Relaxation time of fine magnetic particles in uniaxial symmetry

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Models for fine magnetic particles are shortly reviewed. A method for the solution of the partial differential equation occurring in Brown's model is presented, which in the uniaxial case permits us to calculate numerical solutions for a large range of α values. An approximate formula for the relaxation time is given. The expression obtained is valid for $0 \leq \alpha \leq 60$, which corresponds to all the physical cases (including geological scale). This formula is used to fit experimental results and good agreement is obtained. In addition, the question of the validity of Brown's treatment when the magnetization is not uniform, the meaning of the dissipation constant, and quantum effects are discussed.

I. INTRODUCTION

When a ferromagnetic particle is smaller than a certain critical size, it constitutes a single-domain particle.[1] The direction of its magnetic moment $\mathbf{m}$ when no external field is applied is, at zero temperature, along an axis of easy magnetization corresponding to a minimum of the energy of magnetic anisotropy. The main effect of thermal agitation is a continuous fluctuation of the direction of the vector $\mathbf{m}$ along with a possible overturn of $\mathbf{m}$ from one minimum to another, by overcoming a barrier of energy E_B. Such a set of small particles can be described by one or several relaxation times τ_i necessary for the distribution of orientations of $\mathbf{m}$ to get close to thermal equilibrium.

When τ_i is smaller than the measurement time τ_m the particle is said to be in a superparamagnetic state.[2] A description of the properties of such particles and a model for the relaxation time have been given by Néel,[3] who considered magnetostriction and demagnetization fluctuations induced by vibrations. In Néel's theory,[3] it is assumed that the rotation of the magnetization takes place in unison, i.e., relative directions of the magnetic moments remain unchanged during the rotation.

Using the theory of stochastic processes, by considering a random walk of the magnetization direction analogous to the Brownian motion of a small particle in a liquid, Brown[4] derived a Fokker-Planck type equation for the probability density $W(t,\theta,\phi)$ of the magnetic-moment orientations, the eigenvalues of which are directly related to the above quoted relaxation times τ_i. Unfortunately, Brown[4] did not calculate the eigenvalues of his equation. He obtained an asymptotic formula for high energy barriers, and calculated up to second order in the energy the values for low energy barriers. Therefore, he could not give any reliable estimation for the range of validity of the common high energy-barrier approximation.

Afterwards, Aharoni,[5,6] Aharoni and Eisenstein,[7] and Eisenstein and Aharoni[8] tried to solve the differential equation previously derived by Brown.[4] They obtained numerical and asymptotic expressions for the relaxation time corresponding to small particles of axial or cubic anisotropy. However, the numerical series they used were of slow convergence and the asymptotic expressions not checked for high energy-barrier values.

Recently, Jones and Srivastava[9,10] proposed a theoretical model based upon a formalism developed by Anderson[11] which they applied to the Mössbauer spectra of superparamagnetic particles with uniaxial anisotropy by including in the calculation all possible values of the component of the magnetization along the easy direction axis. They showed that the Mössbauer line shape can be expressed directly in terms of the solution of a differential equation in the continuum limit. A rather arbitrary R factor is introduced to describe possible quantum effects. Moreover, this R factor is used as a parameter to obtain the Mössbauer line shape for a given system of particles. So it is not clear in their theoretical framework how to construct a proper R factor. It is clear that the main feature of their theory is the possibility of treating the Mössbauer lines formation as a multilevel problem. However, a different stochastic description of the magnetization fluctuations (i.e., Anderson[11] instead of Brown[4]) did not lead to a sensitive improvement of their model when compared to Brown's[4] treatment.

More recently, Klik and Gunther,[12,13] derived the relaxation rate for the axially symmetric model previously studied by Brown,[4] following the asymptotic expansion method of Matkowski and Schuss.[14] These authors started with the classical Fokker-Planck equation in spherical coordinates, and argued that the obtained prefactor was unphysical as its $T^{-1/2}$ dependence was claimed to be

due to the lack of saddle points. After that, Klik and Gunther[12] applied the results of the formalism of Dygas, Matkowski, and Schuss[15] and obtained, in the asymptotic limit, the dissipation rate for systems of uniaxial and cubic symmetry. We shall discuss and comment upon these results in Sec. III.

Considering interparticle interactions, Dormann, Bessais, and Fiorani[16] presented phenomenological expressions based upon experimental results they obtained for some species of small particles. Among other results, they claimed, from the dynamical study they performed, the possibility of a phase transition at 0 K for these particles.

Note that an important difficulty that arises in the study of the superparamagnetic relaxation time[16,17] is the lack of a general law (Fulcher, generalized Arrhenius, power-law type, or others) which is able both to describe the relaxation phenomenon of small particles and to allow for a correct and precise reconstitution of the experimental results.

To refine the classical treatment of fine-particle relaxation, we present here a method for the solution of the Fokker-Planck equation derived by Brown[4] [his Eq. (2.10)]. The main advantage of our method when compared to the previously used procedures[4,5] is its ability to provide an analytical formulation of the solution in addition to the numerical results. This advantage arises from the cumulative effects of the great speed of convergence of the method, the matrix formulation of the problem, and, finally, the simplicity of the orthogonal functions used (sine and cosine contrary to the commonly used Legendre polynomials).

In Sec. II, we develop the method; in Sec. III we apply it to the very realistic and simple case of uniaxial particles, and an Arrhenius-type law is derived for the relaxation time. In Sec. IV comparison with experiments is presented, as well as a discussion concerning the validity of the present treatment when the magnetization is not uniform, the role of the dissipation constant, the determination of the preexponential factor τ_0, and, finally, the possible influence of quantum fluctuations.

