic symmetry. Our uniaxial anisotropy model does not necessarily apply in such a case. However, it seems very likely that it should qualitatively hold for cubic anisotropy as well.[4] The relaxation time τ experimentally[18] measured was directly related to the reciprocal of the slope $(dM/dt)^{-1}$ of the magnetization $M(t)$ curve at $M=0$. Therefore, only the contribution of the longest lived mode to the relaxation rate was measured. Thus, we may evaluate the relaxation time τ from the equation

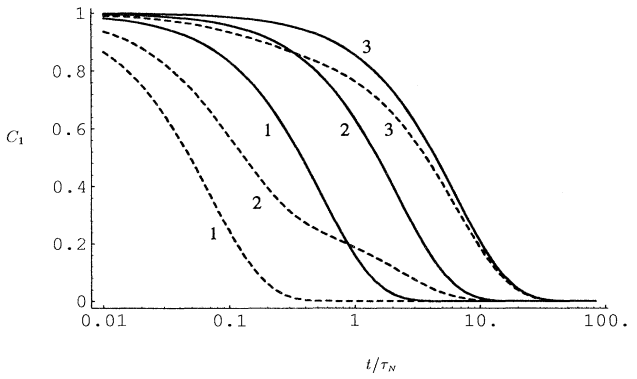

FIG. 3. Comparison of the correlation function $C_1(t)$ (dashed lines) with the single exponential decay function $\exp(-\lambda_1 t/\tau_N)$ (solid lines) for various values of the field parameter h: 0.4, 0.2, and 0.01 (curves 1–3, respectively).

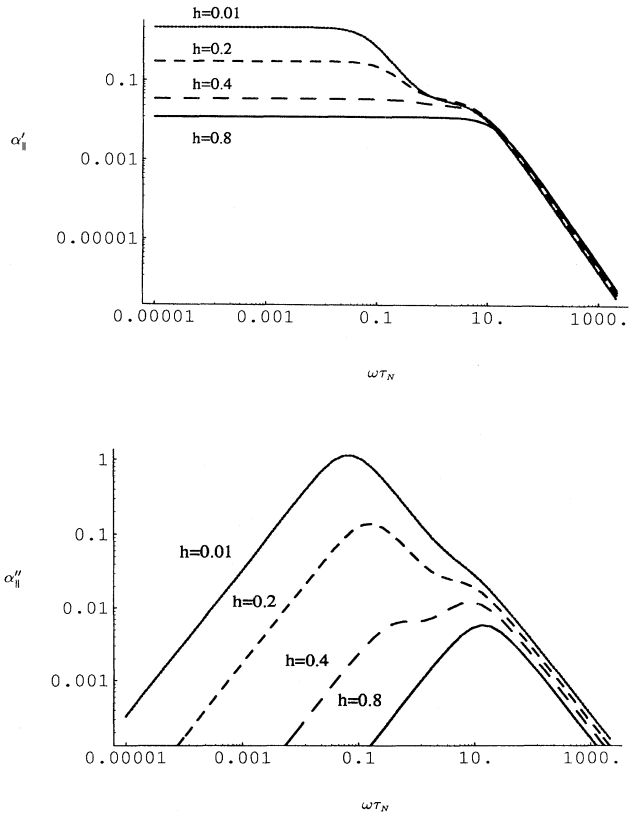

FIG. 4. The real (A) and (B) imaginary parts of the normalized susceptibility $\alpha_{\|}$ as a function of frequency for $\sigma=5$ and $h=0.01$, 0.2, 0.4, and 0.8.

