# Exact analytic formula for the correlation time of a single-domain ferromagnetic particle 

W. T. Coffey, ${ }^{1}$ D. S. F. Crothers, ${ }^{2, *}$ Yu. P. Kalmykov, ${ }^{3}$ E. S. Massawe, ${ }^{1}$ and J. T. Waldron ${ }^{4}$<br>${ }^{1}$ Department of Microelectronics and Electrical Engineering, Trinity College, Dublin 2, Republic of Ireland<br>${ }^{2}$ Harvard Smithsonian Center for Astrophysics, 60 Garden Street, Cambridge, Massachusetts 02138<br>${ }^{3}$ Institute of Radio Engineering and Electronics, Russian Academy of Sciences, Vvedenskii Square 1, Fryazino, Moscow Region 141120, Russia<br>${ }^{4}$ School of Computer Applications, Dublin City University, Dublin 9, Republic of Ireland

(Received 21 June 1993; revised manuscript received 1 October 1993)


#### Abstract

Exact solutions for the longitudinal relaxation time $T_{\|}$and the complex susceptibility $\chi_{\|}(\omega)$ of a thermally agitated single-domain ferromagnetic particle are presented for the simple uniaxial potential of the crystalline anisotropy considered by Brown [Phys. Rev. 130, 1677 (1963)]. This is accomplished by expanding the spatial part of the distribution function of magnetic-moment orientations on the unit sphere in the Fokker-Planck equation in Legendre polynomials. This leads to the three-term recurrence relation for the Laplace transform of the decay functions. The recurrence relation may be solved exactly in terms of continued fractions. The zero-frequency limit of the solution yields an analytic formula for $T_{\|}$as a series of confluent hypergeometric (Kummer) functions which is easily tabulated for all potential-barrier heights. The asymptotic formula for $T_{\|}$of Brown is recovered in the limit of high barriers. On conversion of the exact solution for $T_{\|}$to integral form, it is shown using the method of steepest descents that an asymptotic correction to Brown's high-barrier result is necessary. The inadequacy of the effective-eigenvalue method as applied to the calculation of $T_{\|}$is discussed.


PACS number(s): $05.40 .+\mathrm{j}, 75.60 . \mathrm{Jp}, 76.20 .+\mathrm{q}$

## 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. The direction of the magnetization may undergo a rotation due to thermal agitation, surmounting the barrier, as first described by Neel [1]. He obtained an expression for the relaxation time $T_{\|}$ associated with the transition between two stable orientational states by assuming that the energy barrier between the states is so large compared to the thermal energy $k T$ that the directions of the magnetic moment of the particle are concentrated at the energy minima [2]. In addition, he restricted his analysis to two particular forms for the barrier potential $V$. First

$$
\begin{equation*}
V(\vartheta)=K \sin ^{2} \boldsymbol{\vartheta} \tag{1}
\end{equation*}
$$

and then

$$
\begin{equation*}
V(\vartheta)=K \sin ^{2} \vartheta-H M_{s} \cos \vartheta \tag{2}
\end{equation*}
$$

Equation (1) 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 $\boldsymbol{\vartheta}=0$ and $\boldsymbol{\vartheta}=\pi$, where the

[^0]orientation for $\mathbf{M}$ on a sphere of radius $\boldsymbol{M}_{s}$ (the saturation magnetization) is specified by the spherical polar coordinates $\boldsymbol{\vartheta}$ and $\phi, \boldsymbol{\vartheta}$ being the polar angle. In the presence of an external field $\mathbf{H}$ applied in the $\boldsymbol{\vartheta}=0$ direction the free energy per unit volume of the particle becomes asymmetric so that $V$ obeys Eq. (2).

Néel's calculation of $T_{\|}$was criticized by Brown [3,4] on two counts: (i) the system is not explicitly treated as a gyromagnetic one, and (ii) it relies on a discrete orientation approximation. Brown [3] suggested that both these difficulties could be resolved by constructing the FokkerPlanck equation for the distribution of magnetic-moment orientations on the unit sphere from the underlying Langevin equation. In his analysis Brown took as the Langevin equation the Landau-Lifshitz-Gilbert equation, [ 3,5 ] governing the behavior of $\mathbf{M}$, augmented by random-field terms which are assumed to be white noise. Thus he was able to deduce a Fokker-Planck equation, which for the axially symmetric potentials of Eqs. (1) and (2) becomes [3,5]

$$
\begin{align*}
2 \tau_{N} \frac{\partial W}{\partial t}= & \frac{1}{\sin \vartheta} \frac{\partial}{\partial \vartheta}\left[\sin \vartheta\left(\frac{\partial W}{\partial \vartheta}+\frac{v}{k T} \frac{\partial V}{\partial \vartheta} W\right]\right] \\
& +\frac{1}{\sin \vartheta} \frac{\partial}{\partial \varphi}\left\{\frac{v}{k T}\left(\frac{\partial V}{\alpha \partial \vartheta}\right) W+\frac{1}{\sin \vartheta} \frac{\partial W}{\partial \varphi}\right\} \tag{3}
\end{align*}
$$

where $W(\vartheta, \varphi, t)$ is the probability density of orientations of $\mathbf{M}$ at time $t, v$ is the volume of the particle, $k$ is Boltzmann's constant, and $T$ is the absolute temperature. The characteristic time $\tau_{N}$ and damping parameter $\alpha$ are
defined as [3]

$$
\tau_{N}=\frac{v}{2 \eta k T}\left(1 / \gamma^{2}+\eta^{2} M_{s}^{2}\right), \quad \alpha=\eta \gamma M_{s},
$$

where $\gamma$ is the gyromagnetic ratio and $\eta$ is the damping constant from Gilbert's equation, namely $[3,5]$

$$
\frac{d \mathbf{M}}{d t}=\gamma \mathbf{M} \times\left[\mathbf{H}_{T}-\eta \frac{d \mathbf{M}}{d t}\right]
$$

where $\mathbf{H}_{T}=\mathbf{H}-v \operatorname{grad}(V(\boldsymbol{\vartheta}))$.
Having written down the Fokker-Planck equation, Brown converted it [3] into a Sturm-Liouville problem for the special case of longitudinal relaxation. He did not calculate the eigenvalues for $V \neq 0$, merely assuming [6] that

$$
\begin{equation*}
T_{\|} \cong \frac{2 \tau_{N}}{\lambda_{1}} \tag{4}
\end{equation*}
$$

where $\lambda_{1}$ is the smallest nonvanishing eigenvalue of the Sturm-Liouville equation. He then used perturbation theory to obtain an approximate formula for $\lambda_{1}$ in the low-barrier limit and the Kramers transition-state method $[7,8]$ to obtain the high-barrier approximation. Thus using the simple uniaxial potential of Eq. (1) he was able to deduce that

$$
\begin{equation*}
\frac{T_{\|}}{\tau_{N}} \cong \frac{\sqrt{\pi}}{2} \sigma^{-3 / 2} e^{\sigma}, \quad \sigma=\frac{K v}{k T} \geq 2 . \tag{5}
\end{equation*}
$$

This has recently been rederived rigorously [9] by applying perturbation theory to the singular integral equation arising from Brown's Sturm-Liouville equation.

The assumption [Eq. (4)] used by Brown supposes [6] that, in the set of eigenvalues $\left\{\lambda_{k}\right\}$ of the Sturm-Liouville equation, $\lambda_{1} \ll \lambda_{k}, k \geq 2$, since all the exponential functions $\exp \left(-\lambda_{k} t / 2 \tau_{N}\right), k \geq 2$, are small compared with $\exp \left(-\lambda_{1} t / 2 \tau_{N}\right)$ except in the very early stages of the approach to equilibrium. This is an accurate assumption only for high-energy barriers [6].

Exact numerical calculations for the potential of Eq. (1) were first carried out by Aharoni [2]. He expanded Brown's Sturm-Liouville equation in Legendre polynomials and solved the resulting recursion formula numerically. Aharoni's calculations along with later work have been succinctly reviewed by Scully [6]. An essentially similar procedure was used by Martin, Meier, and Saupe [10] in a study of the analogous problem of dielectric relaxation of nematic liquid crystals.

Lately [11-14] there has been a revival of interest in the problem of calculating $T_{\|}$for the purpose of obtaining an analytical formula for $T_{\|}$, which is valid for all values of the barrier height parameter $\sigma$ in Eq. (1).

The analyses presented by Bessais, Ben Jaffel, and Dormann [11,12] and by Aharoni [13] proceed from the assumption embodied in Eq. (4). Both conclude with simple analytic formulas for $T_{\|}$valid for all values of $\sigma$. These appear to be derived as a result of a curve fitting to the exact $\lambda_{1}$ as determined by the numerical solution of the problem. On the other hand, the analysis presented by Garanin, Ischenko, and Panina [14] and that for the
analogous liquid-crystal problem by Moro and Nordio [15] utilize the definition from linear-response theory of the relaxation time as the correlation time-that is, the area under the curve of the magnetization autocorrelation function [16,17]. The use of the exact definition of the relaxation time enabled these investigators to write down integral expressions for the relaxation time from the Sturm-Liouville equation. However, rather than calculating exact analytic results from their formal equation, they both presented various asymptotic formulas for $T_{\|}$.

It is the principal purpose of this paper to show how the definition of $T_{\|}$as the area under the curve of the autocorrelation function enables one to obtain the exact solution for $T_{\|}$for the simple uniaxial potential of Eq. (1). The solution is presented both as a series of confluent hypergeometric (Kummer) functions [18] [Eq. (40)], which is easily tabulated for all $\sigma$ values, and in integral form [Eq. (54)]. The integral form is particularly suitable for the application of the method of steepest descents [19] for the purpose of obtaining an asymptotic expansion in the high- $\sigma$ limit. The leading term in the asymptotic expansion so derived coincides with the high-barrier formula of Brown [Eq. (5)].

This paper is arranged as follows. In Sec. II we demonstrate how the expansion of the distribution function of orientations in Legendre polynomials leads to the set of differential-difference equations [Eq. (7)] for the aftereffect solution. These equations are then arranged in matrix form [Eq. (9)], with the initial conditions being given as a ratio of two confluent hypergeometric (Kummer) functions [18]. The set of equations is then solved numerically to yield the eigenvalues and their corresponding amplitudes as given in Table I.

Proceeding to Sec. III, $T_{\|}$is now regarded as the zerofrequency limit of the Laplace transform of the autocorrelation function of the magnetization. This definition, used in conjunction with the matrix formulation of the problem, circumvents the solution of the characteristic equation which is required in the lowesteigenvalue method. Furthermore, it allows $T_{\|}$to be calculated by simply calculating the inverse of the system

TABLE I. Amplitudes $A_{2 k+1}$ of the first three modes of the decay of the longitudinal polarization as a function of the barrier height parameter $\sigma$ and corresponding eigenvalues $\lambda_{2 k+1}$, $k=0,1,2$, in the form $\lambda_{2 k+1} / 2 \tau_{N}$. A $12 \times 12$ matrix was used for the amplitudes $A_{2 k+1}$ and a $20 \times 20$ matrix was used to ensure convergence of the eigenvalues $\lambda_{2 k+1}$.

| $\sigma$ | $\boldsymbol{A}_{1}$ | $\boldsymbol{A}_{3}$ | $\boldsymbol{A}_{5}$ | $\lambda_{1}$ | $\lambda_{3}$ | $\lambda_{5}$ |
| ---: | ---: | :--- | :--- | :--- | :--- | :--- |
| 1 | 0.428 | 0.000975 | $9.17 \times 10^{-7}$ | 0.653 | 5.81 | 14.8 |
| 2 | 0.528 | 0.00365 | 0.0000146 | 0.404 | 5.77 | 14.8 |
| 3 | 0.619 | 0.00676 | 0.0000662 | 0.236 | 5.91 | 14.8 |
| 4 | 0.696 | 0.0088 | 0.000171 | 0.13 | 6.23 | 15.0 |
| 5 | 0.755 | 0.00916 | 0.000319 | 0.0677 | 6.74 | 15.4 |
| 6 | 0.799 | 0.00824 | 0.000481 | 0.0336 | 7.45 | 15.9 |
| 7 | 0.832 | 0.00674 | 0.000624 | 0.016 | 8.37 | 16.5 |
| 8 | 0.856 | 0.0052 | 0.00073 | 0.00736 | 9.49 | 17.3 |
| 9 | 0.875 | 0.0039 | 0.000792 | 0.00329 | 10.8 | 18.1 |
| 10 | 0.889 | 0.00289 | 0.000812 | 0.00144 | 12.3 | 19.2 |

matrix A [Eq. (10)], which is a function of $\sigma$ only. It is further shown that the method is not confined to the problem at hand.

