Journal of Magnetism and Magnetic Materials 127 (1993) L254–L260
North-Holland

**Letter to the Editor**

# Exact analytic formulae for the correlation times for single domain ferromagnetic particles

W.T. Coffey [a,*], D.S.F. Crothers [b,1], Yu.P. Kalmykov [c], E.S. Massawe [a], J.T. Waldron [d]

[a] *Department of Microelectronics & Electrical Engineering, Trinity College, Dublin 2, Ireland*
[b] *Harvard Smithsonian Center for Astrophysics, 60 Garden Street, Cambridge MA 02138, USA*
[c] *Institute of Radio Engineering & Electronics, Russian Academy of Sciences, Vvedenskii Sq.1, Fryazino, Moscow Region 141120, Russia*
[d] *School of Computer Applications, Dublin City University, Dublin 9, Ireland*

Received 28 June 1993

Exact solutions for the longitudinal relaxation time $T_{\parallel}$ and the complex susceptibility $\chi_{\parallel}(\omega)$ of a thermally agitated single domain ferromagnetic particle are presented for the simple uniaxial (Maier–Saupe) potential of the crystalline anisotropy considered by Brown [Phys. Rev. 130 (1963) 1677].

## 1. 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 first described by Néel [1]. He obtained an expression for the relaxation time associated with the transition between two stable orientational states by assuming that the energy barrier between stable states is so large compared to $kT$ that the directions of the magnetic moment of the particle are concentrated at the energy minima [2].

Brown [3] improved this treatment, accounting for a continuous distribution of orientations, by constructing the Smoluchowski equation for the distribution of magnetic moment orientations on the unit sphere. He thus obtained a formula using the Kramers transition state theory [4] for the relaxation time in the high barrier limit for the simple uniaxial potential of the crystalline anisotropy

$$V(\vartheta) = K \sin^2\vartheta$$

where $\vartheta$ is the polar angle, $K$ is the anisotropy energy per unit volume.

Subsequently the solution of the Smoluchowski equation was investigated numerically by Aharoni [2] who found close agreement with Brown's results in the limit of high potential barriers. Very recently interest has revived in finding a solution which is valid for all potential barriers as suggested in [5,6]. It is the purpose of this letter to report how an exact solution for the correlation time may be found by reducing [7] the Smoluchowski equation to a three term recurrence relation. In considering this

[1] 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.
* Corresponding author.

0304-8853/93/$06.00 © 1993 – Elsevier Science Publishers B.V. All rights reserved

procedure it is useful to treat rotation in two dimensions as well as the three dimensional problem of Brown as the calculations for rotation in two dimensions are considerably easier.

## 2. Rotation in two dimensions

The Smoluchowski equation for a fine ferromagnetic particle with magnetization vector $\boldsymbol{M}$ confined to rotate in a plane under the influence of an internal uniaxial anisotropy potential and an external steady magnetic field $\boldsymbol{H}$ applied along the anisotropy axis which lies in the same plane is

$$\tau\frac{\partial W}{\partial t}=\frac{\partial}{\partial\phi}\left(W\frac{\nu}{kT}\frac{\partial V}{\partial\phi}\right)+\frac{\partial^2 W}{\partial\phi^2} \tag{1}$$

where $W(\phi, t)$ is the probability density of orientations of $\boldsymbol{M}$ on a circle of radius $M_s$, the plane polar angle $\phi$ specifies the orientation of $\boldsymbol{M}$ and

$$\frac{\nu V(\phi)}{kT}=2\sigma\sin^2\phi-\xi\cos\phi,\qquad \sigma=\frac{K\nu}{2kT},\quad \xi=\frac{\nu HM_s}{kT}. \tag{2}$$

$\sigma$ is so defined for the sake of notational convenience, $\nu$ is the volume of the particle, $M_s$ is the saturation magnetization. The characteristic time $\tau$ is defined as [3]

$$\tau=\frac{\nu}{kT}\left[\frac{1/\gamma^2+\eta^2M_s^2}{2\eta}\right].$$

$\gamma$ is the gyromagnetic ratio and $\eta$ is a phenomenological damping constant.