II. THE METHOD

A. Brown model

Brown[4] considered a small single-domain particle with a uniform mode of rotation of the magnetic moment. The fluctuations of the magnetization vector in such a small particle due to the thermal agitation are important. Although the problem is difficult, it is possible to derive a solution if we consider the correlation times corresponding to the random thermal forces to be smaller than the response time of the system (which is the case of Brownian particles). In this case the random forces reduce to a purely random process with a "white" spectrum, and the Brown[4] hypothesis is valid. This simplification allows us to replace an integral equation (Chapman-Kolmogorov type) by a partial differential one (Fokker-Planck type). In his paper, Brown[4] derived the following equation:

$$\frac{\partial W(t,\theta,\phi)}{\partial t}=\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left[\sin\theta\left[h'E_\theta-g'\frac{1}{\sin\theta}E_\phi\right]W+k'W_\theta\right]+\frac{1}{\sin\theta}\frac{\partial}{\partial\phi}\left[\left[g'E_\theta+h'\frac{1}{\sin\theta}E_\phi\right]W_\theta+k'\frac{1}{\sin\theta}W_\phi\right], \tag{1}$$

where

$$g'=\frac{1/\gamma_0}{M_s(1/\gamma_0^2+\eta^2M_s^2)},$$

$$h'=\frac{\eta}{1/\gamma_0^2+\eta^2M_s^2}, \tag{2a}$$

$$k'=\frac{k_bTh'}{V},$$

and

$$E_\theta=\partial E/\partial\theta \quad \text{and} \quad E_\phi=\partial E/\partial\phi, \tag{2b}$$

$$W_\theta=\frac{\partial W}{\partial\theta} \quad \text{and} \quad W_\phi=\frac{\partial W}{\partial\phi}, \tag{2c}$$

where E is the energy density. Keeping in mind that the form of the solution to Eq. (1) does not depend upon the uniformity of the magnetization, we will use in our treatment the magnetic moment $\mathbf{m}$ ($\mathbf{m}$ per unit volume is noted "$\mathbf{M}$" in the Brown[4] paper) of the particle with the mean magnetization defined as $M=|\mathbf{m}|/V$, where V is the volume of the particle. This change with regards to Brown's notation is of importance and will be discussed in Sec. IV.

B. Solution of Brown's equation

Having used spherical coordinates, it is obvious that any physical quantity $Y(t,\theta,\phi)$, considered as a function of the angle ϕ, is periodic, having a period of at least 2π. Under very weak conditions (Dirichlet's conditions) it is possible to expand the function $Y(t,\theta,\phi)$ as a Fourier series in the variable ϕ as follows:

$$Y(t,\theta,\phi)=\sum_{m=-\infty}^{+\infty}\tilde{Y}_m(t,\theta)e^{jm\phi}, \tag{3}$$

where $0\le\theta\le\pi$ and $0\le\phi\le2\pi$, j being the purely imaginary number $\sqrt{-1}$, and $\tilde{Y}_m(t,\theta)$ are the Fourier coefficients defined as follows:

$$\tilde{Y}_m(t,\theta)=\frac{1}{2\pi}\int_0^{2\pi}Y(t,\theta,\phi)e^{-jm\phi}d\phi. \tag{4}$$

However, depending upon the parity of the index m appearing in Eq. (3), we can expand the Fourier coefficients $\tilde{Y}_m(t,\theta)$ as a sine or cosine series as follows:[18]

$$\tilde{Y}_m(t,\theta)=\sum_{n=0}^{+\infty}\bar{Y}_{n,m}(t)\cos(n\theta) \quad \text{if } m \text{ is even},$$
$$\tilde{Y}_m(t,\theta)=\sum_{n=0}^{+\infty}\bar{Y}_{n,m}(t)\sin(n\theta) \quad \text{if } m \text{ is odd}, \tag{5}$$

where $\bar{Y}_{n,m}(t)$ are the sine or cosine coefficients defined as follows:

$$\bar{Y}_{n,m}(t)=\frac{1}{\pi}\int_0^{\pi}\tilde{Y}_m(t,\theta)\times\begin{Bmatrix}\cos(n\theta) & \text{if } m \text{ is even}\\ \text{or}\\ \sin(n\theta) & \text{if } m \text{ is odd}\end{Bmatrix}d\theta \tag{6}$$
$$\bar{Y}_{0,2p}(t)=\frac{1}{2\pi}\int_0^{\pi}\tilde{Y}_{2p}(t,\theta)d\theta$$

Note finally that $\cos(n\theta)$ and $\sin(n\theta)$ can be considered, respectively, as the Chebyshev polynomials of the first and second kind. We can then use the above properties for the probability density $W(t,\theta,\phi)$ and develop it first as a Fourier series in the ϕ variable. In other words we can write

$$W(t,\theta,\phi)=\sum_{k=-\infty}^{+\infty}W_k(t,\theta)e^{jk.\phi}, \tag{7}$$

where $W_k(t,\theta)$ are the Fourier coefficients defined as follows:

$$W_k(t,\theta)=\frac{1}{2\pi}\int_0^{2\pi}W(t,\theta,\phi)e^{-jk.\phi}d\phi, \tag{8}$$

as already indicated by Eq. (4). We also develop the free energy E as a simple Fourier series, with the coefficients E_k.

Introducing these Fourier series in Eq. (1) and using their orthogonality properties, we obtain a differential equation for each k:

$$\frac{\partial W_k}{\partial t}=k'\left[-\frac{k^2}{\sin^2\theta}+\frac{\partial^2}{\partial\theta^2}+\cot\theta\frac{\partial}{\partial\theta}\right]W_k$$
$$+\sum_m\left[\left[h'\cot\theta+jm\frac{g'}{\sin\theta}\right]W_m+\frac{\partial}{\partial\theta}E_{k-m}+h'W_m\frac{\partial^2}{\partial\theta^2}E_{k-m}\right.$$
$$\left.-jg'\frac{k-m}{\sin\theta}\frac{\partial W_m}{\partial\theta}E_{k-m}-h'k(k-m)\frac{1}{\sin^2\theta}W_mE_{k-m}+h'\frac{\partial W_m}{\partial\theta}\frac{\partial E_{k-m}}{\partial\theta}\right]. \tag{9}$$