In Sec. IV it is shown how the longitudinal susceptibility and correlation time may be obtained exactly from the Laplace transform of the hierarchy of differentialdifference equations in terms of products of infinite continued fractions in the frequency $\omega$ and barrier-height parameter $\sigma$. The definition of $T_{\|}$in the zero-frequency limit is then further exploited, using the final value theorem for Laplace transforms [20], to write down the exact analytic solution for $T_{\|}$[Eq. (34)] in terms of an infinite continued fraction in $\sigma$ alone. This solution in turn (Sec. V) may be written (just as in the corresponding two-dimensional problem [21]) as a series of Kummer functions [Eq. (40)]. The analytic solution rendered by Eqs. (33) and (40) is the central result of the paper, whence a table of values of $T_{\|}$valid for all values of $\sigma$ may be constructed as given in Appendix E.

In Sec. VI the asymptotic form of the Kummer functions is used to demonstrate how the exact solution yields Brown's asymptotic formula [Eq. (5)] in the high-barrier limit. In Sec. VII the representation for the product of two Kummer functions as an integral [22] is used to render the series representation of the exact solution in integral form [Eq. (54)]. The method of steepest descents [19] is then applied to the integral form of the solution in Sec. VIII to obtain correction terms to Brown's asymptotic formula [Eq. (5)]. This procedure reproduces the exact solution to a high degree of accuracy in the high- $\sigma$ limit.

In order to facilitate for the reader, the mathematical details of the calculations, which are very lengthy, have been given in Appendixes A-D at the end of the paper. Appendix E constitutes a table of the exact solution [Eq. (40)].

$$
\mathbf{X}=\left(\begin{array}{c}
f_{1}(t) \\
f_{3}(t) \\
\vdots \\
f_{2 n+1}(t) \\
\cdots
\end{array}\right), \quad \mathbf{A}=-\frac{1}{\tau_{N}}\left[\begin{array}{cc}
\left(1-\frac{2}{5} \sigma\right) & \frac{2}{5} \sigma \\
-\frac{24}{35} \sigma & \left(6-\frac{4}{15} \sigma\right) \\
0 & -\frac{40}{33} \sigma \\
\cdots & \cdots
\end{array}\right.
$$

In Eq. (7), $n$ is taken large enough (equal to $P$ say) to ensure convergence of the set of equations (9).

The lowest eigenvalue, which corresponds to the reciprocal of the longest relaxation time, is then the smallest root of the characteristic equation

$$
\begin{equation*}
\operatorname{det}(\lambda \mathbf{I}-\mathbf{A})=0 \tag{11}
\end{equation*}
$$

The relaxation modes of $f_{1}(t)$ may be found from Eq. (9) by assuming that [25] A has a linearly independent set of $P$ eigenvectors ( $\mathbf{R}_{1}, \ldots, \mathbf{R}_{P}$ ), so that
$\mathbf{X}(t)=b_{1} e^{\lambda_{1} t} \mathbf{R}_{1}+b_{2} e^{\lambda_{2} t} \mathbf{R}_{2}+\cdots+b_{P} e^{\lambda_{P} t} \mathbf{R}_{P}$,
where the $b_{i}$ are to be determined from the initial condi-

## II. REPRESENTATION OF THE LONGITUDINAL RELAXATION PROBLEM AS A SET OF DIFFERENTIAL-DIFFERENCE EQUATIONS

In order to study the longitudinal relaxation behavior we suppose that a small constant field $\mathbf{H}$ ( $v M_{s} H / k T \ll 1$ ) applied along the $z$ axis is switched off at $t=0$, so that we determine the aftereffect solution of Eq. (3). We can disregard the dependence of $W$ on $\varphi$ for the longitudinal relaxation; hence we may assume that the distribution function $W$ is

$$
\begin{equation*}
W(\vartheta, t)=\sum_{n=0}^{\infty} a_{n}(t) P_{n}(\cos \vartheta) \tag{6}
\end{equation*}
$$

where the $P_{n}(x)$ are the Legendre polynomials [18]. On substituting Eq. (6) into Eq. (3) we obtain the differentialdifference equation [5,23,24]

$$
\begin{align*}
& \frac{2 \tau_{N}}{n(n+1)} \dot{f}_{n}+\left[1-\frac{2 \sigma}{(2 n-1)(2 n+3)}\right] f_{n} \\
& \quad=\frac{2 \sigma(n-1)}{(2 n+1)(2 n-1)} f_{n-2}-\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)} f_{n+2}, \tag{7}
\end{align*}
$$

where

$$
\begin{equation*}
f_{n}(t)=\frac{1}{2 n+1} \frac{a_{n}(t)}{a_{0}} \tag{8}
\end{equation*}
$$

By inspection of Eq. (7) it is obvious that it decouples into sets for even and odd $f_{n}(t)$. Here, only the odd $f_{n}(t)=f_{2 k+1}(t)$ are of interest since we seek the relaxation behavior of $f_{1}(t)$.

The set of equations (7) may be solved numerically by forming the matrix equation and writing

$$
\begin{equation*}
\dot{\mathbf{X}}=\mathbf{A} \mathbf{X} \tag{9}
\end{equation*}
$$

where
tions. The initial value vector $\mathbf{X}_{0}$ is determined as follows. At time $t=0$ the steady probe field $\mathbf{H}$ is switched off. Thus the initial value of $f_{2 n+1}(t)$ is in the linear approximation

$$
\begin{align*}
f_{2 n+1}(0) & =\frac{\int_{-1}^{+1} P_{2 n+1}(x) e^{\sigma x^{2}+\xi x} d x}{\int_{-1}^{+1} e^{\sigma x^{2}+\xi x} d x} \\
& =\frac{\xi \int_{-1}^{+1} x P_{2 n+1}(x) e^{\sigma x^{2}} d x}{\int_{-1}^{+1} e^{\sigma x^{2}} d x} \\
& =\xi\left\langle x P_{2 n+1}\right\rangle_{0} \tag{13}
\end{align*}
$$

where $\xi=\nu M_{s} H / k T$. The subscript 0 denotes that the statistical average is taken in the absence of the external field $\mathbf{H}$. Equation (13) is evaluated as a ratio of two confluent hypergeometric (Kummer) functions as described in detail in Appendix B. The Kummer function $M(a, b, z)[18]$ is defined as

$$
\begin{align*}
M(a, b, z)= & 1+\frac{a}{b} z+\frac{a(a+1)}{b(b+1)} \frac{z^{2}}{2!} \\
& +\frac{a(a+1)(a+2)}{b(b+1)(b+2)} \frac{z^{3}}{3!}+\cdots . \tag{14}
\end{align*}
$$

This leads to
$f_{2 n+1}(0)=\frac{\xi \sigma^{n} \Gamma(n+3 / 2) M(n+3 / 2,2 n+5 / 2, \sigma)}{2 \Gamma(2 n+5 / 2) M\left(\frac{1}{2}, \frac{3}{2}, \sigma\right)}$,
so yielding the initial value vector $\mathbf{X}_{0}$. The set of equations (9) may now be solved to any desired degree of accuracy to yield the decay of the longitudinal component of the magnetization as
$m N f_{1}(t)=m N\langle\cos \vartheta\rangle=\frac{m^{2} N H}{k T} \sum_{k=0}^{\infty} A_{2 k+1} e^{-\lambda_{2 k+1} t}$,
where $N$ is the number of particles per unit volume, and

$$
\begin{equation*}
m=M_{s} v \tag{17}
\end{equation*}
$$

The first three eigenvalues in the form $\lambda_{2 k+1} / 2 \tau_{N}$ and the amplitudes $A_{2 k+1}$ are given in Table I as a function of $\sigma$, whence the lowest mode contributes almost all the decay. A $12 \times 12$ matrix was used for the amplitudes $A_{2 k+1}$ and a $20 \times 20$ matrix was used to ensure convergence of the eigenvalues $\lambda_{2 k+1}$. The quantity of most interest to us is the correlation time $T_{\|}$which is $[16,17]$ the area under the curve of the longitudinal autocorrelation function. The longitudinal autocorrelation function of the magnetization in the linear approximation in $\xi$ is

$$
\begin{equation*}
\frac{f_{1}(t)}{f_{1}(0)}=C_{1}(t)=\frac{\langle\cos \vartheta(0) \cos \vartheta(t)\rangle_{0}}{\left\langle\cos ^{2} \vartheta(0)\right\rangle_{0}} \tag{18}
\end{equation*}
$$

so that the correlation time $T_{\|}$is from Eq. (16)

$$
\begin{equation*}
T_{\|}=\int_{0}^{\infty} C_{1}(t) d t=\frac{\sum_{k} A_{2 k+1} \lambda_{2 k+1}^{-1}}{\sum_{k} A_{2 k+1}} \tag{19}
\end{equation*}
$$

By inspection of Table $I, T_{\|}$is effectively the reciprocal of the lowest eigenvalue. Another quantity which we shall require is the effective eigenvalue defined as $[16,17]$

$$
\begin{equation*}
\lambda_{\mathrm{ef}}=-\frac{\dot{f}(0)}{f(0)}=\frac{\sum_{k} A_{2 k+1} \lambda_{2 k+1}}{\sum_{k} A_{2 k+1}} \tag{20}
\end{equation*}
$$

which may also be evaluated from Table I. $\lambda_{\text {ef }}$ is the reciprocal time constant associated with the initial slope of the magnetization decay. It also contains contributions from all the eigenvalues just as the correlation time $T_{\|}$. The behavior of $T_{\|}$and $\lambda_{\text {ef }}^{-1}$ is sometimes similar. In fact [17], if a single eigenvalue dominates the decay of the
correlation function and

$$
\begin{equation*}
A_{1} \lambda_{1} \gg A_{2 k+1} \lambda_{2 k+1}, \quad k>1 \tag{21}
\end{equation*}
$$

then $T_{\|}=\lambda_{\text {ef }}^{-1}$. However, if different time scales are involved, $T_{\|}$and $\lambda_{\text {ef }}^{-1}$ may not be similar and in this case $\lambda_{\text {ef }}$ gives precise information on the initial decay of the correlation function.