In order to study the relaxation behaviour we suppose that $\boldsymbol{H}$ is switched off at $t=0$. Let us now suppose that

$$W(\phi, t)=\sum_{p=-\infty}^{\infty}a_p(t)\,\mathrm{e}^{\mathrm{i}p\phi}. \tag{3}$$

Whence $\langle\cos(2p+1)\phi\rangle=f_{2p+1}(t)=a_{2p+1}(t)/a_0$ satisfies

$$\dot f_{2p+1}(t)+\frac{(2p+1)^2}{\tau}f_{2p+1}(t)-\frac{\sigma(2p+1)}{\tau}\left[f_{2p-1}(t)-f_{2p+3}(t)\right], \tag{4}$$

with $f_{-p}(t)=f_p(t)$, $f_0(t)=1$.

We wish to determine the relaxation behaviour of the decay functions $f_1(t)$ in the linear response approximation where $\xi\ll 1$. By taking the Laplace transform of Eq. (4) and using the methods described by Coffey et al. [8] one may show that

$$\tilde f_1(s)=\frac{\tau}{s\tau+(1-\sigma)+\sigma\tilde S_3(s)}\left[f_1(0)+\sum_{p=1}^{\infty}\frac{(-1)^p}{(2p+1)}f_{2p+1}(0)\prod_{k=1}^{p}\tilde S_{2k+1}(s)\right] \tag{5}$$

where

$$\tilde S_p(s)=\frac{\sigma p}{\tau}\Big/\left\{s+\frac{p^2}{\tau}+\frac{\sigma p}{\tau}\tilde S_{p+2}(s)\right\}. \tag{6}$$

which is an infinite continued fraction.

Eq. (5) allows us to determine the complex susceptibility $\chi_{\|}(\omega)$ using linear response theory. The most important quantity in the present application is however the correlation time which may be obtained from Eq. (5) as follows:

On noting that

$$T_{\|} = \lim_{s\to 0} \int_0^\infty C_1(t)\, \mathrm{e}^{-st}\, \mathrm{d}t = \lim_{s\to 0} \tilde{C}_1(s) = \tilde{C}_1(0) \tag{7}$$

and that for the linear response the autocorrelation function is $C_1(t) = f_1(t)/f_1(0)$, we see that $T_{\|}$ may be determined from Eq. (5) by setting $s = 0$. That is

$$T_{\|} = \frac{\tilde{f}_1(0)}{f_1(0)}. \tag{8}$$

Thus on inserting the initial conditions explicitly, we have

$$T_{\|} = \frac{\tau}{(1-\sigma)+\sigma\tilde{S}_3(0)}\left[1 + \sum_{p=1}^{\infty} \frac{(-1)^p}{(2p+1)} \left\{ \frac{I_{p+1}(\sigma)+I_p(\sigma)}{I_1(\sigma)+I_0(\sigma)} \right\} \prod_{k=1}^{p} S_{2k+1}(0)\right]. \tag{9}$$

By inspection of Eq. (6),

$$\tilde{S}_{2k+1}(0) = I_{k+1/2}(\sigma)/I_{k-1/2}(\sigma) \tag{10}$$

where the $I_k$ are the modified Bessel functions of the first kind. Thus

$$T_{\|} = \frac{\tau}{(1-\sigma)+\sigma I_{3/2}(\sigma)/I_{1/2}(\sigma)}\left[1 + \frac{1}{f_1(0) I_{1/2}(\sigma)} \sum_{p=1}^{\infty} \frac{(-1)^p}{(2p+1)} f_{2p+1}(0) I_{p+1/2}(\sigma)\right] \tag{11}$$

where

$$\frac{f_{2p+1}(0)}{f_1(0)} = \frac{I_{p+1}(\sigma)+I_p(\sigma)}{I_1(\sigma)+I_0(\sigma)}. \tag{12}$$