Now, the next step is to introduce the Chebyshev polynomial expansion [see Eq. (5)] for each Fourier coefficient $W_k(t,\theta)$ appearing in Eq. (9), which gives (see the Appendix)

$$\frac{\partial W_{s,k}(t)}{\partial t}=\sum_r W_{r,k}k'(-k^2I-rJ-r^2K)$$
$$+\sum_{\substack{m\\ \text{even}}}\sum_{r,q}W_{r,m}E_{q,k-m}[-h'qL-jmg'qL'-h'q^2M+jg'(k-m)rH-h'k(k-m)P+h'rqN]$$
$$+\sum_{\substack{m\\ \text{odd}}}\sum_{r,q}W_{r,m}E_{q,k-m}[h'qQ+jmg'qQ'-h'q^2R+jg'(k-m)rT-h'k(k-m)U+h'rqS], \tag{10}$$

where I, J, K, L, L', M, N, H, P, Q, Q', R, S, T, and U are well-defined functions both of the indices (r,q,s) and the parity of m (see the Appendix). It is easy to see that Eq. (10) can be written in a more compact form as follows:

$$\frac{dW_{s,k}(t)}{dt}=-k'\sum_r W_{r,k}(k^2G+rJ+r^2K)+\sum_{m,r,q}W_{r,m}E_{q,k-m}h'\{rq\tilde{N}-k(k-m)\tilde{P}-q^2\tilde{M}-q\tilde{L}$$
$$+jg'[-m\tilde{L}'+(k-m)r\tilde{H}]\}, \tag{11}$$

where $(\tilde{L},\tilde{L}',\tilde{M},\tilde{H},\tilde{P}$, and $\tilde{N})$ correspond to the same functions without a tilde for even values of m and are defined as follows for the odd values: $(\tilde{L}=-Q;\ \tilde{L}'=-Q';\ \tilde{M}=R;\ \tilde{H}=-T;\ \tilde{P}=U$; and $\tilde{N}=S)$.

Now, the previous results enable us to conclude that we have reduced the multidimensional Fokker-Planck equation to an infinite system of one dimension but coupled equations for the coefficients $W_{s,k}(t)$. The correlation between the coefficients $W_{s,k}$ via the right-hand part of Eq. (11) can be decoupled by considering a finite Fourier and Chebyshev sum instead of infinite series in Eqs. (4) and (5) (called the spectral approximation)

$$W^{(M,N)}(t,\theta,\phi)=\sum_{k=-M}^{M}W_k^{(N)}(t,\theta)e^{jk\phi}, \tag{12}$$

and

$$W_k(t,\theta)=\sum_{n=0}^{N}W_{n,k}(t)\times\begin{matrix}\cos(n\theta) & \text{if } k \text{ is even}\\ \sin(n\theta) & \text{if } k \text{ is odd}\end{matrix}, \tag{13}$$

where $W_k^{(N)}(t,\theta)$ and $W_{n,k}^{(M,N)}(t)$ are, respectively, the

coefficients $W_k(t,\theta)$ and $W_{n,k}(t)$ resulting from the truncated series. The convergence of the truncated series is assured by spectral analysis as shown by Canuto *et al.*[18] We therefore consider in all the following finite Fourier and Chebyshev series.

C. Matrix analysis

To obtain the relaxation times τ_i which are directly related to the eigenvalues of the Fokker-Planck equation, we consider the following matrix notation:

$$W^{(N,M)}=\begin{pmatrix} W_{0,-M} \\ \vdots \\ W_{n,-M} \\ \vdots \\ W_{0,k} \\ \vdots \\ W_{N,k} \\ \vdots \\ W_{0,M} \\ \vdots \\ W_{N,M} \end{pmatrix}, \tag{14}$$

where $W^{(N,M)}$ is a vector of Fourier and Chebyshev coefficients appearing in Eq. (11). Each element of this vector is defined as follows:

$$W_r^{(N,M)}=W_{s,k}^{(N,M)}, \tag{15}$$

with $r=s+(M+k)(N+1)$; $0\le s\le N$; $-M\le k\le M$.

Taking into account of the above notation, we can rewrite Eq. (11) in a more compact form

$$(d/dt)W^{(N,M)}=-A^{(N,M)}W^{(N,M)}. \tag{16}$$

It is clear that we now deal with a differential linear equation, where $A^{(N,M)}$ is a time independent matrix which elements are defined in the following manner:

$$A_{i,j}^{(N,M)}=A_{(s_i,k_i;s_j,k_j)}^{(N,M)} \tag{17a}$$

and

$$A_{(s_i,k_i;s_j,k_j)}^{(N,M)}=k'(k_i^2 I+s_j J+s_j^2 K)\delta_{k_i,k_j}+\sum_q E_{q,(k_i-k_j)}h'\{-s_j q\tilde{N}+k_i(k_i-k_j)\tilde{P}+q^2\tilde{M}+q\tilde{L}+jg'[k_j q\tilde{L}'-(k_i-k_j)s_i\tilde{H}]\}. \tag{17b}$$

The matrix being time independent, the solution of Eq. (16) reads

$$W^{(N,M)}(t)=e^{-tA^{(N,M)}}W^{(N,M)}(t=0). \tag{18}$$

In the following we shall show that the problem resides now in the calculation of $e^{-A^{(N,M)}}$ and that the relaxation times are simply equal to the inverse of the real parts of the eigenvalues of $A^{(N,M)}$. Therefore, it is sufficient to diagonalize this matrix to get both the relaxation times and the probability density via Eqs. (12) and (13).

We can generally write

$$A^{(N,M)}=P\Gamma P^{-1}, \tag{19}$$

where P is the nodal matrix associated with $A^{(N,M)}$, Γ is the diagonal (or Jordan) matrix of the eigenvalues, and P^{-1} is the inverse of P. Using the above decomposition [Eq. (19)], we can write

$$e^{-A^{(N,M)}}=Pe^{-\Gamma}P^{-1}, \tag{20}$$

where to simplify the notation we have dropped the indices (N,M) in the new decomposition of $A^{(N,M)}$. It follows that the computation of $e^{-A^{(N,M)}}$ is equivalent to the eigenvalue problem of $A^{(N,M)}$ and thus to the calculation of P, Γ, and P^{-1}.

To calculate the matrix Γ and thus all the eigenvalues, we used the formalism developed by Ben Jaffel and Vidal-Madjar,[19] which allows for the computation of the eigenvalues and the eigenvectors disregarding their multiplicities. We refer the readers to this paper for further details.

Having Γ, P, and P^{-1}, we can easily write,

$$e^{-tA^{(N,M)}}=Pe^{-t\Gamma}P^{-1}, \tag{21}$$

where $\Gamma_{i,j}=p_i\delta_{i,j}$.