## III. MATRIX FORMULA FOR THE CORRELATION TIME

The definition of the correlation time given above suggests a simpler way of finding $T_{\|}$than solving the characteristic Eq. (11). First we note that according to Eq. (19)

$$
\begin{equation*}
T_{\|}=\lim _{s \rightarrow 0} \int_{0}^{\infty} C_{1}(t) e^{-s t} d t=\widetilde{C}_{1}(0) \tag{22}
\end{equation*}
$$

where $\widetilde{C}_{1}(s)$ is the Laplace transform of $C_{1}(t)$ and $\widetilde{C}_{1}(0)$ is the value of that quantity at zero frequency. The Laplace transform of Eq. (9) is

$$
s \widetilde{\mathbf{X}}(s)-\mathbf{X}(0)=\mathbf{A} \widetilde{\mathbf{X}}(s)
$$

which, noting the final value theorem for Laplace transforms, namely [20],

$$
\begin{equation*}
\lim _{s \rightarrow 0} s \widetilde{f}(s)=\lim _{t \rightarrow \infty} f(t) \tag{23}
\end{equation*}
$$

becomes for $s=0$,

$$
\begin{equation*}
\mathbf{X}(\infty)-\mathbf{X}(0)=\mathbf{A} \tilde{\mathbf{X}}(0) \tag{24}
\end{equation*}
$$

Furthermore,

$$
\begin{equation*}
\mathbf{X}(\infty)=0 \tag{25}
\end{equation*}
$$

because all the $f_{2 n+1}(\infty)$ vanish. Thus Eq. (24) becomes

$$
\begin{equation*}
-\mathbf{X}(0)=\mathbf{A} \widetilde{\mathbf{X}}(0) \tag{26}
\end{equation*}
$$

with the solution

$$
\begin{equation*}
\widetilde{\mathbf{X}}(0)=-\mathbf{A}^{-1} \mathbf{X}(0) \tag{27}
\end{equation*}
$$

The relaxation time $T_{\|}$may be extracted from this set merely by calculating $\mathbf{A}^{-1}$. This is much easier than approximating the correlation time as in Eq. (4) by the reciprocal of the lowest eigenvalue obtained from Eq. (11). This always requires one to solve the high-order polynomial equation [Eq. (11)]. Such a procedure also yields $T_{\|}$ from a matrix which is merely a function of $\sigma$ - not, as in the lowest eigenvalue method, a function of $\sigma$ and $s$. We remark that the method is not confined to the set of differential-difference equations for the $f_{2 n+1}$. It may be applied to the aftereffect solution whenever the set of equations for the expansion coefficients in the corresponding Fokker-Planck equation may be written in the matrix form [Eq. (9)]. It also allows us to calculate all the functions $\widetilde{f}_{2 n+1}(0)$ and thus the correlation times of the other $f_{2 n+1}(t)$. We shall now demonstrate how an exact analytical formula for the Laplace transform of the aftereffect function $\widetilde{f}_{1}(s)$, and hence $T_{\|}$, may be obtained

## IV. ANALYTIC FORMULA FOR THE LONGITUDINAL SUSCEPTIBILITY AND CORRELATION TIME

Consider the Laplace transform of the recurrence relation for $\widetilde{f}_{n}(s)$ [Eq. (7)]. We have

$$
\begin{equation*}
\widetilde{R}_{n}(s)\left[\frac{2 \tau_{N} s}{n(n+1)}+1-\frac{2 \sigma}{(2 n-1)(2 n+3)}+\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)} \widetilde{R}_{n+2}(s)\right]=\frac{2 \tau_{N}}{n(n+1)} \frac{f_{n}(0)}{\widetilde{f}_{n-2}(s)}+\frac{2 \sigma(n-1)}{4 n^{2}-1}, \tag{28}
\end{equation*}
$$

where

$$
\begin{equation*}
\widetilde{R}_{n}(s)=\widetilde{f}_{n}(s) / \widetilde{f}_{n-2}(s) \tag{29}
\end{equation*}
$$

The solution of Eq. (28) will allow us to determine $\tilde{f}_{1}(s)$. The homogeneous equation (28) [i.e., with $f_{n}(0)=0$ ] may be readily solved in terms of the continued fraction

$$
\begin{equation*}
\widetilde{S}_{n}(s)=\frac{\frac{2 \sigma(n-1)}{4 n^{2}-1}}{\frac{2 \tau_{N} s}{n(n+1)}+1-\frac{2 \sigma}{(2 n-1)(2 n+3)}+\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)} \widetilde{S}_{n+2}(s)} . \tag{30}
\end{equation*}
$$

As shown in Appendix A, we can then solve [21,27-29] the inhomogeneous equation (28) in terms of the $\widetilde{S}_{n}(s)$ by successively eliminating the other variables to get

$$
\begin{equation*}
\frac{\tilde{f}_{1}(s)}{f_{1}(0)}=\frac{\tau_{N}}{\tau_{N} s+1-\frac{2}{5} \sigma+\frac{2}{5} \sigma \widetilde{S}_{3}(s)}\left[1+\frac{4}{3} \sum_{n=1}^{\infty}(-1)^{n} \frac{f_{2 n+1}(0)}{f_{1}(0)} \frac{\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2) \Gamma\left(\frac{1}{2}\right)} \prod_{k=1}^{n} \widetilde{S}_{2 k+1}(s)\right] \tag{31}
\end{equation*}
$$

This exact formula allows one to calculate the frequency dependence of the longitudinal susceptibility $\chi_{\|}(\omega)=\chi^{\prime}(\omega)-i \chi^{\prime \prime}(\omega)$, since according to linear-response theory [30]

$$
\begin{equation*}
\frac{\chi_{\|}(\omega)}{\chi_{\|}^{\prime}(0)}=1-i \omega \int_{0}^{\infty} e^{-i \omega t} C_{1}(t) d t=1-i \omega \frac{\widetilde{f}_{1}(i \omega)}{f_{1}(0)} \tag{32}
\end{equation*}
$$

Thus we have

$$
\begin{align*}
\frac{\chi_{\|}(\omega)}{\chi_{\|}^{\prime}(0)}= & \frac{1}{i \omega \tau_{N}+1-\frac{2}{5} \sigma+\frac{2}{5} \sigma \widetilde{S}_{3}(i \omega)} \\
& \times\left[1-\frac{2}{5} \sigma+\frac{2}{5} \sigma \widetilde{S}_{3}(i \omega)-i \omega \tau_{N} \frac{4}{3} \sum_{n=1}^{\infty}(-1)^{n} \frac{f_{2 n+1}(0)}{f_{1}(0)} \frac{\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2) \Gamma\left(\frac{1}{2}\right)} \prod_{k=1}^{n} \widetilde{S}_{2 k+1}(i \omega)\right], \tag{33}
\end{align*}
$$

where

$$
\chi_{\|}^{\prime}(0)=m N f_{1}(0) / H=\frac{m^{2} N}{3 k T} \frac{M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)}{M\left(\frac{1}{2}, \frac{3}{2}, \sigma\right)} .
$$

The most significant feature of Eq. (31) is however that it yields an exact expression for the correlation time. We have, on setting $s=0$ in Eq. (31),

$$
\begin{equation*}
T_{\|}=\frac{\widetilde{f}_{1}(0)}{f_{1}(0)}=\frac{\tau_{N}}{1-\frac{2}{5} \sigma+\frac{2}{5} \sigma \widetilde{S}_{3}(0)}\left[1+\frac{4}{3} \sum_{n=1}^{\infty}(-1)^{n} \frac{f_{2 n+1}(0)}{f_{1}(0)} \frac{\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2) \Gamma\left(\frac{1}{2}\right)} \prod_{k=1}^{n} \widetilde{S}_{2 k+1}(0)\right] \tag{34}
\end{equation*}
$$

Equation (34) is an exact analytical formula which allows $T_{\|}$to be calculated to any desired degree of accuracy by computing successive convergents of the continued fraction $\widetilde{S}_{2 k+1}(0)$. The results obtained from Eq. (34) are coincident with those of the matrix inversion method of Sec. III above. We shall now demonstrate how $T_{\|}$may be written as a series of Kummer functions, which allows one to easily deduce the asymptotic behavior of $T_{\|}$.

## V. ANALYTIC FORMULA FOR $T_{\|}$ IN TERMS OF KUMMER'S FUNCTIONS

The mathematical procedure involved in expressing $T_{\|}$ in terms of Kummer's functions is rather lengthy and so is described in Appendix C. We have, using the results of Appendix C, an expression for $\widetilde{S}_{2 k+1}(0)$ in terms of Kummer's functions of ascending order
$\widetilde{S}_{2 k+1}(0)=\frac{4 k \sigma}{(4 k+1)(4 k+3)} \frac{M\left(k+1,2 k+\frac{5}{2}, \sigma\right)}{M\left(k, 2 k+\frac{1}{2}, \sigma\right)}$.
It is also shown in Appendix $C$ that

$$
\begin{equation*}
\frac{1}{1-\frac{2 \sigma}{5}+\frac{2 \sigma}{5} \widetilde{S}_{3}(0)}=M\left(1, \frac{5}{2}, \sigma\right) \tag{35}
\end{equation*}
$$

Equation (15) yields the ratio $f_{2 n+1}(0) / f_{1}(0)$ as a ratio of two Kummer functions

$$
\begin{equation*}
\frac{f_{2 n+1}(0)}{f_{1}(0)}=\frac{3 \sigma^{n} \Gamma\left(n+\frac{3}{2}\right) M\left(n+\frac{3}{2}, 2 n+\frac{5}{2}, \sigma\right)}{2 \Gamma\left(2 n+\frac{5}{2}\right) M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \tag{37}
\end{equation*}
$$

so that with the aid of Eqs. (35)-(37) we have the explicit formula

$$
\begin{align*}
\frac{T_{\|}}{\tau_{N}}=\boldsymbol{M}\left(1, \frac{5}{2}, \sigma\right)\left\{1+\frac{4}{3} \sum_{n=1}^{\infty}\right. & (-1)^{n} \frac{3 \sigma^{n} \Gamma\left(n+\frac{3}{2}\right) \boldsymbol{M}\left(n+\frac{3}{2}, 2 n+\frac{5}{2}, \sigma\right)}{2 \Gamma\left(2 n+\frac{5}{2}\right) \boldsymbol{M}\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \\
& \left.\times \frac{\left(n+\frac{3}{2}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2) \Gamma\left(\frac{1}{2}\right)} \prod_{k=1}^{n} \frac{4 k \sigma}{(4 k+1)(4 k+3)} \frac{M\left(k+1,2 k+\frac{5}{2}, \sigma\right)}{M\left(k, 2 k+\frac{1}{2}, \sigma\right)}\right\} . \tag{38}
\end{align*}
$$

This may be further simplified by noting that

$$
\prod_{k=1}^{n} \frac{M\left(k+1,2 k+\frac{5}{2}, \sigma\right)}{M\left(k, 2 k+\frac{1}{2}, \sigma\right)}=\frac{M\left(n+1,2 n+\frac{5}{2}, \sigma\right)}{M\left(1, \frac{5}{2}, \sigma\right)},
$$

and

$$
\prod_{k=1}^{n} \frac{4 k \sigma}{(4 k+1)(4 k+3)}=\frac{n!\sigma^{n} 48(2 \pi)^{-1 / 2} \Gamma\left(\frac{3}{4}\right) \Gamma\left(\frac{1}{4}\right)}{(4 n+3)(4 n+1) \Gamma\left(2 n+\frac{1}{2}\right)} .
$$

Using the $\Gamma$-function recurrence relations

$$
\begin{equation*}
z \Gamma(z)=\Gamma(z+1), \quad \Gamma(2 z)=(2 \pi)^{-1 / 2} 2^{2 z-1 / 2} \Gamma(z) \Gamma\left(z+\frac{1}{2}\right) \tag{39}
\end{equation*}
$$

we have

$$
\begin{align*}
\frac{T_{\|}}{\tau_{N}}=M\left(1, \frac{5}{2}, \sigma\right)+\frac{\frac{3}{2}}{M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \sum_{n=1}^{\infty} & \frac{\left(-\sigma^{2}\right)^{n}\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{3}{2}\right) \Gamma\left(n+\frac{1}{2}\right)}{(n+1)\left[\Gamma\left(2 n+\frac{5}{2}\right)\right]^{2}} \\
& \times M\left(n+\frac{3}{2}, 2 n+\frac{5}{2}, \sigma\right) M\left(n+1,2 n+\frac{5}{2}, \sigma\right) \tag{40}
\end{align*}
$$

which is the exact solution in terms of known functions for the longitudinal relaxation time $T_{\|}$for the $K \sin ^{2} \vartheta$ potential. This function is tabulated for a wide range of values of $\sigma$ in Appendix E. It is apparent, using the series definition of the Kummer function Eq. (14), that $T_{\|} / \tau_{N}=1$ for $\sigma=0$.