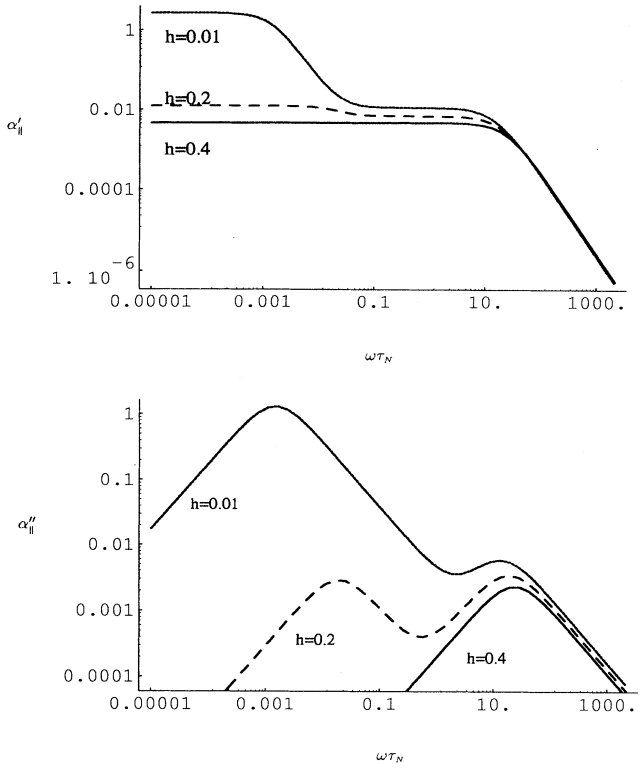

FIG. 5. The same as in Fig. 3 for $\sigma=10$ and $h=0.01$, 0.2, and 0.4.

$$\tau=\frac{\tau_N}{\lambda_1},$$

where λ_1 is given by Eq. (53). The comparison is made in Fig. 7 for two temperatures 8 and 10 K, when the contribution of the quantum tunneling to the relaxation rate may be neglected. As observed by Barbara *et al.*[18] the plot of $\log_{10}(1/T_{\parallel})$ vs $1/H$ at different temperatures as a parameter may be approximated by a family of straight lines with a focal point. Figure 7 shows clearly that these

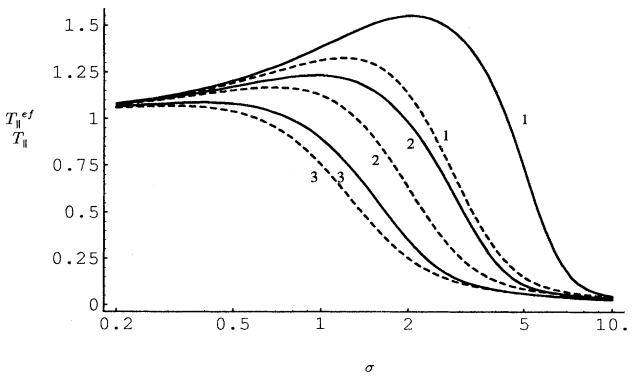

FIG. 6. Co. parison of the correlation time $T_{\parallel}$ (solid lines) and effective correlation time $T_{\parallel}^{ef}$ (dashed lines) for various values of the field parameter $h=0.4$, 0.6, and 1.0 (curves 1–3, respectively).

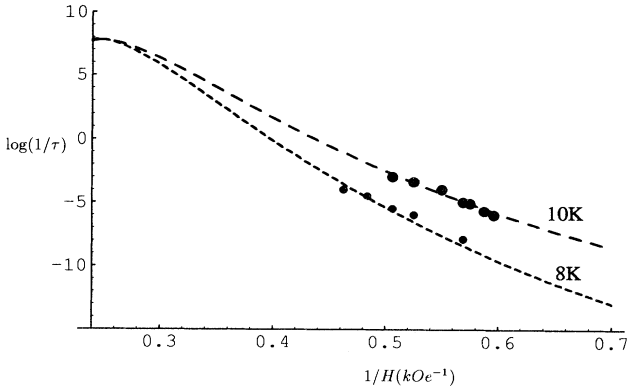

FIG. 7. Comparison of theoretical [dashed lines, Eq. (53)] and experimental [filled circles (Ref. 18)], $\log_{10}(1/T_{11})$ vs $1/H$ at 8 and 10 K in $Tb_{0.5}Cl_{0.5}Fl_2$.

observations are in qualitative agreement with the theoretical results. The values of model parameters σ and h yielded by the best fit are $\sigma=900$ (K)/T and $h=0.222$ (kOe^{-1})H.