Thus utilizing the properties of the $I_{k+1/2}(\sigma)$,

$$T_{\|} = \frac{\tau}{2\sigma}(\mathrm{e}^{2\sigma}-1)\left[1 + \sum_{p=1}^{\infty} \frac{(-1)^p}{(2p+1)} \frac{f_{2p+1}(0)}{f_1(0)} \frac{I_{p+1/2}(\sigma)}{I_{1/2}(\sigma)}\right]. \tag{13}$$

This is an exact analytic formula for $T_{\|}$ which is valid for all $\sigma$. If $\sigma$ is very large, one easily finds using the asymptotic formulae for the $I_p$ that [8]

$$T_{\|} \cong \left(\frac{\tau}{2\sigma}\, \mathrm{e}^{2\sigma}\right) \sum_{p=0}^{\infty} \frac{(-1)^p}{(2p+1)} = \frac{\tau}{2\sigma}\, \mathrm{e}^{2\sigma}\beta(1) = \left(\frac{\tau}{2\sigma}\, \mathrm{e}^{2\sigma}\right)\frac{\pi}{4} \tag{14}$$

by the properties of the Riemann Zeta function [8]. This is the result of Lauritzen and Zwanzig [10] obtained by perturbation theory in the context of dielectric relaxation [9].

Eq. (13) agrees completely with the results of numerical solutions for $T_{\|}$ for all values of the barrier height $2\sigma$.

### 3. Rotation in three dimensions

This is the original problem considered by Brown [3]. The appropriate Smoluchowski equation is

$$2\tau\frac{\partial W}{\partial t}=\frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left[\sin\vartheta\left(\frac{\partial W}{\partial\vartheta}+\frac{\nu}{kT}\frac{\partial V}{\partial\vartheta}W\right)\right] \tag{15}$$

where

$$\frac{\nu V}{kT}=\sigma\sin^2\vartheta-\xi\cos\vartheta \tag{16}$$

and this time the barrier height parameter $\sigma=K\nu/kT$. $W(\vartheta, t)$ is the probability density of orientations of $\boldsymbol{M}$ on a sphere of radius $M_s$ and the orientation of $\boldsymbol{M}$ is specified by the spherical polar coordinates $\vartheta$ and $\varphi$. We assume that the after-effect solution of Eq. (15) is of the form

$$W(\vartheta, t)=\sum_{n=0}^{n=\infty}a_n(t)P_n(\cos\vartheta) \tag{17}$$

where $P_n$ is the Legendre polynomial of order $n$, which on using the orthogonality properties of the $P_n$ leads to (having switched off the steady field $\boldsymbol{H}$ at time $t=0$)

$$\dot{f}_{2n+1}(t)=\frac{(2n+1)(n+1)}{\tau}\left[\frac{2\sigma}{(4n+1)(4n+5)}-1\right]f_{2n+1}(t)$$
$$+\frac{4\sigma n(2n+1)}{(4n+3)\tau}\frac{(n+1)}{(4n+1)}f_{2n-1}(t)-\frac{2\sigma(2n+1)(2n+3)(n+1)}{(4n+3)(4n+5)\tau}f_{2n+3}(t) \tag{18}$$

with $f_0(t)=1$ where $f_{2n+1}(t)=\langle P_{2n+1}(x)\rangle=(a_{2n+1}(t)/a_0)/(4n+3)$. Thus

$$\tilde{f}_1(s)=\frac{\tau}{\tau s+1-\frac{2}{5}\sigma+\frac{2}{5}\sigma\tilde{S}_3(s)}\left[f_1(0)+\tfrac{4}{3}\sum_{n=1}^{\infty}(-1)^n\frac{f_{2n+1}(0)}{f_1(0)}\frac{(n+\frac{3}{4})(\frac{1}{2})_n}{\Gamma(n+2)}\prod_{k=1}^{n}\tilde{S}_{2k+1}(s)\right] \tag{19}$$