We remark that all the Kummer functions appearing in Eq. (40) may be expressed in terms of the more familiar error functions of the real and imaginary arguments [18]

$$
\begin{aligned}
& \operatorname{erf}(x)=\frac{2}{\sqrt{\pi}} \int_{0}^{x} \exp \left(-t^{2}\right) d t \\
& \operatorname{erf}(x)=\frac{2}{\sqrt{\pi}} \int_{0}^{x} \exp \left(t^{2}\right) d t
\end{aligned}
$$

In particular ([36], p. 580)

$$
\begin{aligned}
& M\left(1, \frac{5}{2}, z\right)=\frac{3}{2 z}[\sqrt{\pi / 4 z} \exp (z) \operatorname{erf}(\sqrt{z})-1] \\
& M\left(\frac{3}{2}, \frac{5}{2}, z\right)=\sqrt{\pi / 4 z} \operatorname{erfi}(\sqrt{z})
\end{aligned}
$$

Equations for the other $M$ functions occurring in Eq. (40) may be obtained from Table 7.11.2 of Ref. [36] and the recurrence relation for the Kummer functions.

We shall now demonstrate how the asymptotic formula of Brown [Eq. (5)] may be recovered from Eq. (40) in the high-barrier limit.

## VI. RECOVERY OF BROWN'S ASYMPTOTIC FORMULA FROM THE HIGH-BARRIER LIMIT OF EQ. (40)

In order to obtain the high- $\sigma$ limit of Eq. (40) we note that the asymptotic form of $M(a, b, z)$ as $|z| \rightarrow \infty$ is [cf.

Eq. (13.1.4) of [18]].

$$
M(a, b, z)=\frac{\Gamma(b)}{\Gamma(a)} e^{z} z^{a-b}\left[1+O\left(|z|^{-1}\right)\right], \quad \operatorname{Re}(z)>0
$$

Thus Eq. (40) becomes in the high- $\sigma$ limit
$\frac{T_{\|}}{\tau_{N}}=e^{\sigma} \sigma^{-3 / 2} \sum_{n=0}^{\infty} \frac{(-1)^{n}}{(n+1)!} \Gamma\left(n+\frac{1}{2}\right)\left(n+\frac{3}{4}\right)$.
We sum this series by expressing it in terms of Gauss hypergeometric functions ${ }_{2} F_{1}(a, b ; c ; z)[18]$ as follows:

$$
\begin{align*}
\sum_{n=0}^{\infty} & \frac{(-1)^{n}}{(n+1)!} \Gamma\left(n+\frac{1}{2}\right)\left(n+\frac{3}{4}\right) \\
& =\sqrt{\pi}\left[\sum_{n=0}^{\infty} \frac{\left(\frac{1}{2}\right)_{n}(-1)^{n}}{n!}-\frac{1}{4} \sum_{n=0}^{\infty} \frac{\left(\frac{1}{2}\right)_{n}(1)_{n}(-1)^{n}}{(2)_{n} n!}\right] \\
& =\sqrt{\pi}\left[{ }_{2} F_{1}\left(\frac{1}{2}, b ; b ;-1\right)-\left(\frac{1}{4}\right){ }_{2} F_{1}\left(\frac{1}{2}, 1 ; 2 ;-1\right)\right]  \tag{42}\\
& =\sqrt{\pi} / 2 \tag{43}
\end{align*}
$$

where [18]

$$
\begin{align*}
& { }_{2} F_{1}(a, b ; c ; z)=\sum_{n=0}^{\infty} \frac{(a)_{n}(b)_{n}}{(c)_{n}} \frac{z^{n}}{n!}  \tag{44}\\
& (a)_{n}=a(a+1)(a+2) \cdots(a+n-1) \\
& { }_{2} F_{1}(a, b ; b ; z)=(1-z)^{-a}  \tag{45}\\
& { }_{2} F_{1}\left(a, a+\frac{1}{2} ; 1+2 a ; z\right)=2^{2 a}\left[1+(1-z)^{1 / 2}\right]^{-2 a}
\end{align*}
$$

Thus, Eq. (41) becomes

$$
\begin{equation*}
T_{\|} / \tau_{N} \cong(\sqrt{\pi} / 2) e^{\sigma} \sigma^{-3 / 2}, \tag{46}
\end{equation*}
$$

which is the asymptotic formula of Brown [Eq. (5)]. It is apparent from our previous work on the two-dimensional problem [2i] and that of Storonkin [31,32] that this formula (46) will not exactly reproduce the asymptotic behavior, as is demonstrated in Fig. 1 and Table II. Thus it is necessary to calculate correction terms to Eq. (46). This is accomplished by writing the exact solution for $T_{\|}$ [Eq. (40)] in integral form and utilizing the method of steepest descents [19].

## VII. INTEGRAL FORM OF THE EXACT SOLUTION

In order to write our series solution for $T_{\|} / \tau_{N}$ [Eq. (40)] in integral form we note the formula from Bateman [ [22] Vol. 1: 6.15.3, Eq. (18)]


FIG. 1. Exact solution [Eq. (40)] for the longitudinal correlation time $T_{\|} / \tau_{N}$ (solid line) compared with the asymptotic solution [Eq.(5)] (large dashed line) of Brown. The small dashed line is the solution rendered by the asymptotic formula (59).

$$
\begin{equation*}
M(a, b, z) M(a, b,-z)=\frac{[\Gamma(b)]^{2}, z^{1-b}}{\Gamma(a) \Gamma(b-a)} \int_{-\infty}^{\infty} \operatorname{sech} t I_{b-1}(z \operatorname{sech} t) e^{(b-2 a) t} d t, \quad \operatorname{Re}\{a\}>0, \quad \operatorname{Re}\{b-a\}>0 \tag{47}
\end{equation*}
$$

where $I_{v}(z)$ is the modified Bessel function of the first kind of order $v$ [18]. Thus the product of two Kummer functions may be expressed as an integral. In order to apply the above formula to Eq. (40) we note the Kummer transformation [Eq. (13.1.27) of Ref. [18]]

$$
\begin{equation*}
M(a, b, z)=e^{z} M(b-a, b,-z) \tag{48}
\end{equation*}
$$

so that Eq. (48), taking $a=n+\frac{3}{2}$, and $b=2 n+\frac{5}{2}$, be-

## comes

$$
\begin{gather*}
M\left(n+\frac{3}{2}, 2 n+\frac{5}{2}, \sigma\right) M\left(n+1,2 n+\frac{5}{2}, \sigma\right) \\
=e^{\sigma} M\left(n+\frac{3}{2}, 2 n+\frac{5}{2}, \sigma\right) \\
\quad \times M\left(n+\frac{3}{2}, 2 n+\frac{5}{2},-\sigma\right), \tag{49}
\end{gather*}
$$

so casting Eq. (40) into a form suitable for conversion to

TABLE II. Comparison of various asymptotic formulas for $T_{\|} / \tau_{N}$ with the exact solution [Eq. (40)]. Equation (60) is Brown's formula with asymptotic $\sigma^{-1}$ and $\sigma^{-2}$ corrections. Equation (59) is Brown's formula with $\sigma^{-1}$ correction; Eq. (5) is Brown's formula without correction.

| $\sigma$ | Eq. $(40)$ | Eq. $(60)$ | Eq. $(59)$ | Eq. $(5)$ |
| :--- | :---: | :---: | :---: | :---: |
| 0 | 1.0 | $\infty$ | $\infty$ | $\infty$ |
| 1.0 | 1.528 | 8.4316 | 4.818 | 2.409 |
| 1.5 | 1.9254 | 5.0446 | 3.6033 | 2.162 |
| 2.0 | 2.4603 | 4.341 | 3.4728 | 2.3152 |
| 2.5 | 3.1899 | 4.4793 | 3.8238 | 2.7313 |
| 3.0 | 4.1982 | 5.1385 | 4.5676 | 3.4257 |
| 3.5 | 5.6091 | 6.3114 | 5.7626 | 4.482 |
| 4.0 | 7.6061 | 8.1274 | 7.5604 | 6.0483 |
| 4.5 | 10.463 | 10.833 | 10.214 | 8.357 |
| 5.0 | 14.589 | 14.823 | 14.117 | 11.764 |
| 6.0 | 29.43 | 29.395 | 28.381 | 24.327 |
| 10.0 | 691.02 | 688.28 | 679.02 | 617.29 |
| 14.0 | 21986.0 | 21955.0 | 21799.0 | 20346.0 |
| 18.0 | $8.0835 \times 10^{5}$ | $8.0783 \times 10^{5}$ | $8.043 \times 10^{5}$ | $7.6197 \times 10^{5}$ |
| 22.0 | $3.2294 \times 10^{7}$ | $3.2283 \times 10^{7}$ | $3.2188 \times 10^{7}$ | $3.0789 \times 10^{7}$ |
| 26.0 | $1.3619 \times 10^{9}$ | $1.3616 \times 10^{9}$ | $1.3587 \times 10^{9}$ | $1.3084 \times 10^{9}$ |
| 30.0 | $5.9662 \times 10^{90}$ | $5.9654 \times 10^{10}$ | $5.9558 \times 10^{10}$ | $5.7636 \times 10^{10}$ |
| 34.0 | $2.6885 \times 10^{12}$ | $2.6883 \times 10^{12}$ | $2.6849 \times 10^{12}$ | $2.6082 \times 10^{12}$ |
| 38.0 | $1.2382 \times 10^{14}$ | $1.2382 \times 10^{14}$ | $1.2369 \times 10^{14}$ | $1.2052 \times 10^{14}$ |
| 42.0 | $5.8028 \times 10^{15}$ | $5.8026 \times 10^{15}$ | $5.7977 \times 10^{15}$ | $5.6629 \times 10^{15}$ |
| 46.0 | $2.7581 \times 10^{17}$ | $2.758 \times 10^{17}$ | $2.7561 \times 10^{17}$ | $2.6975 \times 10^{17}$ |
| 50.0 | $1.3264 \times 10^{19}$ | $1.3264 \times 10^{19}$ | $1.3256 \times 10^{19}$ | $1.2996 \times 10^{19}$ |
| 54.0 | $6.4425 \times 10^{20}$ | $6.4424 \times 10^{20}$ | $6.4391 \times 10^{20}$ | $6.322 \times 10^{20}$ |
| 58.0 | $3.1558 \times 10^{22}$ | $3.1557 \times 10^{22}$ | $3.1543 \times 10^{22}$ | $3.1009 \times 10^{22}$ |

an integral, namely,

$$
\begin{align*}
\frac{T_{\|}}{\tau_{N}}= & M\left(1, \frac{5}{2}, \sigma\right)+3\left[M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)\right]^{-1} e^{\sigma} \sigma^{-3 / 2} \\
\times & \sum_{n=1}^{\infty} \frac{(-1)^{n}\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2)} \\
& \times \int_{0}^{\infty} I_{2 n+3 / 2}(\sigma \operatorname{sech} t) \frac{\cosh (t / 2)}{\cosh t} d t \tag{50}
\end{align*}
$$

which, using the change of variable $\operatorname{sech} t=\sin \theta$, reduces to

$$
\begin{align*}
\frac{T_{\|}}{\tau_{N}}= & M\left(1, \frac{5}{2}, \sigma\right) \\
& +\frac{3 e^{\sigma} \sigma^{-3 / 2}}{M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \int_{0}^{\pi / 2} d \theta\left(\frac{1+\sin \theta}{2 \sin \theta}\right)^{1 / 2} F(\theta) \tag{51}
\end{align*}
$$

where

$$
\begin{equation*}
F(\theta)=\sum_{n=1}^{\infty} \frac{(-1)^{n}\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2)} I_{2 n+3 / 2}(\sigma \sin \theta) \tag{52}
\end{equation*}
$$

It is shown in Appendix D that the series Eq. (52) is

$$
\begin{equation*}
F(\theta)=\frac{1}{2 \sqrt{2 \sigma \sin \theta}}\left[\cosh (\sigma \sin \theta)-\frac{3 \sinh (\sigma \sin \theta)}{\sigma \sin \theta}+2\right] \tag{53}
\end{equation*}
$$

Thus

$$
\begin{equation*}
\frac{T_{\|}}{\tau_{N}}=M\left(1, \frac{5}{2}, \sigma\right)-\frac{3 e^{\sigma} \sigma^{-2}}{4 M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \int_{0}^{\pi / 2} d \theta \frac{\sqrt{1+\sin \theta}}{\sin \theta}\left[\frac{1}{2}\left(e^{\sigma \sin \theta}+e^{-\sigma \sin \theta}\right)-\frac{3}{2}\left[\frac{e^{\sigma \sin \theta}-e^{-\sigma \sin \theta}}{\sigma \sin \theta}\right]+2\right] \tag{54}
\end{equation*}
$$

which is the exact solution rendered in integral form.