We can now deduce the solution for the density of probability $W(t,\theta,\phi)$ using Eqs. (12) and (13)

$$W(t,\theta,\phi)=\sum_{n=0}^{N}\sum_{m=-M}^{M}W_{n,m}^{(N,M)}(t)e^{jm\phi}T_{n,m}(\theta), \tag{22a}$$

where, depending on the parity of m, $T_{n,m}$ represent the sine or cosine functions or, respectively, the Chebyshev polynomials of the first and second kind, and where

$$W_{n_i,m_i}^{(N,M)}(t)=W_i^{(N,M)}(t)=\sum_{k,l}P_{ik}e^{-p_k t}P_{kl}^{-1}W_l^{(N,M)}(t=0). \tag{22b}$$

Using Eqs. (21) and (22), we can show that $W(t,\theta,\phi)$ can be written in the following form:

$$W(t,\theta,\phi)=\sum_k e^{-\mathrm{Re}(p_k)t}\left[\sum_{il} T_{n_i}(\theta)[e^{-j\,\mathrm{Im}(p_k)t}P_{ik}P_{kl}^{-1}e^{jm_i\phi}W_l^{(N,M)}(t=0)+e^{j\,\mathrm{Im}(p_k)t}\bar{P}_{ik}\bar{P}_{kl}^{-1}e^{-jm_i\phi}\bar{W}_l^{(N,M)}(t=0)]\right], \tag{23a}$$

where to each index $s=i,k,l\ldots$, corresponds m_s,n_s in a way that s verifies

$$s=n_s+(N+1)(m_s-1) \tag{23b}$$

and where Re and Im mean real and imaginary part. Defining

$$F_k(\theta,\phi)=\sum_{il} T_{n_i,m_i}(\theta)[e^{-j\,\mathrm{Im}(p_k)t}P_{ik}P_{kl}^{-1}e^{jm_i\phi}W_l^{(N,M)}(t=0)+e^{j\,\mathrm{Im}(p_k)t}\bar{P}_{ik}\bar{P}_{kl}^{-1}e^{-jm_i\phi}\bar{W}_i^{(N,M)}(t=0)], \tag{24}$$

and $\tilde{p}_k=\mathrm{Re}(p_k)$, we can write using Eq. (23):

$$W(t,\theta,\phi)=\sum_k e^{-\tilde{p}_k t}F_k(\theta,\phi)\ . \tag{25}$$

Therefore we can deduce that the $F_k(\theta,\phi)$ are the eigenfunctions corresponding to the Fokker-Planck equation [see Brown,[4] Eq. (4.3)] and that the $\tilde{p}_k$ are the corresponding eigenvalues.

The relaxation times τ_k are simply defined as

$$\tau_k=1/\tilde{p}_k\ . \tag{26}$$

At this point, it is important to recall that using the Fourier and Chebyshev series, we have reduced the multidimensional Fokker-Planck equation to a simple differential equation. Using spectral methods and matrix algebra, we have deduced the corresponding eigenvalues and eigenfunctions. This only requires the diagonalization of a relatively low dimensional matrix (~ 80). Note that we have considered the most general form for the free energy $E(\theta,\phi)$.

III. APPLICATION TO UNIAXIAL SYMMETRY PARTICLES

The anisotropy energy corresponding to a superparamagnetic particle state is very complex, and no self-consistent theory nor experimental data are available to unambiguously define its real form. Furthermore, this anisotropy depends strongly on the nature (topology, constitution, structure, . . .) of the particles.

By symmetry considerations, it is possible[20] to construct analytical expressions for the anisotropy energy as a function of some unknown (to be provided or to be deduced from laboratory experiments) anisotropy coefficients. However, considering the complexity of the phenomena, there is no solid basis for such a formalism to hold for real samples of superparamagnetic particles.

Nevertheless, one way to overcome this difficulty is still to consider the symmetry derived formulas as a starting model to get some insight on the real samples through laboratory studies: This may help to get some experimental evidence on the departure of these simple models from the physics of the problem. It follows that there is *a priori* no reason to favor a specific symmetry, among others, the criteria in any particular choice generally are based on computational simplifications.

At this level, it is important to clarify some ambiguous points relevant to the uniaxial symmetry, mostly criticized by Klik and Gunther.[12] These authors[12,13] strongly claimed that the uniaxial symmetry was unphysical and that all results derived on this basis were completely faulty. We do not agree with these conclusions. Furthermore, we consider it is somewhat difficult to make such strong statements from an asymptotic solution to the problem.

Klik and Gunther draw their conclusions from two apparent difficulties of the uniaxial symmetry: the lack of a saddle point, and the so-called mathematical anomaly of the spherical coordinate system.

It is true that a completely uniaxial system (becomes a one-dimensional model) has no saddle point. However, we do not think that this lack leads directly to the $T^{-1/2}$ dependence of the relaxation rate prefactor, nor does the mathematical anomaly of the coordinate system. Moreover, we believe it is the asymptotic behavior associated with the mathematical method used by some authors[4,6,9] to solve the Fokker-Planck equation that leads to this dependence. In this paper, we clearly show (see the Appendix), that spectra methods avoid the pseudosingularities the spherical coordinates system introduces, and thus the mathematical anomaly problem raised by Klik and Gunther does not exist in our case.

Basically, we consider the uniaxial symmetry as a starting model that can be refined afterwards. In this particular case (the simplest one), the anisotropy energy $E(\theta,\phi)$, together with the density of probability $W(t,\theta,\phi)$ are ϕ (azimuthal angle) independent.

Noting that $E(\theta)$ can be written as

$$E(\theta)=K_t\sin^2(\theta)\ , \tag{27}$$

and that the matrix $\tilde{A}^{(N,M)}$ described in Sec. II is now just a function of N, we can deduce from Eq. (17)

$$\tilde{A}_{i,j}^N=k'(s_jJ+s_j^2K)+\sum_q E_q h'(-s_jq\tilde{N}+q^2\tilde{M}+q\tilde{L})\ . \tag{28}$$

The related eigenvalue problem thus becomes

$$\tilde{A}^N X=pX\ , \tag{29}$$

where X is an eigenvector of $\tilde{A}^N$ and p is the corresponding eigenvalue (which represents the inverse of the relaxation time).

Defining

$$\lambda=\frac{pV}{k_bTh'} \quad \text{and} \quad h'=\frac{\eta}{1/\gamma_0^2+\eta^2m^2/V^2} , \tag{30}$$

and

$$\alpha=K_tV/k_bT , \tag{31}$$

we rewrite Eq. (28) as follows:

$$\hat{A}^N X=\lambda X , \tag{32}$$

where $\hat{A}^N$ is now defined from Eqs. (28) and (29) as

$$\hat{A}_{ij}^N=\frac{V}{k_bTh'}\tilde{A}_{ij}^N . \tag{33}$$

We thus deal with the diagonalization problem of the real matrix $\hat{A}^N$ for increasing value of the dimension N until the stability of the eigenvalues and the eigenvectors is reached.