where $(\frac{1}{2})_n=\Gamma(n+\frac{1}{2})/\Gamma(\frac{1}{2})$ and the continued fraction

$$\tilde{S}_n(s)=\cfrac{\frac{2\sigma(n-1)}{4n^2-1}}{1+\frac{2\tau s}{n(n+1)}-\cfrac{2\sigma}{(2n-1)(2n+3)}+\cfrac{2\sigma(n+2)}{(2n+1)(2n+3)}\tilde{S}_{n+2}(s)}. \tag{20}$$

$\Gamma$ denotes the gamma function and the initial conditions satisfy

$$f_{2n+1}(0)=\frac{\xi\sigma^n\Gamma(n+\frac{3}{2})M(n+\frac{3}{2}, 2n+\frac{5}{2}, \sigma)}{2\Gamma(2n+\frac{5}{2})M(\frac{1}{2}, \frac{3}{2}, \sigma)} \tag{21}$$

where $M(a, b, z)$ is Kummer's function (the confluent hypergeometric function) [9]. The exact formula (Eq. (19)) allows one to calculate the longitudinal susceptibility since [11] ($s=\mathrm{i}\omega$)

$$\frac{\chi_\parallel(\omega)}{\chi_\parallel'(0)}=1-\mathrm{i}\omega\int_0^\infty \mathrm{e}^{-\mathrm{i}\omega t}C_1(t)\,\mathrm{d}t=1-\mathrm{i}\omega\frac{\tilde{f}_1(\omega)}{f_1(0)}. \tag{22}$$

Thus

$$\frac{\chi_{\parallel}(\omega)}{\chi_{\parallel}'(0)} = \frac{1}{i\omega\tau + 1 - \frac{2}{5}\sigma + \frac{2}{5}\sigma\tilde{S}_3(\omega)} \times\left[1 - \tfrac{2}{5}\sigma + \tfrac{2}{5}\sigma\tilde{S}_3(\omega) - i\omega\tau\tfrac{4}{3}\sum_{n=1}^{\infty}(-1)^n\frac{f_{2n+1}(0)}{f_1(0)}\frac{(n+\frac{3}{4})(\frac{1}{2})_n}{\Gamma(n+2)}\prod_{k-1}^{n}\tilde{S}_{2k+1}(\omega)\right]. \tag{23}$$

Eq. (19) with $s = 0$ yields

$$T_{\parallel} = \frac{\tilde{f}_1(0)}{f_1(0)} = \frac{\tau}{1 - \frac{2}{5}\sigma + \frac{2}{5}\sigma\tilde{S}_3(0)}\left[1 + \tfrac{4}{3}\sum_{n=1}^{\infty}(-1)^n\frac{f_{2n+1}(0)}{f_1(0)}\frac{(n+\frac{3}{4})(\frac{1}{2})_n}{\Gamma(n+2)}\prod_{k-1}^{n}\tilde{S}_{2k+1}(0)\right]. \tag{24}$$

Eq. (24) is an exact analytical formula which allows $T_{\parallel}$ to be calculated to any desired degree of accuracy by computing successive convergents of the continued fraction $\tilde{S}_{2k+1}(0)$. The $\tilde{S}_{2k+1}(0)$ are in terms of Kummer's functions

$$\tilde{S}_{2k+1}(0) = -\frac{4k\sigma}{(4k+1)(4k+3)}\frac{M\left(k+1, \frac{4k+5}{2}, \sigma\right)}{M\left(k, \frac{4k+1}{2}, \sigma\right)} \tag{25}$$