## VIII. APPLICATION OF THE METHOD OF STEEPEST DESCENTS TO OBTAIN THE ASYMPTOTIC EXPANSION OF THE EXACT SOLUTION

In order to apply the method of steepest descents [19], we note that the exact solution [Eq. (54)] has no singularity at $\theta=0$ and has a saddle point at $\theta=\pi / 2$. Since the saddle point is at $\theta=\pi / 2$, it will be convenient to replace $\theta$ by $\pi / 2-\theta$ in Eq. (54), so that $\theta=0$ is now the saddle. Thus

$$
\begin{equation*}
\frac{T_{\|}}{\tau_{N}}=M\left(1, \frac{5}{2}, \sigma\right)-\frac{3 e^{\sigma} \sigma^{-2}}{4 M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)} \int_{0}^{\pi / 2} d \theta \frac{\sqrt{1+\cos \theta}}{\cos \theta}\left[\frac{1}{2}\left(e^{\sigma \cos \theta}+e^{-\sigma \cos \theta}\right)+2-\frac{3}{2}\left(\frac{e^{\sigma \cos \theta}-e^{-\sigma \cos \theta}}{\sigma \cos \theta}\right)\right] \tag{55}
\end{equation*}
$$

Let us now write

$$
\begin{equation*}
J=\int_{0}^{\pi / 2} d \theta e^{\sigma \cos \theta} \boldsymbol{G}(\theta) \tag{56}
\end{equation*}
$$

where

$$
\begin{equation*}
G(\theta)=\frac{\sqrt{1+\cos \theta}}{2 \cos \theta}\left(1-\frac{3}{\sigma \cos \theta}\right) \tag{57}
\end{equation*}
$$

Thus, referring to Appendix D , we have in accordance with the method of steepest descents,

$$
\begin{align*}
J= & \int_{0}^{\pi / 2}\left[G(0)+\frac{\theta^{2}}{2} G^{\mathrm{II}}(0)+\frac{\theta^{4}}{24} G^{\mathrm{IV}}(0)+\cdots\right] \\
& \times \exp \left[\sigma-\frac{\sigma \theta^{2}}{2}+\frac{\sigma \theta^{4}}{24}\right] d \theta \\
= & \frac{\sqrt{\pi} e^{\sigma}}{2 \sqrt{\sigma}}\left[1-\frac{5}{2 \sigma}\right] \tag{58}
\end{align*}
$$

whence, on using Eqs. (41) and (58), we obtain

$$
\begin{equation*}
\frac{T_{\|}}{\tau_{N}} \cong \frac{\sqrt{\pi}}{2} e^{\sigma} \sigma^{-3 / 2}\left(1+\frac{1}{\sigma}\right) \tag{59}
\end{equation*}
$$

in agreement with Storonkin $[31,32$ ] and Brown [33]
when their results are truncated at the term of order $\sigma^{-1}$.
Equation (59) is compared with the exact solution and Brown's asymptotic formula [Eq. (46)] in Table II and Fig. 1. It is apparent that Eq. (59) reproduces the asymptote more accurately than Eq. (46) for $\sigma \leq 2.5$ (see Table II). Brown's formula [Eq. (5)] yields a closer approximation to the exact solution for $\sigma$ in the range 1.5-2.5. If the $1 / \sigma^{2}$ term is included in the asymptotic expansion one finds after a tedious calculation that

$$
\begin{equation*}
\frac{T_{\|}}{\tau_{N}} \cong \frac{\sqrt{\pi}}{2} e^{\sigma} \sigma^{-3 / 2}\left[1+\frac{1}{\sigma}+\frac{7}{4 \sigma^{2}}\right] \tag{60}
\end{equation*}
$$

This formula is in agreement with Brown's calculation [33] and is shown in Table II. It provides an even closer approximation to the asymptotic behavior for large $\sigma$. Another approximation that has been used to estimate the relaxation time is the inverse of the effective eigenvalue $[5,23,24]$. The effective relaxation time is found by evaluating Eq. (7) for $n=1$ at $t=0$ and Eq. (20). We have

$$
\begin{equation*}
\tau_{N} \dot{f}_{1}(0)+\left[1-\frac{2 \sigma}{5}\right] f_{1}(0)=-\frac{2}{5} \sigma f_{3}(0) \tag{61}
\end{equation*}
$$

The effective eigenvalue is then [cf. Eq. (20)]

$$
\begin{equation*}
\lambda_{\mathrm{ef}}=-\frac{\dot{f}_{1}(0)}{f_{1}(0)}=\frac{1}{\tau_{N}}\left(1-\frac{2 \sigma}{5}+\frac{2 \sigma}{5} \frac{f_{3}(0)}{f_{1}(0)}\right) . \tag{62}
\end{equation*}
$$

Thus according to Eq. (15) the effective relaxation time $\tau_{\text {ef }}=\lambda_{\text {ef }}^{-1}$ is given by

$$
\begin{equation*}
\frac{\tau_{\mathrm{ef}}}{\tau_{N}}=\frac{1}{1-\frac{2 \sigma}{5}+\frac{2 \sigma}{5} \frac{6 \sigma}{35} \frac{M\left(\frac{5}{2}, \frac{9}{2}, \sigma\right)}{M\left(\frac{3}{2}, \frac{5}{2}, \sigma\right)}} \tag{63}
\end{equation*}
$$

It is apparent from Fig. 2 that the effective-eigenvalue method is inadequate when applied to the longitudinal relaxation, as noted in [23,24], because $\tau_{\text {ef }}$ cannot reproduce the behavior of $T_{\|}$in the large- $\sigma$ limit since the asymptotic behavior of $f_{3}(0) / f_{1}(0)$ in Eq. (62) and $M(1,5 / 2, \sigma)$ in Eq. (40) differs by a factor of $e^{\sigma}$. One can obtain from the data of Table I that the condition (21) does not hold in the case under consideration.

## IX. CONCLUSIONS

We have shown in this paper how one may have exact solutions [Eqs. (33) and (40)] for the longitudinal susceptibility $\chi_{\|}(\omega)$ and correlation time $T_{\|}$for a single-domain ferromagnetic particle for the simple uniaxial potential $K \sin ^{2} \vartheta$. The formula for $T_{\|}$contains the previous result of Brown [3] as the limiting case of high potential barriers and for low potential barriers it reduces to that of perturbation theory in $\sigma$. The crucial steps which yield the solution in closed form are, first, the representation of the correlation time as the zero-frequency limit of the Laplace transform of the aftereffect functions, and second, the fact that the aftereffect solution is governed by a three-term recurrence relation for $\widetilde{f}_{2 n+1}(s)$. This allows us to express the $\widetilde{f}_{2 n+1}(0)$ in terms of Kummer's functions. Thus the method may be extended to the correlation times of the higher-order averages of the aftereffect solution when these are of interest.

We remark that the above method will apply to any problem where the solution of the Fokker-Planck equation may be reduced to a three-term recurrence relation (see e.g., $[16,21,29]$ ). Moreover, our method of finding the Laplace transform of the aftereffect function, described in detail in Appendix A, may be easily extended when the $\widetilde{S}_{n}(s)$ are matrix continued fractions. This is useful in problems involving diffusion in more complex potentials $[34,35]$ and in phase space.
The matrix representation of the problem [Eq. (9)] is inconvenient for the recognition of the existence of a solution in closed form. However, it is extremely useful for numerical calculations because it is not subject to the restriction (as the continued fraction method is) that the differential-difference equations constitute a three-term recurrence relation. Thus the matrix procedure of Eq. (9) is far more general than the continued fraction one. For example, it may be used to calculate the correlation time for uniaxial anisotropy in the presence of an external field H. Here it is not obvious that $T_{\|}$can be expressed in terms of hypergeometric functions since the underlying recurrence relation for arbitrary field strength $|\mathbf{H}|$ is a five-term one. We reiterate in connection with the matrix


FIG. 2. Exact solution [Eq. (40)] for the longitudinal correlation time $T_{\|} / \tau_{N}$ (solid line) compared with the solution rendered by the effective eigenvalue [Eq. (63)] (dashed line).
formulation of the problem that the calculation of $T_{\|}$ from Eq. (27) simply requires one to calculate $\mathbf{A}^{-1}$. On the other hand, the representation of the correlation time as $2 \tau_{N} \lambda_{1}^{-1}$ compels one to solve numerically a high-order polynomial equation-Eq. (11) in $s$. The calculation of $\mathbf{A}^{-1}$ is in general much easier than solving such a polynomial equation.

It is apparent from the results [Eqs. (59) and (60)] of the method of steepest descents that the asymptotic corrections to Brown's formula [Eq. (46)] given in Eqs. (59) and (60) are necessary in order that the asymptotic expansion should accurately represent the solution for large $\sigma$. In view of the ease of computation of the exact solution, which in effect is just

$$
\frac{T_{\|}}{\tau_{N}}=g(\sigma)
$$

it appears that the previously used formulas are now redundant.

The exact solution of this problem is of particular importance in the context of the remarks of Klik and Gunther [37] concerning the application of the uniaxial model to real superparamagnets, in particular the $T^{-1 / 2}$ behavior of the relaxation rate prefactor $\sigma^{3 / 2} \tau_{N}^{-1}$ in Brown's asymptote [Eq. (5)]. The reader is referred to their paper [37] and that of Bessais, Ben Jaffel, and Dormann [12] for a detailed discussion. A description of the difficulties accompanying the comparison of theoretical formulas for $T_{\|}$with experiment is given in Refs. [12] and [13].

Our results, with a few changes in notation, govern the longitudinal relaxation behavior in the theory of dielectric relaxation of nematic liquid crystals given by Martin, Meier, and Saupe [10]. We note that the present method yields an analytic expression for the transverse relaxation time in the theory of Martin, Meier, and Saupe [10]. This result, however, does not carry over to magnetic relaxation, unless the gyromagnetic term in $\sigma / \alpha$ in Brown's equation (3) can be ignored.

In conclusion, we emphasize that we have confined ourselves in this paper to the resolution of the purely mathematical question posed by the exact calculation of the longitudinal relaxation time for the simple uniaxial anisotropy from Brown's equation (3). This problem may
now be considered as completely solved due to the existence of the series solution [Eq. (40)]. In view of the idealized nature of the simple uniaxial potential model as discussed in [37], the method described in the paper should properly be regarded as a convenient starting point for the analytical treatment of more realistic potentials which incorporate the azimuthal angle dependence, e.g., cubic anisotropy.

## ACKNOWLEDGMENTS

We would like to thank the British Council and EOLAS for grants in support of this work. D.S.F.C. acknowledges UK-SERC rolling grant support under GR/H 59862, and partial support by the National Science Foundation through a grant at the Institute for Theoretical Atomic and Molecular Physics at Harvard University and the Smithsonian Astrophysics Observatory. E.S.M. would like to thank the University of Dar-

Es-Salaam for granting him study leave and HEDCO for financial support. J.T.W. would like to thank Dublin City University for granting him study leave and financial support. We thank Professor B. K. P. Scaife for introducing us to this problem. We further thank Professor R. W. Chantrell, Dr. M. El. Hilo, Dr. P. C. Fannin, Dr. A. Lyberatos, and Dr. M. San. Miguel for helpful conversations.