IV. RESULTS AND DISCUSSION

The smallest eigenvalue λ_1 seems of particular interest for superparamagnetic particles study. It corresponds to the dominant relaxation time. We have plotted in Fig. 1 the variation of λ_1 vs $\alpha=K_tV/k_bT$. The exact numerical calculation of this eigenvalue was performed for a wide domain of variation of α contrary to the previous publications.[5,6]

From the calculations we carried out, it appears that the convergence process is very rapid, except for very high values of α, where it decreases slightly. Then, we have obtained all the eigenvalues with a precision better than 10^{-5} and for matrix dimension no higher than 80.

Another advantage of the present method is the possibility of deriving a very precise analytical approximation for the eigenvalues and especially for the most important of them, λ_1. This possibility arises both from the successive analytical approximations we obtain for small matrix dimension, and the asymptotic behavior ($\alpha \gg 1$) we deduce from numerical calculations.

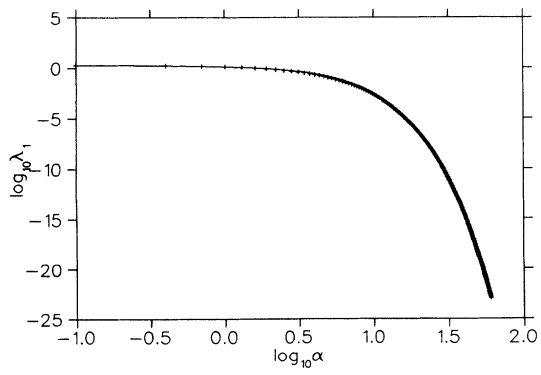

FIG. 1. First positive eigenvalue λ_1 of the Fokker-Planck equation, which is inversely proportional to the relaxation time τ, plotted as a function of the reduced energy barrier $\alpha=K_tV/k_bT$. The full curve corresponds to the exact numerical calculation and the (+) correspond to the analytical formulation.

Considering λ_1, we have derived the following analytical expression:

$$\lambda_1=2(1+\alpha/4)^{5/2}\exp(-\alpha) . \tag{34}$$

As shown in Fig. 1, the proposed analytical solution reproduces quite perfectly the numerical solution. The accuracy is better than 1% for $0\leq\alpha\leq60$. The upper limit corresponds to a relaxation time higher than one billion years. That means that this analytical expression is valid for geological scales. We therefore deduce from Eqs. (26) and (29) that the corresponding relaxation time τ_1 is given by

$$\tau_1=\frac{V}{k_bTh'}\frac{1}{2(1+\alpha/4)^{5/2}}\exp(\alpha) , \tag{35a}$$

which can be written in an Arrhenius-type form:

$$\tau_1=\tau_0e^{\alpha} , \tag{35b}$$

where

$$\tau_0=\frac{V}{k_bTh'}\frac{1}{2(1+\alpha/4)^{5/2}} . \tag{35c}$$

It is interesting to see here that our dissipation rate expression has a $T^{-3/2}$ temperature dependence in the asymptotic limit of high energy barrier, contrary to the $T^{-1/2}$ dependence previously deduced.[4,6,10,12] Contrary to Klik and Gunther's[12] claim, our result is, at least, an indication that the $T^{-1/2}$ dependence is not a direct consequence of the lack of saddle points.

Now, in order to compare the above formula for τ_1 with the experimental results, it is important to state precisely the τ_0 value. This leads us to the question of the meaning of the parameters included in h', particularly η. We can remark that the modified Gilbert's equation of Brown[4] mix spin parameters (η,γ_0) with particle terms [the free energy of the particles, the random field $\mathbf{h}(t)$]. This difficulty was overcome by Brown by supposing the uniformity of the particle magnetization, i.e., that each spin magnetic moment sees the same interactions and then has the same value, direction, the same γ_0 and η parameters. Unfortunately, the particle magnetization is never uniform due to surface effects. The common description for a ferromagnetic (or a ferrimagnetic) particle leads to the existence of a hard core with normal spin arrangement and a surface part for which the noncollinearity of the spin is generally concluded[21] and for which the value of the spin magnetic moment can be different from the bulk value.

Nevertheless, the validity of Brown's treatment can be maintained by considering the magnetic moment of the particle ($\mathbf{M}$ is replaced by $\mathbf{m}/V$ in Brown's notation, where $\mathbf{m}$ and V are respectively the magnetic moment and the volume of the particle). In this case, the parameters γ_0 and η are relative to the particle and correspond

to a kind of mean value of spin parameters. It is probable that the γ_0 value is very nearly the single spin value because experimental values deduced from different bulk measurements are very close. On the other hand, η values are probably much larger due particularly to the defects present in small particles and the influence of the surface which causes noncollinearity of spins.

This nonuniformity of the magnetization makes questionable the Brown[4] hypothesis of a uniform mode of rotation, in other words, the rigid coupling of the spins. The fact that exchange interactions inside the particle are much larger than other interactions leads to this mode probably occurring for spherical particles and to a good approximation for large particles, but it seems that some spin rearrangements occur during the rotation for very small ellipsoidal particles. In our opinion, that does not change the validity of Brown's treatment, at least in first approximation, but in this case the anisotropy constant which determines the energy barrier probably deviates from the bulk magnetocrystalline anisotropy constant.[22]