VI. CONCLUSIONS

The analysis of Néel relaxation of single-domain magnetic particles has usually proceeded using the assumption of Brown[2] that except in the very early stages of an approach to equilibrium the only appreciable time-dependent term in the solution of the Fokker-Planck equation [Eq. (14)] will be that corresponding to the inverse of the lowest nonvanishing eigenvalue. The statement is certainly true in the case of zero applied field $\xi=0$. However, when $\xi\neq 0$, we have demonstrated that the higher-order decay modes have a significant role to play in the relaxation process. This effect manifests itself in two ways (i) in the large difference between $T_{\parallel}$ and λ_1^{-1} for values of $h>0.15$, (ii) in the existence of a high-frequency loss which for small values of h displays itself as a shoulder in the conventional low-frequency absorption peak for $h=0$ and then predominates as h increases. We remark in conclusion that we have assumed throughout (in accordance with the work described in Refs. 2 and 3) that the field **H** is applied along the easy axis. In practice the easy axis is in a random position so that the calculation must also be carried out when **H** is at an arbitrary angle ψ to that axis. The preliminary results of such a calculation are described in Ref. 19.

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[1]L. Néel, Ann. Géophys. **5**, 99 (1949).
[2]W. F. Brown, Jr., Phys. Rev. **130**, 1677 (1963).
[3]A. Aharoni, Phys. Rev. **135A**, 447 (1964).
[4]A. Aharoni, Phys. Rev. **177**, 793 (1969).
[5]W. T. Coffey, D. S. F. Crothers, Yu. P. Kalmykov, E. S. Massawe, and J. T. Waldron, Phys. Rev. E **49**, 1869 (1994).
[6]H. A. Kramers, Physica **7**, 248 (1940).
[7]D. A. Garanin, V. V. Ischenko, and L. V. Panina, Teor. Mat. Fiz. **82**, 242 (1990) [Theor. Math. Phys. **82**, 169 (1990)].
[8]W. T. Coffey, Yu. P. Kalmykov, E. S. Massawe, and J. T. Waldron, J. Chem. Phys. **99**, 4011 (1993).
[9]J. T. Waldron, Yu. P. Kalmykov, and W. T. Coffey, Phys. Rev. E **49**, 3976 (1994).
[10]W. T. Coffey, Yu. P. Kalmykov, and J. T. Waldron, Physica A **208**, 462 (1994).
[11]W. T. Coffey, P. J. Cregg, and Yu. P. Kalmykov, Adv. Chem. Phys. **83**, 263 (1993).
[12]*Handbook of Mathematical Functions,* edited by M. Abramowitz and I. Stegun (Dover, New York, 1964).
[13]H. Risken, *The Fokker-Planck Equation* (Springer, Berlin, 1984).
[14]M. Krasnov, A. Kiselev, G. Makarenko, and E. Shikin, *Mathematical Analysis for Engineers* (Mir, Moscow 1990), Vol. 2.
[15]B. K. P. Scaife, *Principles of Dielectrics* (Oxford University, Oxford, 1989).
[16]A. J. Martin, G. Meier, and S. Saupe, Symp. Faraday Soc. **5**, 119 (1971).
[17]W. T. Coffey, Yu. P. Kalmykov, and E. S. Massawe, Adv. Chem. Phys. **85**, (part 2), 667 (1993).
[18]B. Barbara, L. C. Sampaio, J. E. Wegrowe, B. A. Ratnam, A. Marchand, C. Paulsen, M. A. Novak, J. L. Tholence, M. Uehara, and D. Fruchart, J. Appl. Phys. **73**, 6703 (1993).
[19]W. T. Coffey, D. S. F. Crothers, J. L. Dormann, L. J. Geoghegan, Yu. P. Kalmykov, J. T. Waldron, and A. W. Wickstead, J. Magn. Magn. Mater. **145**, L263 (1995).