## APPENDIX A: CALCULATION <br> OF THE LAPLACE TRANSFORM $\tilde{f}_{1}(s)$ OF THE AFTEREFFECT FUNCTION

Following Cresser et al. [27] and Coffey [28] we seek a solution of Eq. (28) in the form

$$
\begin{equation*}
\widetilde{R}_{n}(s)=\widetilde{S}_{n}(s)+\widetilde{Q}_{n}(s), \tag{A1}
\end{equation*}
$$

where $\widetilde{S}_{n}$ is given by Eq. (30). Equation (28) then becomes

$$
\begin{equation*}
\left[\frac{2 \tau_{N} s}{n(n+1)}+1-\frac{2 \sigma}{(2 n-1)(2 n+3)}\right] \widetilde{f}_{n-2} \widetilde{Q}_{n}+\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)}\left[\widetilde{Q}_{n+2} \widetilde{f}_{n-2}\left(\widetilde{S}_{n}+\widetilde{Q}_{n}\right)+\widetilde{Q}_{n} \widetilde{S}_{n+2} \widetilde{f}_{n-2}\right]=\frac{2 \tau_{N} f_{n}(0)}{n(n+1)} . \tag{A2}
\end{equation*}
$$

We now introduce

$$
\begin{equation*}
q_{n}=\widetilde{Q}_{n} \widetilde{f}_{n-2} \tag{A3}
\end{equation*}
$$

hence

$$
\begin{equation*}
\left[\frac{2 \tau_{N} s}{n(n+1)}+1-\frac{2 \sigma}{(2 n-1)(2 n+3)}\right] q_{n}+\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)}\left[q_{n+2}+q_{n} \tilde{S}_{n+2}\right]=\frac{2 \tau_{N} f_{n}(0)}{n(n+1)} \tag{A4}
\end{equation*}
$$

Equation (A4) may be solved for $q_{n}$ to get

$$
\begin{align*}
q_{n} & =\frac{\frac{2 \tau_{N} f_{n}(0)}{n(n+1)}-\frac{2 \sigma(n+2)}{(2 n-1)(2 n+3)} q_{n+2}}{\frac{2 \tau_{N} s}{n(n+1)}+1-\frac{2 \sigma}{(2 n-1)(2 n+3)}+\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)} \widetilde{S}_{n+2}(s)} \\
& =a_{n}\left[\left(\tau_{N} / \sigma\right) f_{n}(0)-b_{n} q_{n+2}\right] \widetilde{S}_{n}(s), \tag{A5}
\end{align*}
$$

where

$$
\begin{equation*}
a_{n}=\frac{4 n^{2}-1}{n\left(n^{2}-1\right)}, \quad b_{n}=\frac{n(n+1)(n+2)}{(2 n+1)(2 n+3)} \tag{A6}
\end{equation*}
$$

However, from Eqs. (A3) and (A5) we obtain

$$
\begin{equation*}
f_{n}(s)=\left\{\widetilde{f}_{n-2}(s)+a_{n}\left[\left(\tau_{N} / \sigma\right) f_{n}(0)-b_{n} q_{n+2}\right]\right\} \widetilde{S}_{n}(s) \tag{A7}
\end{equation*}
$$

In particular, for $n=1$ we have

$$
\begin{equation*}
\widetilde{f}_{1}(s)=\frac{1}{G(\sigma, s)}\left[\tau_{N} f_{1}(0)-\frac{2}{5} \sigma q_{3}\right] \tag{A8}
\end{equation*}
$$

where

$$
\begin{equation*}
G(\sigma, s)=s \tau_{N}+1-\frac{2}{5} \sigma+\frac{2}{5} \sigma \widetilde{S}_{3}(s) . \tag{A9}
\end{equation*}
$$

Substituting for $q_{3}$ in Eq. (A8) from Eq. (A5) for $n=3$ we have

$$
\begin{equation*}
\widetilde{f}_{1}(s)=\frac{1}{G(\sigma, s)}\left\{\tau_{N} f_{1}(0)-\sigma a_{3} b_{1}\left[\left(\tau_{N} / \sigma\right) f_{3}(0)-b_{3} q_{5}\right] \widetilde{S}_{3}(s)\right\} \tag{A10}
\end{equation*}
$$

so that

$$
\begin{equation*}
\tilde{f}_{1}(s)=\frac{\tau_{N}}{G(\sigma, s)}\left[f_{1}(0)+\sum_{n=0}^{\infty}(-1)^{n+1} f_{2 n+3}(0) \prod_{k=1}^{n} a_{2 k+3} b_{2 k+1} \tilde{S}_{2 k+3}(s)\right] \tag{A11}
\end{equation*}
$$

Equation (A11) may be further simplified if we write out the product $a_{2 k+3} b_{2 k+1}$ explicitly. On using

$$
\begin{aligned}
\prod_{k=0}^{n} a_{2 k+3} b_{2 k+1} & =\prod_{k=0}^{n} \frac{(4 k+7)(2 k+1)}{2(k+2)(4 k+3)} \\
& =\frac{4}{3}\left(n+\frac{7}{4}\right)\left(n+\frac{1}{2}\right) \frac{\Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+3) \Gamma\left(\frac{1}{2}\right)}
\end{aligned}
$$

we can reduce Eq. (A11) to Eq. (31).

## APPENDIX B: EVALUATION OF THE INITIAL CONDITIONS AS A RATIO OF TWO KUMMER FUNCTIONS

The purpose of this appendix is to demonstrate how the initial conditions [Eq. (13)] may be written as a ratio of two Kummer functions. In order to evaluate the integral in the numerator of Eq. (13) we first expand the exponential in powers of $\sigma$ and next expand $x^{2 r+1}$ as a finite series of the Legendre polynomials $P_{2 n+1}(x)$ using Eq. (12.4.6b) of Arfken [26], namely,
$x^{2 r+1}=\sum_{n=0}^{r} \frac{2^{2 n+1}(4 n+3)(2 r+1)!(r+n+1)!}{(2 r+2 n+3)!(r-n)!} P_{2 n+1}(x)$.
(B1)
Thus, on using the orthogonality properties of the Legendre polynomials and equation ([36], p. 580)

$$
\begin{equation*}
\int_{0}^{1} \exp \left(\sigma x^{2}\right) d x=M\left(\frac{1}{2}, \frac{3}{2}, \sigma\right), \tag{B2}
\end{equation*}
$$

we have
$f_{2 n+1}(0)=\xi \frac{2^{2 n+1}}{M\left(\frac{1}{2}, \frac{3}{2}, \sigma\right)} \sum_{r=n}^{\infty} \frac{\sigma^{r}}{r!} \frac{(2 r+1)!(r+n+1)!}{(2 r+2 n+3)!(r-n)!}$.

We now eliminate the summation in Eq. (B3) by writing $r=n+N$ and using the recurrence relations (39) and the definition of the Kummer function [Eq. (14)]. The desired result is Eq. (15).

## APPENDIX C: REPRESENTATION OF $\widetilde{S}_{2 k+1}(0)$ <br> AS A RATIO OF TWO KUMMER FUNCTIONS

We require $\widetilde{S}_{2 k+1}(0)$ as a ratio of two Kummer functions. This is accomplished by noting that Eq. (30) can be rearranged to yield after simple algebra

$$
\begin{equation*}
1-\widetilde{S}_{n}(0)=\frac{1}{1+\frac{2 \sigma(n-1)}{1-\frac{2 \sigma(n+2)}{(2 n+1)(2 n+3)}\left[1-\widetilde{S}_{n+2}(0)\right]}} \tag{C1}
\end{equation*}
$$

On comparing Eq. (C1) with the continued fraction ([38], p. 347)
$\frac{M(a+1, b+1, z)}{M(a, b, z)}$

$$
\begin{equation*}
=\frac{1}{1+\frac{\frac{z(b-a)}{b(b+1)}}{1-\frac{z(a+1)}{(b+1)(b+2)} \frac{M(a+2, b+3, z)}{M(a+1, b+2, z)}}}, \tag{C2}
\end{equation*}
$$

we obtain $a=n / 2$ and $b=n-1 / 2$ so that

$$
\begin{align*}
\widetilde{S}_{n}(0) & =1-\frac{M\left(1+n / 2, n+\frac{1}{2},-\sigma\right)}{M\left(n / 2, n-\frac{1}{2},-\sigma\right)} \\
& =1-\frac{M\left((n-1) / 2, n+\frac{1}{2}, \sigma\right)}{M\left((n-1) / 2, n-\frac{1}{2}, \sigma\right)} . \tag{C3}
\end{align*}
$$

Here we have used Eq. (48). Further on, using the recurrence relation ([18], Eq. 13.4.4)
$M(a, b-1, z)-M(a, b, z)=\frac{a z}{b(b-1)} M(a+1, b+1, z)$,
we have from Eq. (C3)

$$
\begin{equation*}
\widetilde{S}_{n}(0)=\frac{2(n-1) \sigma}{\left(4 n^{2}-1\right)} \frac{M\left((n+1) / 2, n+\frac{3}{2}, \sigma\right)}{M\left((n-1) / 2, n-\frac{1}{2}, \sigma\right)} . \tag{C4}
\end{equation*}
$$

Equation (C5) reduces to Eq. (36) at $n=2 k+1$. Also on using Eq. (C3) and the properties of Kummer's functions [18]
$M(0, b, z)=1$,
$b M(a, b, z)-b M(a-1, b, z)-z M(a, b+1, z)=0$,
we can express the leading term of Eq. (34) as follows:

$$
\begin{align*}
\frac{1}{1-\frac{2 \sigma}{5}+\frac{2 \sigma}{5} \widetilde{S}_{3}(0)} & =\frac{1}{1-\frac{2 \sigma}{5} \frac{M\left(1, \frac{7}{2}, \sigma\right)}{M\left(1, \frac{5}{2}, \sigma\right)}} \\
& =M\left(1, \frac{5}{2}, \sigma\right) \tag{C8}
\end{align*}
$$

## APPENDIX D: DERIVATION OF A CLOSED-FORM EXPRESSION FOR THE SERIES $\boldsymbol{F}(\theta)$

In our derivation of the integral form of the exact solution [Eq. (55)] from the series solution Eq. (40) we require proof that Eq. (52) may be expressed in the closed form of Eq. (53). In order to accomplish this we recall that [[18] Eq. (11.1.1)]

$$
\begin{equation*}
\int_{0}^{z} t^{\mu} J_{v}(t) d t=\frac{z^{\mu} \Gamma\left(\frac{v+\mu+1}{2}\right)}{\Gamma\left(\frac{v-\mu+1}{2}\right)} \sum_{k=0}^{\infty} \frac{(v+2 k+1) \Gamma\left(\frac{v-\mu+1}{2}+k\right)}{\Gamma\left(\frac{v+\mu+3}{2}+k\right)} J_{v+2 k+1}(z) \tag{D1}
\end{equation*}
$$

where $J_{v}(t)$ is the Bessel function of the first kind of order $v$ and $\operatorname{Re}(v+\mu+1)>1$. Equation (D1) holds for complex $z$. Let us replace $z$ by $i z$ in Eq. (D1) and suppose that $k=n-1$, whence Eq. (D1) becomes

$$
\begin{equation*}
\int_{0}^{z} t^{\mu} I_{v}(t) d t=z^{\mu} \frac{\Gamma\left(\frac{v+\mu+1}{2}\right)}{\Gamma\left(\frac{v+\mu+1}{2}\right)} \sum_{n=1}^{\infty}(-1)^{n-1} \frac{(v+2 n-1) \Gamma\left(\frac{v-\mu+1}{2}+n\right)}{\Gamma\left(\frac{v+\mu+1}{2}+n\right)} I_{v+2 n-1}(z), \tag{D2}
\end{equation*}
$$

where we have used Eq. (9.6.3) of Ref. [18], namely,

$$
\begin{equation*}
J_{v}(i z)=e^{i \pi v / 2} I_{v}(z) \tag{D3}
\end{equation*}
$$