Now we can discuss the η values. In his paper Brown[4] has supposed $\eta=1/\gamma_0 M_s$ which represents the maximum possible value of h' with respect to η. This expression is questionable though the proportionality to $1/M_s$, and the dimensionality seem correct.[23] For a bulk sample, it is easy to determine the η value from the classical formula giving the resonance line width[23,24] ΔH. For example, for bulk iron $\Delta H \simeq 30$ Oe for $\nu=9$GHz at 300 K (Ref. 23) and the deduced η value is equal to $\simeq 3.7\times 10^{-10}/M_s$ to compare to $5\times 10^{-8}/M_s$ deduced from Brown's suggestion. In fact, the η value for bulk iron is lower than the value cited above because ΔH is dominated by the exchange-conductivity mechanism. A better expression for η will be therefore $\eta=aV/\gamma_0|\mathbf{m}|$ in agreement with the dependence of the Brown treatment to $\mathbf{m}$, a being a dimensionless parameter. In this case the h' factor can be expressed as

$$\frac{1}{h'}=\frac{|\mathbf{m}|}{\gamma_0}V\frac{1+a^2}{a} . \tag{36}$$

The parameter a could be determined from τ experimental values, but that is difficult because of the number of unknown parameters and their possible variation with temperature. For example in the case of magnetically interacting particles, if the a.c. susceptibility results are considered, one obtains a straight line in the classical plot $\log_{10}\nu=f(1/T_B)$, where ν is the frequency and T_B is the blocking temperature, with an intercept with the $\log_{10}\nu$ axis corresponding to $\tau_0\simeq 10^{-18}$ s.[16] This is due to the effect of magnetic dipolar interactions between particles through an additional term to τ_0[16] which complicates seriously the determination of h'. On the other hand, for small particles, ΔH values seem much larger than for bulk samples[23] which lead to a values with an order of magnitude of one. This estimate would be confirmed by an experimental determination[16] of τ_0, but the available experimental data is incomplete to estimate its order of magnitude. A consequence of this relation [Eq. (36)] is evident for particles made of antiferromagnetic materials. For these particles, disorder occurs as it does in the particle core (except for large particles). Experiments indicate that $|\mathbf{m}|$ proportional to n^p with $\frac{1}{3}\leq p\leq\frac{1}{2}$, depends upon the particle size, n being the number of spins.[25] From the above discussion, there are no special reasons which prevent Brown's model application for this type of particle as long as we use the actual value of $|\mathbf{m}|$ and the modified anisotropy constant. In this case, the value of the preexponential factor τ_0 will be lowered with regard to ferromagnetic particles as it is proportional to $|\mathbf{m}|/V$. We can remark that the antiferromagnetic particle terms are not adequate, as the magnetic state is indeed very far from a pure antiferromagnetic state.

From formula (35), it is clear that a correct estimate of τ_0 is necessary to determine the parameters inside the exponential and their eventual variation with temperature. On the other hand, this evaluation and the verification (or not) of the τ_0 expression is surely easier when the exponential has a value near 1, i.e., when α is small. Therefore it is interesting to discuss the τ_1 limit values at high temperature.

The first question concerns the thermal variation of the dissipation constant η. ΔH measurements versus temperature have shown that ΔH remains constant until around $0.8T_c$, then exhibits a sharp increase with temperature and reaches a constant value[23,26] above T_c the Curie temperature. For the $1/h'$ expression (36), we can consider that $\eta=a(T)V/\gamma_0|\mathbf{m}(0)|$, where $|\mathbf{m}(0)|/V$ is the mean magnetization at zero temperature. In this case,

$$\frac{1}{h'}=\frac{|\mathbf{m}(0)|}{\gamma_0 V}\frac{1+a(T)^2|\mathbf{m}(T)|^2/|\mathbf{m}(0)|^2}{a(T)} . \tag{37}$$

From this formula we can infer that for $a\leq 1$ (the more probable case), $1/h'$ is roughly independent of temperature with $1/h'\simeq|\mathbf{m}(0)|/\gamma_0 V a(T)$. On the contrary, for $a\geq 1$, $1/h'$ roughly decreases between $a(0)|\mathbf{m}(0)|/\gamma_0 V$ (low temperature) and $|\mathbf{m}(0)|/\gamma_0 V a(T)$ $(T=T_c)$. We can conclude that $1/h'$ remains finite when $1/T\rightarrow 1/T_c$. Therefore, from Eq. (35), $\tau_1\rightarrow\tau_0\rightarrow 0$ when $1/T\rightarrow 0$. Indeed the limit $1/T\rightarrow 0$ has no real physical meaning as in this case the magnetic state of the particle becomes paramagnetic, with a spin-relaxation time[10] around 10^{-14} s.

However, it is important to determine the relaxation time in this limit in order to specify its asymptotic behavior. Unfortunately, one of Brown's hypotheses, i.e., the unison rotation mode for the spins, becomes inappropriate in the paramagnetic state. Therefore it is not possible to derive any relaxation time valid for this latter state from this model. In addition, it is not definite that Brown's treatment is valid for weak values of α. Formally, it is always possible to derive τ_1, but the first eigenvector which describes the equilibrium state indicates that the probability density of the particle magnetic moment is almost equal over the unit sphere. In this case, it is difficult to keep the picture of particle magnetic moments relaxing between two preferred directions.

The asymptotic behavior of $\tau_0\rightarrow 0$ arises from the existence in our formulae of the ratio V/kT, also occurring

in the low energy approximation of Brown.[4] (The high energy approximation[4,5] is not valid when α is small.) It is important to remark that the factor V/kT appearing in Eqs. (35) arises from the semiclassical description of the particle magnetization of Brown[4] through a Langevin equation, which implies that the relaxation time is entirely independent of the microscopic origins of the relaxation. On the contrary, Jones and Srivastava[9] derived an expression with a "free" parameter R, which represents the strength of the random field (R should contain information on the microscopic origin of the relaxation), but their expression is not valid when $1/T \rightarrow 0$. However, and contrary to all theoretical considerations, it appears from experimental results,[16] that τ_1 approaches a finite and nonzero value of τ_0 as $1/T \rightarrow 0$.

A way to raise the difficulty could be the addition to τ_0 of some constant value around 10^{-14} s (relaxation time in paramagnetic state). Another way consists of determining an approximative formula, valid for α not too small, which extrapolated for $1/T \rightarrow 0$ gives a nonzero value for τ_0. For this purpose, we can write

$$\tau_1 = \frac{1}{h'} \frac{1}{K_t} g(\alpha) , \tag{38}$$

and approximate the $g(\alpha)$ function by $c_1 e^{\delta_1 \alpha}$, with $c_1 \simeq 0.3$ and $\delta_1 \simeq 0.96$, valid for a large α interval ($0.3 \leq \alpha \leq 60$). The Brown[4] formalism being inadequate to describe a paramagnetic state ($T > T_c$), the only way we have to reach our aim is to evaluate τ_1 around $T \leq T_c$ where the Brown model is at its limit of validity, and to suppose that the behavior of τ_1 for $T > T_c$ can be deduced from that approximation. This assumes that the relaxation time is a continuous function at $T = T_c$. We think that such an assumption is a good one as far as we can imagine the relaxation process taking place continuously from a particle relaxation form for $T < T_c$ temperature range to a spin-relaxation form for the $T > T_c$ range. In this case, we find for the preexponential factor τ_0 from a development of τ_1 in the neighborhood of $1/T = 1/T_c$ as a Taylor series, which is valid only as far as we approximate the effect of the term V/kT through the function $g(\alpha)$:

$$\tau_0 = \begin{cases} c_1 \left[\dfrac{1}{K_c} + \left[\delta_1 - 1 + \dfrac{\zeta}{4} \right] \dfrac{V}{k_b} \left[\dfrac{1}{T} - \dfrac{1}{T_c} \right] \right] \dfrac{1}{h'} \dfrac{1}{(1+\alpha/4)^{\zeta}} & \text{if } T \leq T_c , \\ c_1 \left[\dfrac{1}{K_c} \right] \dfrac{1}{h'} \dfrac{1}{(1+\alpha/4)^{\zeta}} & \text{if } T > T_c \end{cases} \tag{39}$$

and that

$$\lim_{1/T \rightarrow 0} \tau = \tau_0 (1/T = 0) = c_1 \frac{1}{h'(1/T = 1/T_c) K_c} , \tag{40}$$

where K_c is magnetocrystalline energy constant and $\zeta = \frac{5}{2}$. We have tried to fit Eq. (40) to the results obtained on small iron particles embedded in an alumina matrix.[16] As shown by Dormann, Bessais, and Fiorani,[16] a dynamical model for the superparamagnetic particles describing their mutual magnetic interactions is necessary to recover the experimental results. This model leads to the K_t expression:

$$K_t V = K_u V + E_{in} \quad \text{with} \quad E_{in} = M^2 V \sum_j b_j L \left[\frac{M^2 V a_j}{k_b T} \right] , \tag{41}$$

where $L(x)$ is the Langevin function. Three parameters have been considered in the fit: M^2V/k_b, K_uV/k_b, and the ratio b_j/a_j, which has been supposed independent of j. On the other hand, a_j has been determined from the ratio V/d^3, considering a compact arrangement of particles. The ratio V/d^3, where d is the mean distance between particles, is known from microprobe analysis results. Finally, the a parameter included in h' formulae (36) has been taken equal to 1. A very good fit is obtained with $M^2V/k_b = 300$ K, $K_uV/k_b = 380$ K, and $b_j/a_j = 1.15$ and the resulting curve is plotted in Fig. 2.

From electron microscopy measurements, the mean particle diameter can be evaluated as 55 ± 5 Å for the $S13$ sample (Ref. 16). For the same sample the magnetization is equal to 760 ± 80 emu/cm^3 (Ref. 16). This leads to $M^2V/k_b = 390 \pm 180$ K, which is in agreement with the fitted value. Concerning K_u, the main anisotropies, outside those resulting from mutual magnetic interactions, are magnetocrystalline anisotropy (cubic for metallic iron with $K_c = 5 \times 10^5$ ergs/cm^3) and magnetostatic anisotropy. This former is typically of uniaxial form because the particles are spherical as seen by electron microscopy,

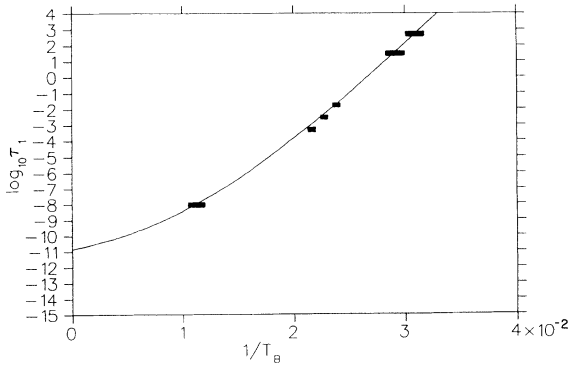

FIG. 2. Variation of the blocking temperature T_B in the classical plot of $\log_{10}\tau$ vs $1/T_B$ for the $S13$ sample. The full curve corresponds to the fit with the analytical formulation and the error bars correspond to the experimental results.

and the correspondent anisotropy constant K_m is equal to $K_m = \frac{1}{2}M^2(N_a - N_c)V/V_{Fe^0}$, where N_c and N_a are the demagnetizing factors of the particles, and V_{Fe^0} is the volume corresponding to the metallic iron (our particles are oxidized on the surface). It has been shown[16] that in first approximation the anisotropy is of uniaxial form when $K_m > K_c/2$ and the resulting K_u is equal to K_m if $K_m \geq K_c$. From the fitted values $K_u/M^2 = 1.27$ and it is possible to deduce (considering $V_{Fe^0}/V = 0.55$ as deduced from Mössbauer spectroscopy[16]) that K_m is higher than K_c and that $N_a - N_c = 1.35$ is a very reasonable value for our spherical particles. All these parameters differ only slightly from previous determinations.[16]

The good agreement, obtained for the parameters values, between the determination from our model considering the uniaxial symmetry and those from various techniques (electron microscopy, microprobe analysis, Mössbauer spectroscopy, magnetization and susceptibility measurement) indicates, in our opinion, a strong support for uniaxial symmetry as a good starting model for the study of fine magnetic particles. However, experimental results, not too far from T_c would be necessary to verify the validity of the suggested methods. Unfortunately, experimental results exist only at low temperature (generally very far from T_c). As can be seen in Fig. 2, the last experimental point toward the boundary $1/T \rightarrow 1/T_c$ comes from Mössbauer data $\tau_m \simeq 10^{-8}$ s. As noted by the authors,[16] the determination of the blocking temperature corresponding to this data is difficult: The related uncertainties are fairly large. Consideration of new experimental techniques that reach higher temperatures and probe shorter time scales seems essential to determine τ not too far from T_c.

The existence of quantum fluctuations (such as tunneling[27] or noise[10]) is also an interesting question. But a precise τ formulation as well as accurate experiments at low temperatures are necessary to determine the strength of their effect. We think that our formula, valid over a large range of α values, is useful for this purpose. However, it is not clear to us whether quantum fluctuations, which arise probably below a certain critical volume, can be expressed only via a change in τ_0, as suggested by Jones and Srivastava,[9] which relate the R parameter to the microscopic origin of the relaxation, or via a τ change. Let us recall that τ represents the probability of magnetic moment reversals. Then a formula such as $1/\tau = 1/\tau_{qu} + 1/\tau_{ba}$, where τ_{qu} and τ_{ba} are related to relaxation times corresponding respectively to quantum and barrier effects, would seem to be a more adequate approximation.