so that Eq. (24) with Eq. (21) becomes

$$\frac{T_{\parallel}}{\tau} = M(1, \tfrac{5}{2}, \sigma) + \frac{\frac{3}{2}}{M(\frac{3}{2}, \frac{5}{2}, \sigma)}\sum_{n=1}^{\infty}\frac{(-\sigma^2)^n(n+\frac{3}{4})\Gamma(n+\frac{3}{2})\Gamma(n+\frac{1}{2})}{(n+1)\left[\Gamma(2n+\frac{5}{2})\right]^2} \times M(n+\tfrac{3}{2}, 2n+\tfrac{5}{2}, \sigma)M(n+1, 2n+\tfrac{5}{2}, \sigma) \tag{26}$$

which is the *exact solution in terms of known functions for the longitudinal relaxation time* for the $K\sin^2\vartheta$ potential.

On using the asymptotic form of $M(a, b, z)$ as $|z| \to \infty$, namely [9]

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

Eq. (26) becomes in the high barrier limit

$$\frac{T_{\parallel}}{\tau} = e^{\sigma}\sigma^{-3/2}\sqrt{\pi}\sum_{n=0}^{\infty}\frac{(-1)^n}{(n+1)!}(1/2)_n(n+\tfrac{3}{4}). \tag{27}$$

Now by the properties of Gauss's hypergeometric function

$$F(a, b; c; z) = \sum_{n=0}^{\infty}\frac{(a)_n(b)_n}{(c)_n}\frac{z^n}{n!} \tag{28}$$

given in Ref. [9], the sum of the series in Eq. (27) is $\frac{1}{2}$, so that finally

$$\frac{T_{\parallel}}{\tau} \cong \frac{\sqrt{\pi}}{2}e^{\sigma}\sigma^{-3/2}, \tag{29}$$

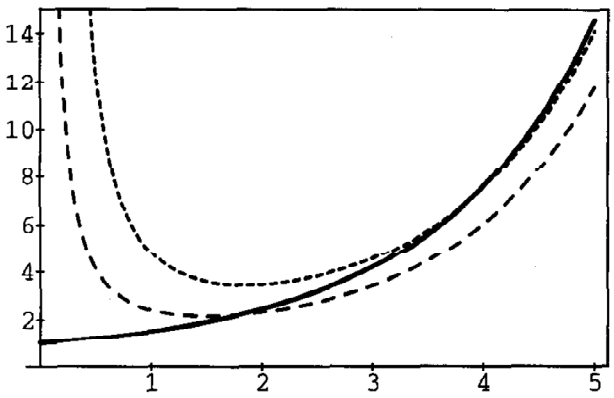



Fig. 1. Exact solution (Eq. (26)) for the longitudinal correlation time in the form $T_{\|}/\tau$ (solid line) compared with the asymptotic solution (Eq. (29)) (large dashed line) of Brown. The small dashed line is the solution rendered by the asymptotic formula (Eq. (30)).

which is the asymptotic formula of Brown [3]. In the opposite limit where $\sigma \to 0$, Eq. (26) reduces to

$$\frac{T_{\|}}{\tau} = 1$$

using the definition of $M(1, \frac{5}{2}, \sigma)$.

Table 1
Comparison of various asymptotic formulae for $T_{\|}/\tau$ with the exact solution (Eq. (26)). Eq. (30) is Brown's formula with $\sigma^{-1}$ correction, Eq. (29) Brown's formula without correction. It is apparent that Eq. (30) yields the closest approach to the exact solution for large $\sigma$.