Hence referring to Eq. (52) we have

$$
\begin{equation*}
\int_{0}^{\sigma \sin \theta} t^{1 / 2} I_{5 / 2}(t) d t=-2 \sqrt{\sigma \sin \theta} \frac{\Gamma(2)}{\Gamma\left(\frac{3}{2}\right)} \sum_{n=1}^{\infty} \frac{(-1)^{n}\left(n+\frac{3}{4}\right) \Gamma\left(n+\frac{1}{2}\right)}{\Gamma(n+2)} I_{2 n+3 / 2}(\sigma \sin \theta) \tag{D4}
\end{equation*}
$$

so that
$F(\theta)=-\frac{1}{2} \frac{\Gamma\left(\frac{3}{2}\right)}{\sqrt{\sigma \sin \theta} \Gamma(2)} \int_{0}^{\sigma \sin \theta} t^{1 / 2} I_{5 / 2}(t) d t$.
This may be further simplified using the properties of the spherical Bessel functions [18]. We have

$$
\begin{equation*}
\int_{0}^{x} t^{1 / 2} I_{5 / 2}(t) d t=\sqrt{2 / \pi} \int_{0}^{x}\left[\left(\frac{3}{t^{3}}+\frac{1}{t}\right) \sinh t-\frac{3}{t^{2}} \cosh t\right] t d t=\sqrt{2 / \pi}\left(\cosh x-3 \frac{\sinh x}{x}+2\right) \tag{D6}
\end{equation*}
$$

so that finally

$$
\begin{equation*}
F(\theta)=\frac{1}{\sqrt[3]{2} \sqrt{\sigma \sin \theta}}\left[\cosh (\sigma \sin \theta)-\frac{3 \sinh (\sigma \sin \theta)}{\sigma \sin \theta}+2\right] \tag{D7}
\end{equation*}
$$

which is the desired closed-form expression for the series $F(\theta)$.
We now briefly sketch the calculation of the derivatives of the function $G(\theta)$ from Eq. (59) used in the evaluation of the integral Eq. (58) by the method of steepest descents since it is tedious for the reader to reproduce the calculation. We arrange $G(\theta)$ as

$$
\begin{equation*}
G(\theta)=\frac{1}{\sqrt{2}} \frac{\cos (\theta / 2)}{\cos \theta}-\frac{3}{\sigma \sqrt{2}} \frac{\cos (\theta / 2)}{\cos ^{2} \theta}, \tag{D8}
\end{equation*}
$$

hence

$$
\begin{equation*}
G(0)=\frac{1}{\sqrt{2}}-\frac{3}{\sigma \sqrt{2}} \tag{D9}
\end{equation*}
$$

Further,

$$
\begin{equation*}
G^{\prime}(\theta)=\frac{1}{\sqrt{2}}\left[-\frac{\sin (\theta / 2)}{2 \cos \theta}+\frac{\sin \theta \cos (\theta / 2)}{\cos ^{2} \theta}\right)-\frac{3}{\sigma \sqrt{2}}\left(-\frac{\sin (\theta / 2)}{2 \cos ^{2} \theta}+\frac{2 \sin \theta \cos (\theta / 2)}{\cos ^{3} \theta}\right) \tag{D10}
\end{equation*}
$$

so that

$$
\begin{equation*}
G^{\prime}(0)=0 . \tag{D11}
\end{equation*}
$$

Now

$$
\begin{align*}
G^{\prime \prime}(\theta)= & \frac{1}{\sqrt{2}}\left[\frac{3 \cos (\theta / 2)}{4 \cos \theta}-\frac{\sin \theta \sin (\theta / 2)}{\cos ^{2} \theta}+\frac{2 \sin ^{2} \theta \cos (\theta / 2)}{\cos ^{3} \theta}\right) \\
& -\frac{3}{\sigma^{\sqrt{2}}}\left(\frac{7 \cos (\theta / 2)}{4 \cos ^{2} \theta}-\frac{2 \sin \theta \sin (\theta / 2)}{\cos ^{3} \theta}+\frac{6 \sin ^{2} \theta \cos (\theta / 2)}{\cos ^{4} \theta}\right) \tag{D12}
\end{align*}
$$

Thus

$$
\begin{equation*}
G^{\prime \prime}(0)=\frac{3 \sqrt{2}}{8}-\frac{21 \sqrt{2}}{8 \sigma} \tag{D13}
\end{equation*}
$$

Further,

$$
\begin{align*}
& \text { ther, } \begin{aligned}
G^{\prime \prime \prime}(\theta)= & \frac{1}{\sqrt{2}}\left[-\frac{11 \sin (\theta / 2)}{8 \cos \theta}+\frac{17 \sin \theta \cos (\theta / 2)}{4 \cos ^{2} \theta}-\frac{3 \sin ^{2} \theta \cos (\theta / 2)}{\cos ^{3} \theta}+\frac{6 \sin ^{3} \theta \cos (\theta / 2)}{\cos ^{4} \theta}\right] \\
& -\frac{3}{\sigma \sqrt{2}}\left[-\frac{23 \sin (\theta / 2)}{8 \cos ^{2} \theta}+\frac{29 \sin \theta \cos (\theta / 2)}{2 \cos ^{3} \theta}-\frac{9 \sin ^{2} \theta \sin (\theta / 2)}{\cos ^{4} \theta}+\frac{24 \sin ^{3} \theta \cos (\theta / 2)}{\cos ^{5} \theta}\right]
\end{aligned},
\end{align*}
$$

TABLE III. Table of values $T_{\|} / \tau_{N}$ from the exact solution [Eq. (40)].

| $\sigma$ | $T_{\\|} / \tau_{N}$ | $\sigma$ | $T_{\\|} / \tau_{N}$ | $\sigma$ | $T_{\\|} / \tau_{N}$ | $\sigma$ | $T_{\\|} / \tau_{N}$ | $\sigma$ | $T_{\\|} / \tau_{N}$ | $\sigma$ | $T_{\\|} / \tau_{N}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| 0.1 | 1.041 | 5.1 | 15.616 | 10.1 | 751.43 | 15.1 | 58605.0 | 20.1 | $5.5576 \times 10^{6}$ | 25.1 | $5.8463 \times 10^{8}$ |
| 0.2 | 1.0843 | 5.2 | 16.724 | 10.2 | 817.27 | 15.2 | 64098.0 | 20.2 | $6.0949 \times 10^{6}$ | 25.2 | $6.4217 \times 10^{8}$ |
| 0.3 | 1.1299 | 5.3 | 17.919 | 10.3 | 889.03 | 15.3 | 70111.0 | 20.3 | $6.6843 \times 10^{6}$ | 25.3 | $7.0538 \times 10^{8}$ |
| 0.4 | 1.1779 | 5.4 | 19.208 | 10.4 | 967.26 | 15.4 | 76693.0 | 20.4 | $7.3311 \times 10^{6}$ | 25.4 | $7.7484 \times 10^{8}$ |
| 0.5 | 1.2286 | 5.5 | 20.601 | 10.5 | 1052.5 | 15.5 | 83899.0 | 20.5 | $8.0407 \times 10^{6}$ | 25.5 | $8.5115 \times 10^{8}$ |
| 0.6 | 1.282 | 5.6 | 22.104 | 10.6 | 1145.5 | 15.6 | 91788.0 | 20.6 | $8.8194 \times 10^{6}$ | 25.6 | $9.35 \times 10^{8}$ |
| 0.7 | 1.3385 | 5.7 | 23.728 | 10.7 | 1246.9 | 15.7 | $1.0043 \times 10^{5}$ | 20.7 | $9.6739 \times 10^{6}$ | 25.7 | $1.0271 \times 10^{9}$ |
| 0.8 | 1.3982 | 5.8 | 25.483 | 10.8 | 1357.5 | 15.8 | $1.0988 \times 10^{5}$ | 20.8 | $1.0612 \times 10^{7}$ | 25.8 | $1.1284 \times 10^{9}$ |
| 0.9 | 1.4613 | 5.9 | 27.38 | 10.9 | 1478.1 | 15.9 | $1.2024 \times 10^{5}$ | 20.9 | $1.1641 \times 10^{7}$ | 25.9 | $1.2396 \times 10^{9}$ |
| 1.0 | 1.528 | 6.0 | 29.43 | 11.0 | 1609.6 | 16.0 | $1.3158 \times 10^{5}$ | 21.0 | $1.277 \times 10^{7}$ | 26.0 | $1.3619 \times 10^{9}$ |
| 1.1 | 1.5986 | 6.1 | 31.647 | 11.1 | 1753.1 | 16.1 | $1.44 \times 10^{5}$ | 21.1 | $1.4009 \times 10^{7}$ | 26.1 | $1.4963 \times 10^{9}$ |
| 1.2 | 1.6733 | 6.2 | 34.046 | 11.2 | 1909.7 | 16.2 | $1.5761 \times 10^{5}$ | 21.2 | $1.5369 \times 10^{7}$ | 26.2 | $1.6439 \times 10^{9}$ |
| 1.3 | 1.7525 | 6.3 | 36.641 | 11.3 | 2080.5 | 16.3 | $1.7251 \times 10^{5}$ | 21.3 | $1.6862 \times 10^{7}$ | 26.3 | $1.8062 \times 10^{9}$ |
| 1.4 | 1.8364 | 6.4 | 39.45 | 11.4 | 2266.9 | 16.4 | $1.8883 \times 10^{5}$ | 21.4 | $1.85 \times 10^{7}$ | 26.4 | $1.9845 \times 10^{9}$ |
| 1.5 | 1.9254 | 6.5 | 42.49 | 11.5 | 2470.4 | 16.5 | $2.067 \times 10^{5}$ | 21.5 | $2.0299 \times 10^{7}$ | 26.5 | $2.1804 \times 10^{9}$ |
| 1.6 | 2.0199 | 6.6 | 45.783 | 11.6 | 2692.5 | 16.6 | $2.2629 \times 10^{5}$ | 21.6 | $2.2273 \times 10^{7}$ | 26.6 | $2.3958 \times 10^{9}$ |
| 1.7 | 2.1202 | 6.7 | 49.349 | 11.7 | 2934.9 | 16.7 | $2.4774 \times 10^{5}$ | 21.7 | $2.4439 \times 10^{7}$ | 26.7 | $2.6325 \times 10^{9}$ |
| 1.8 | 2.2267 | 6.8 | 53.212 | 11.8 | 3199.6 | 16.8 | $2.7124 \times 10^{5}$ | 21.8 | $2.6818 \times 10^{7}$ | 26.8 | $2.8927 \times 10^{9}$ |
| 1.9 | 2.3399 | 6.9 | 57.398 | 11.9 | 3488.5 | 16.9 | $2.9699 \times 10^{5}$ | 21.9 | $2.9428 \times 10^{7}$ | 26.9 | $3.1786 \times 10^{9}$ |
| 2.0 | 2.4603 | 7.0 | 61.935 | 12.0 | 3804.0 | 17.0 | $3.2521 \times 10^{5}$ | 22.0 | $3.2294 \times 10^{7}$ | 27.0 | $3.4929 \times 10^{9}$ |
| 2.1 | 2.5884 | 7.1 | 66.853 | 12.1 | 4148.6 | 17.1 | $3.5612 \times 10^{5}$ | 22.1 | $3.5441 \times 10^{7}$ | 27.1 | $3.8383 \times 10^{9}$ |
| 2.2 | 2.7248 | 7.2 | 72.186 | 12.2 | 4524.9 | 17.2 | $3.8999 \times 10^{5}$ | 22.2 | $3.8895 \times 10^{7}$ | 27.2 | $4.218 \times 10^{9}$ |
| 2.3 | 2.8701 | 7.3 | 77.971 | 12.3 | 4935.9 | 17.3 | $4.2711 \times 10^{5}$ | 22.3 | $4.2687 \times 10^{7}$ | 27.3 | $4.6353 \times 10^{9}$ |
| 2.4 | 3.0249 | 7.4 | 84.245 | 12.4 | 5384.8 | 17.4 | $4.6779 \times 10^{5}$ | 22.4 | $4.6851 \times 10^{7}$ | 27.4 | $5.0941 \times 10^{9}$ |
| 2.5 | 3.1899 | 7.5 | 91.053 | 12.5 | 5875.2 | 17.5 | $5.1237 \times 10^{5}$ | 22.5 | $5.1422 \times 10^{7}$ | 27.5 | $5.5984 \times 10^{9}$ |
| 2.6 | 3.366 | 7.6 | 98.442 | 12.6 | 6411.0 | 17.6 | $5.6123 \times 10^{5}$ | 22.6 | $5.6441 \times 10^{7}$ | 27.6 | $6.1527 \times 10^{9}$ |
| 2.7 | 3.5539 | 7.7 | 106.46 | 12.7 | 6996.5 | 17.7 | $6.1479 \times 10^{5}$ | 22.7 | $6.1952 \times 10^{7}$ | 27.7 | $6.762 \times 10^{9}$ |
| 2.8 | 3.7546 | 7.8 | 115.17 | 12.8 | 7636.1 | 17.8 | $6.7348 \times 10^{5}$ | 22.8 | $6.8003 \times 10^{7}$ | 27.8 | $7.4318 \times 10^{9}$ |
| 2.9 | 3.969 | 7.9 | 124.62 | 12.9 | 8335.2 | 17.9 | $7.3782 \times 10^{5}$ | 22.9 | $7.4648 \times 10^{7}$ | 27.9 | $8.1682 \times 10^{9}$ |
| 3.0 | 4.1982 | 8.0 | 134.89 | 13.0 | 9099.1 | 18.0 | $8.0835 \times 10^{5}$ | 23.0 | $8.1944 \times 10^{7}$ | 28.0 | $8.9777 \times 10^{9}$ |
| 3.1 | 4.4434 | 8.1 | 146.05 | 13.1 | 9934.1 | 18.1 | $8.8566 \times 10^{5}$ | 23.1 | $8.9956 \times 10^{7}$ | 28.1 | $9.8676 \times 10^{9}$ |
| 3.2 | 4.7056 | 8.2 | 158.17 | 13.2 | 10847.0 | 18.2 | $9.7041 \times 10^{5}$ | 23.2 | $9.8754 \times 10^{7}$ | 28.2 | $1.0846 \times 10^{10}$ |
| 3.3 | 4.9863 | 8.3 | 171.34 | 13.3 | 11845.0 | 18.3 | $1.0633 \times 10^{6}$ | 23.3 | $1.0842 \times 10^{8}$ | 28.3 | $1.1922 \times 10^{10}$ |
| 3.4 | 5.287 | 8.4 | 185.65 | 13.4 | 12935.0 | 18.4 | $1.1652 \times 10^{6}$ | 23.4 | $1.1903 \times 10^{8}$ | 28.4 | $1.3104 \times 10^{10}$ |
| 3.5 | 5.6091 | 8.5 | 201.21 | 13.5 | 14128.0 | 18.5 | $1.2769 \times 10^{6}$ | 23.5 | $1.3068 \times 10^{8}$ | 28.5 | $1.4404 \times 10^{10}$ |
| 3.6 | 5.9544 | 8.6 | 218.13 | 13.6 | 15432.0 | 18.6 | $1.3994 \times 10^{6}$ | 23.6 | $1.4348 \times 10^{8}$ | 28.6 | $1.5834 \times 10^{10}$ |
| 3.7 | 6.3247 | 8.7 | 236.52 | 13.7 | 16857.0 | 18.7 | $1.5336 \times 10^{6}$ | 23.7 | $1.5754 \times 10^{8}$ | 28.7 | $1.7405 \times 10^{10}$ |
| 3.8 | 6.7219 | 8.8 | 256.53 | 13.8 | 18416.0 | 18.8 | $1.6809 \times 10^{6}$ | 23.8 | $1.7297 \times 10^{8}$ | 28.8 | $1.9133 \times 10^{10}$ |
| 3.9 | 7.1483 | 8.9 | 278.29 | 13.9 | 20122.0 | 18.9 | $1.8424 \times 10^{6}$ | 23.9 | $1.8993 \times 10^{8}$ | 28.9 | $2.1033 \times 10^{10}$ |
| 4.0 | 7.6061 | 9.0 | 301.97 | 14.0 | 21986.0 | 19.0 | $2.0194 \times 10^{6}$ | 24.0 | $2.0856 \times 10^{8}$ | 29.0 | $2.3122 \times 10^{10}$ |
| 4.1 | 8.0979 | 9.1 | 327.73 | 14.1 | 24026.0 | 19.1 | $2.2136 \times 10^{6}$ | 24.1 | $2.2901 \times 10^{8}$ | 29.1 | $2.5419 \times 10^{10}$ |
| 4.2 | 8.6263 | 9.2 | 355.77 | 14.2 | 26258.0 | 19.2 | $2.4266 \times 10^{6}$ | 24.2 | $2.5148 \times 10^{8}$ | 29.2 | $2.7944 \times 10^{10}$ |
| 4.3 | 9.1945 | 9.3 | 386.29 | 14.3 | 28698.0 | 19.3 | $2.6602 \times 10^{6}$ | 24.3 | $2.7617 \times 10^{8}$ | 29.3 | $3.0721 \times 10^{10}$ |
| 4.4 | 9.8054 | 9.4 | 419.51 | 14.4 | 31369.0 | 19.4 | $2.9164 \times 10^{6}$ | 24.4 | $3.0328 \times 10^{8}$ | 29.4 | $3.3775 \times 10^{10}$ |
| 4.5 | 10.463 | 9.5 | 455.68 | 14.5 | 34291.0 | 19.5 | $3.1974 \times 10^{6}$ | 24.5 | $3.3307 \times 10^{8}$ | 29.5 | $3.7133 \times 10^{10}$ |
| 4.6 | 11.17 | 9.6 | 495.06 | 14.6 | 37487.0 | 19.6 | $3.5056 \times 10^{6}$ | 24.6 | $3.6579 \times 10^{8}$ | 29.6 | $4.0826 \times 10^{10}$ |
| 4.7 | 11.932 | 9.7 | 537.96 | 14.7 | 40985.0 | 19.7 | $3.8437 \times 10^{6}$ | 24.7 | $4.0173 \times 10^{8}$ | 29.7 | $4.4886 \times 10^{10}$ |
| 4.8 | 12.752 | 9.8 | 584.68 | 14.8 | 44813.0 | 19.8 | $4.2146 \times 10^{6}$ | 24.8 | $4.4122 \times 10^{8}$ | 29.8 | $4.9351 \times 10^{10}$ |
| 4.9 | 13.636 | 9.9 | 635.57 | 14.9 | 49002.0 | 19.9 | $4.6215 \times 10^{6}$ | 24.9 | $4.8461 \times 10^{8}$ | 29.9 | $5.4262 \times 10^{10}$ |
| 5.0 | 14.589 | 10.0 | 691.02 | 15.0 | 53587.0 | 20.0 | $5.0679 \times 10^{6}$ | 25.0 | $5.3227 \times 10^{8}$ | 30.0 | $5.9662 \times 10^{10}$ |