V. CONCLUSION

In this paper we have presented a method for the solution of the partial differential equation resulting from Brown's model of relaxation of small particle magnetization in the case of uniaxial symmetry.

This method permits us to extend numerical calculations of the relaxation time τ_1 and leads to only one approximate formula for τ_1 valid for all experimental cases, when previous calculations gave two approximations (high and low energy) which did not cover all the relaxation time range. Good agreement is obtained with experimental results. On the other hand, it is concluded that Brown's treatment is valid when the magnetization is not uniform, which is always the case for small particles, and the meaning of the dissipation constant and its effect on the preexponential factor τ_0 is discussed.

Finally, the asymptotic behavior of τ_0 when the temperature is near the Curie value and the possible influence of quantum fluctuations are discussed.

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APPENDIX

To obtain Eq. (10), depending on the parity of the Fourier index k, we have, using Eq. (9), to replace each quantity $W_k(t,\theta)$ and E_{k-m} by the corresponding expansion series, to multiply the entire equation by $\cos(n\theta)$ if k is even and by $\sin(n\theta)$ if k is odd, and finally to integrate the resulting equation over the interval $[0,\pi]$, which gives the required equations if we use the following identities corresponding to some general properties of the integrals of trigonometric functions. To clarify the notation, δ denotes the Dirac delta function.

When the index k is even we obtain:

$$I(l,s) \equiv \frac{1}{\pi}\int_0^{\pi} \frac{\cos(l\theta)\cos(s\theta)}{\sin^2\theta} d\theta = \begin{cases} -\frac{1}{8}[(l+s)(l+s-2)+(l-s)(l-s-2)] & \text{if } (1+s) \text{ is even} \\ 0 & \text{if } (1+s) \text{ is odd} , \end{cases} \tag{A1}$$

$$J(l,s) \equiv \frac{1}{\pi}\int_0^{\pi} \sin(l\theta)\cos(s\theta)\cot\theta\, d\theta = \frac{1}{4}[\epsilon(l+s+1)+\epsilon(l+s-1)+\epsilon(l-s-1)+\epsilon(l-s+1)] , \tag{A2}$$

where $\epsilon(l+s+1)$ denotes the sign of $(l+s+1)$.

$$K(l,s)\equiv\frac{1}{\pi}\int_0^{\pi}\cos(l\theta)\cos(s\theta)d\theta=\tfrac{1}{2}(\delta_{l+s,0}+\delta_{l-s,0})\ , \tag{A3}$$

$$\begin{aligned}L(l,q,s)\equiv&\frac{1}{\pi}\int_0^{\pi}\cos(l\theta)\cos(s\theta)\cot\theta\sin(q\theta)d\theta\\=&\tfrac{1}{8}\{\epsilon[q-(l+1)-s]+\epsilon[q+(l+1)+s]+\epsilon[q+(l+1)-s]+\epsilon[q-(l+1)+s]+\epsilon[q+(l-1)-s]\\&+\epsilon[q+(l-1)+s]+\epsilon[q-(l-1)-s]+\epsilon[q-(l-1)+s]\}\ ,\end{aligned} \tag{A4}$$

$$\begin{aligned}L'(l,q,s)\equiv&\frac{1}{\pi}\int_0^{\pi}\frac{\cos(l\theta)\cos(s\theta)\sin(q\theta)}{\sin\theta}d\theta\\=&\tfrac{1}{4}[\epsilon(l+s+q)+\epsilon(l+s-q)+\epsilon(l-s-q)+\epsilon(l-s+q)]\ ,\end{aligned} \tag{A5}$$

$$\begin{aligned}M(l,q,s)\equiv&\frac{1}{\pi}\int_0^{\pi}\cos(l\theta)\cos(s\theta)\cos(q\theta)d\theta\\=&\tfrac{1}{4}(\delta_{l+q+s,0}+\delta_{l+q-s,0}+\delta_{l-q-s,0}+\delta_{l-q+s,0})\ ,\end{aligned} \tag{A6}$$

$$\begin{aligned}N(l,q,s)\equiv&\frac{1}{\pi}\int_0^{\pi}\sin(l\theta)\cos(s\theta)\sin(q\theta)d\theta\\=&\tfrac{1}{4}(\delta_{l+q+s,0}+\delta_{l+q-s,0}+\delta_{-l+q-s,0}+\delta_{l-q-s,0})\ ,\end{aligned} \tag{A7}$$

$$O(l,q,s)\equiv L'(q,l,s)\ , \tag{A8}$$

$$\begin{aligned}P(l,q,s)\equiv&\frac{1}{\pi}\int_0^{\pi}\frac{\cos(l\theta)\cos(s\theta)\cos(q\theta)}{\sin^2\theta}d\theta\\=&-\tfrac{1}{32}[(l+q+s)+(l+q+s-2)+(l+q-s)+(l+q-s-2)+(l-q+s)+(l-q+s-2)\\&+(l-q-s)+(l-q-s-2)]\ \text{if } (1+q+s) \text{ is even}\ ,\\=&0\ \text{if } (l+q+s) \text{ is odd}\ ,\end{aligned} \tag{A9}$$

$$Q(l,q,s)\equiv L(q,l,s)\ , \tag{A10}$$

$$Q'(l,q,s)\equiv L'(q,l,s)\ , \tag{A11}$$

$$R(l,q,s)\equiv N(l,q,s)\ , \tag{A12}$$

$$S(l,q,s)\equiv M(l,q,s)\ , \tag{A13}$$

$$T(l,q,s)\equiv L'(l,q,s)\ , \tag{A14}$$

$$\begin{aligned}U(l,q,s)\equiv&\frac{1}{\pi}\int_0^{\pi}\frac{\sin(l\theta)\cos(s\theta)\sin(q\theta)}{\sin^2\theta}\\=&-\tfrac{1}{16}[(l-q+s)+(l-q+s-2)-(l+q-s)+(l+q-s-2)+(l-q-s)+(l-q-s-2)\\&-(l+q+s)+(l+q+s-2)]\ \text{if } (l+q+s) \text{ is even}\ ,\\=&0\ \text{if } (l+q+s) \text{ is odd}\ .\end{aligned} \tag{A15}$$

Similar identities can be derived without serious difficulties when the index k appearing in Eq. (9) is odd.

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