| $\sigma$ | Eq. (26) | Eq. (30) | Eq. (29) |
|---|---|---|---|
| 1 | 1.5280 | 4.8180 | 2.4090 |
| 1.5 | 1.9254 | 3.6033 | 2.1620 |
| 2 | 2.4603 | 3.4728 | 2.3152 |
| 2.5 | 3.1899 | 3.8238 | 2.7313 |
| 3 | 4.1982 | 4.5676 | 3.4257 |
| 3.5 | 5.6091 | 5.7626 | 4.4820 |
| 4 | 7.6061 | 7.5604 | 6.0483 |
| 4.5 | 10.4630 | 10.2140 | 8.3570 |
| 5 | 14.5890 | 14.1170 | 11.7640 |
| 6 | 29.4300 | 28.3810 | 24.3270 |
| 10 | 691.0200 | 679.0200 | 617.2900 |
| 14 | 21986.0000 | 21799.0000 | 20346.0000 |
| 18 | $8.083500\times10^{5}$ | $8.04300\times10^{5}$ | $7.61970\times10^{5}$ |
| 22 | $3.229400\times10^{7}$ | $3.21880\times10^{7}$ | $3.07890\times10^{7}$ |
| 26 | $1.361900\times10^{9}$ | $1.35870\times10^{9}$ | $1.30840\times10^{9}$ |
| 30 | $5.96620\times10^{10}$ | $5.95580\times10^{10}$ | $5.76360\times10^{10}$ |
| 34 | $2.68850\times10^{12}$ | $2.68490\times10^{12}$ | $2.60820\times10^{12}$ |
| 38 | $1.23820\times10^{14}$ | $1.23690\times10^{14}$ | $1.20520\times10^{14}$ |
| 42 | $5.80280\times10^{15}$ | $5.79770\times10^{15}$ | $5.66290\times10^{15}$ |
| 46 | $2.75810\times10^{17}$ | $2.75610\times10^{17}$ | $2.69750\times10^{17}$ |
| 50 | $1.32640\times10^{19}$ | $1.32560\times10^{19}$ | $1.29960\times10^{19}$ |
| 54 | $6.44250\times10^{20}$ | $6.43910\times10^{20}$ | $6.32200\times10^{20}$ |
| 58 | $3.15580\times10^{22}$ | $3.15430\times10^{22}$ | $3.10090\times10^{22}$ |

The behaviour of the exact solution is shown in Fig. 1 and Table 1. In particular in Table 1 the exact solution is compared with Brown's formula (Eq. (29)) and the equations

$$\frac{T_{\|}}{\tau} \cong \frac{\sqrt{\pi}}{2}\, e^{\sigma}\sigma^{-3/2}\left(1+\frac{1}{\sigma}\right) \tag{30}$$

obtained by writing the exact solution in integral form and applying the method of steepest descents. It is apparent that the asymptotic correction yields a closer approximation to the exact solution than Brown's asymptotic formula in the high barrier limit. A table of the exact solution for all values of $\sigma$ is available from the authors. In order to ensure convergence to five significant digits for values of $\sigma$ up to 60 it is necessary to take the first 16 terms in the infinite series in Eq. (26).

## References

[1] L. Néel, Ann. Géophys. 5 (1949) 99.
[2] A. Aharoni, Phys. Rev. 135 (1964) A447.
[3] W.F. Brown Jr., Phys. Rev. 130 (1963) 1677.
[4] H.A. Kramers, Physica 7 (1940) 284.
[5] L. Bessais, L. Ben-Jaffel and J.L. Dormann, Phys. Rev. B 45 (1992) 7805.
[6] A. Aharoni, Phys. Rev. B 46 (1992) 534.
[7] W.T. Coffey, P.J. Cregg and Yu.P. Kalmykov, On the Theory of Debye and Néel Relaxation of Single Domain Ferromagnetic Particles, Adv. Chem. Phys., Vol. 83 (Wiley Interscience, New York, 1993) p. 263.
[8] W.T. Coffey, Yu.P. Kalmykov and E.S. Massawe, Phys. Rev. E, In press.
[9] M. Abramowitz and I. Stegun, Handbook of Mathematical Functions (Dover, New York, 1964).
[10] J.I. Lauritzen Jr. and R. Zwanzig, Adv. Molec. Rel. Interact. Proc. 5 (1973) 339.
[11] B.K.P. Scaife, Principles of Dielectrics (Oxford University, Oxford, 1989).