so that

$$
\begin{equation*}
G^{\prime \prime \prime}(0)=0 . \tag{D15}
\end{equation*}
$$

## APPENDIX E: TABLE OF THE EXACT SOLUTION

In order to facilitate comparison with the results of Brown's asymptotic formula [Eq. (46)] and with experimental observations it is useful to present the exact solution [Eq. (40)] in tabular form. This function is tabulated in Table III for values of $\sigma$ from 0.1 to 30 .

All numerical calculations were performed on a Macintosh II SI with Motorolla 68882 coprocessor running MATHEMATICA 1.2. In the calculation of Eq. (40), it was necessary to take the fist 16 terms from the infinite summation to ensure convergence to five significant digits for values of $\sigma$ up to 60 . The MATHEMATICA computer program for the computation of Eq. (40) is available from the authors [39].
[1] L. Néel, Ann. Geophys. 5, 99 (1949).
[2] A. Aharoni, Phys. Rev. 135A, 447 (1964).
[3] W. F. Brown, Jr., Phys. Rev. 130, 1677 (1963).
[4] W. F. Brown, Jr., Suppl. J. Appl. Phys. 30, 130S (1959).
[5] W. T. Coffey, P. J. Cregg, and Yu. P. Kalmykov, Adv. Chem. Phys. 83, 263 (1993).
[6] C. N. Scully, Ph. D. thesis, The Queen's University of Belfast, 1993.
[7] H. A. Kramers, Physica 7, 387 (1940).
[8] P. Hänggi, P. Talkner, and M. Borkovec, Rev. Mod. Phys. 62, 251 (1990).
[9] C. N. Scully, P. J. Cregg, and D. S. F. Crothers, Phys. Rev. B 45, 474 (1992).
[10] A. J. Martin, G. Meier, and A. Saupe, Symp. Faraday Soc. 5, 119 (1971).
[11] L. Bessais, L. Ben Jaffel, and J. L. Dormann, J. Magn. Magn. Mater. 104, 1565 (1992).
[12] L. Bessais, L. Ben Jaffel, and J. L. Dormann, Phys. Rev. B 45, 7805 (1992).
[13] A. Aharoni, Phys. Rev. B 46, 5434 (1992).
[14] D. A. Garanin, V. V. Ischenko, and L. V. Panina, Teor. Mat. Fiz. 82, 242 (1990) [Theor. Math. Phys. 82, 169 (1990)].
[15] G. Moro and P. L. Nordio, Mol. Phys. 56, 255 (1985).
[16] W. T. Coffey, Yu. P. Kalmykov, and E. S. Massawe, Adv. Chem. Phys. II 85, 667 (1993), p. 667.
[17] M. San Miguel, L. Pesquera, M. A. Rodrigues, and A. Hernández-Machado, Phys. Rev. A 35, 208 (1987).
[18] Handbook of Mathematical Functions, edited by M. Abramowitz and I. Stegun (Dover, New York, 1964).
[19] H. Jeffreys and B. S. Jeffreys, Methods of Mathematical Physics, 2nd ed. (Cambridge University, Cambridge, 1950).
[20] M. R. Spiegel, Laplace Transforms (Schaum, New York, 1965).
[21] W. T. Coffey, Yu. P. Kalmykov, E. S. Massawe, and J. T. Waldron, J. Chem. Phys. 99, 4011 (1993).
[22] H. Bateman, A. Erdélyi, W. Magnus, F. Oberhettinger, and F. G. Tricomi, Higher Transcendental Functions, Bateman Manuscript Project Vol. 1 (McGraw Hill, New York, 1953).
[23] Yu. L. Raikher and M. I. Shliomis, Zh. Eksp. Teor. Fiz. 67, 1060 (1974) [Sov. Phys. JETP. 40, 526 (1974)].
[24] Yu. L. Raǐkher and M. I. Shliomis, Adv. Chem. Phys. (to be published).
[25] S. Roman, An Introduction to Linear Algebra (Saunders, Philadelphia, 1984).
[26] G. Arfken, Mathematical Methods for Physicists, 2nd ed. (Academic, New York, 1970).
[27] J. D. Cresser, D. Hammonds, W. H. Louisell, P. Meystre, and H. Risken, Phys. Rev. 25A, 2226 (1982).
[28] W. T. Coffey, Adv. Chem. Phys. 63, 69 (1985).
[29] W. T. Coffey, Yu. P. Kalmykov, and E. S. Massawe, Phys. Rev. E 48, 77 (1993).
[30] B. K. P. Scaife, Principles of Dielectrics (Oxford University, Oxford, 1989).
[31] B. A. Storonkin, Kristallografiya 30, 841 (1985) [Sov. Phys. Crystallogr. 30, 489 (1985)].
[32] B. A. Storonkin, Theor. Math. Phys. 41, 1098 (1979).
[33] W. F. Brown, Jr., IEEE Trans. Magn. 15, 1196 (1979).
[34] I. Eisenstein and A. Aharoni, Phys. Rev. B 16, 1278 (1977).
[35] I. Eisenstein and A. Aharoni, Phys. Rev. B 16, 1285 (1977).
[36] A. Prudinikov, Yu. A. Brychkov, and O. I. Marichev, Integrals and Series, More Special Functions (Gordon and Breach, New York, 1990), Vol. 3.
[37] I. Klik and L. Gunther, J. Stat. Phys. 60, 473 (1990).
[38] H. S. Wall, Continued Fractions (Van Nostrand, Princeton, 1948).
[39] E-mail address for requests for the computer program: JWALDRON@COMPAPP.DCU.IE


[^0]:    ${ }^{*}$ Permanent address: Theoretical and Computational Physics Research Division, Department of Applied Mathematics and Theoretical Physics, The Queen's University of Belfast, Belfast BT7 1NN, Northern Ireland.

