Longitudinal complex magnetic susceptibility and relaxation times of superparamagnetic particles with triaxial anisotropy

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The longitudinal relaxation time and spectrum of the complex magnetic susceptibility of single domain ferromagnetic particles with triaxial (orthorhombic) anisotropy are calculated by averaging the Gilbert-Langevin equation for the magnetization of an individual particle and by reducing the problem to that of solving a system of linear differential-recurrence relations for the appropriate equilibrium correlation functions. The solution of this system is obtained in terms of matrix continued fractions. It is shown that in contrast to the linear magnetic response of particles with uniaxial anisotropy, there is an inherent geometric dependence of the complex susceptibility and the relaxation time on the damping parameter arising from coupling of longitudinal and transverse relaxation modes. Simple analytic equations, which allow one to understand the qualitative behavior of the system and to accurately predict the spectrum of the longitudinal complex susceptibility in wide ranges of the barrier height and dissipation parameters, are proposed.

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I. INTRODUCTION

The single domain ferromagnetic particles, which are characterized by an internal anisotropy potential, may have several positions of local equilibrium of the magnetization with potential barriers separating them. If the particles are small (∼100 Å), thermal fluctuations may cause the magnetization vector \( \mathbf{M} \) to reorient itself over the barriers from one equilibrium position to another. The instability of the magnetization due to thermal agitation results in superparamagnetism because each fine particle behaves like an enormous paramagnetic atom having a magnetic moment \( ∼10^4–10^5 \) Bohr magnetons. The superparamagnetism of single-domain ferromagnetic nanoparticles is important in the context of rock magnetism and technology because of the ever decreasing size of the particles used in magnetic recording.

The pioneering theory of thermal fluctuations of the magnetization \( \mathbf{M}(t) \) of a single domain ferromagnetic particle due to Néel was further developed by Brown using the theory of the Brownian motion. Brown proceeded by taking as Langevin equation, Gilbert’s equation for the distribution function \( W(\mathbf{M},t) \) of the orientations of the magnetization vector \( \mathbf{M} \).

where \( \mathbf{u} = \mathbf{M}/M_s \) is the unit vector directed along \( \mathbf{M} \), \( M_s \) is the saturation magnetization, \( \gamma \) is the gyromagnetic ratio, \( \alpha \) is the dimensionless damping (dissipation) parameter, \( \mathbf{H}_g = -\gamma V/\partial \mathbf{M} \), \( V \) is the free energy per unit volume (characterizing the magnetic anisotropy and Zeeman energy density of the particle), and a random field \( \mathbf{h}(t) \) with white noise properties, accounting for the thermal fluctuations of the magnetization of an individual particle. Brown derived from the Gilbert-Langevin Eq. (1), the Fokker-Planck equation for the distribution function \( W(\mathbf{M},t) \) of the orientations of the magnetization vector \( \mathbf{M} \).
magnetic characteristics on the value of the damping parameter \( \alpha \) arising from coupling of the longitudinal and transverse relaxation modes. For nonaxially symmetric potentials, Fokker-Planck Eq. (2) can be solved by expanding \( W \) as a series of spherical harmonics so yielding an infinite hierarchy of differential-recurrence equations for the statistical moments (averaged spherical harmonics or appropriate correlation functions). The system of moment equations can be solved by calculating the eigenvalues and eigenvectors of the system matrix (e.g., Refs. 16 and 17) or by a matrix continued fraction method. The last approach has been used recently for the study of the magnetization dynamics of particles with cubic anisotropy and uniaxial anisotropy in the presence of an external dc magnetic field, which breaks the axial symmetry.

In the present paper, we use the continued fraction method to calculate the longitudinal complex magnetic susceptibility and relaxation time of single domain particles with triaxial (i.e., orthorhombic) anisotropy, where the free energy density is given by

\[
V = -K_1 \sin^2 \vartheta \cos^2 \varphi - K_2 \sin^2 \vartheta \sin^2 \varphi - K_3 \cos^2 \vartheta + \text{const},
\]

(\( K_1 < K_2 < K_3 \) are the anisotropy constants). In spite of the practical importance of orthorhombic anisotropy, which may yield an essential contribution to the free energy density of magnetic nanoparticles, the orthorhombic case is to some extent incomplete. The only available appropriate formula for the relaxation time of the magnetization of orthorhombic crystals has been given by Smith and de Rozario in the low-temperature limit and intermediate-to-high damping [(HDD) \( \alpha \gg 1 \)]. A very similar problem of magnetization reversal in elongated particles (where easy and hard-axis anisotropy terms present) in the presence of a strong dc magnetic field has been treated by Braun but also in the HDD limit only. Some quantum and field effects for magnetic relaxation of biaxial particles have been treated, for instance, in Refs. 28–30. Here we present the results of a study of the longitudinal complex magnetic susceptibility \( \chi_1(\omega) \) and relaxation time \( \tau_1 \) of single domain particles with triaxial anisotropy for wide ranges of the anisotropy energy and dissipation parameters. Numerical results obtained with the help of matrix continued fractions are compared with asymptotic estimates based on Kramers’ escape rate theory.

II. LONGITUDINAL DYNAMIC SUSCEPTIBILITY AND RELAXATION TIMES

According to linear response theory (Ref. 14, Chap. 2), the decay of the longitudinal component of the magnetization \( \langle M_x(t) \rangle \) of a single domain particle, when a small constant external field \( H_1 \), \( \beta (\mathbf{M} - H_1) \ll 1 \), applied along the \( z \) axis (which is the easy axis of the particle) has been switched off at time \( t=0 \), is

\[
\langle M_x(t) \rangle = M_S \int_0^\pi \int_0^{2\pi} \cos \theta W(\theta, \varphi, t) \sin \theta d\theta d\varphi = \chi_1(\omega) C_0(t),
\]

where

\[
C_0(t) = \frac{\langle \cos \theta(0) \cos \theta(t) \rangle_0}{\langle \cos^2 \theta(0) \rangle_0} = \sum_k c_k e^{-\lambda_k t}
\]

is the normalized equilibrium autocorrelation function of the longitudinal component of the magnetization, \( \lambda_k \) are the eigenvalues of the Fokker-Planck operator \( L_{FP} \) from Eq. (2), \( \sum_k \lambda_k = 1, \chi_1 = \beta M_S^2 \langle \cos^2 \theta \rangle_0 \) is the static longitudinal magnetic susceptibility of the particle, and the brackets \( \langle \cdot \rangle_0 \) designate the equilibrium ensemble average defined as

\[
\langle A \rangle_0 = \frac{1}{Z} \int_0^\pi \int_0^{2\pi} A(\theta, \varphi) e^{-\beta V(\theta, \varphi)} \sin \theta d\theta d\varphi
\]

(\( Z \) is the partition function). The correlation function \( C_0(t) \) completely determines the transient longitudinal relaxation of the magnetization. Moreover, it allows one to evaluate the ac response of the system to a small ac perturbing magnetic field, namely, the longitudinal complex susceptibility \( \chi_1(\omega) = \chi_1(\omega) - i \chi_1'(\omega) \), which is given by

\[
\chi_1(\omega)/\chi_1 = 1 + i \omega \int_0^\infty e^{-i\omega t} C_0(t) dt.
\]

According to Eq. (8), the behavior of \( \chi_1(\omega) \) in the frequency domain is completely determined by the time behavior of \( C_0(t) \). In order to characterize quantitatively the time behavior of \( C_0(t) \), one may formally introduce two time constants. These are the integral relaxation (or correlation) time \( \tau_1 \) defined as the area under \( C_0(t) \), viz. 14

\[
\tau_1 = \int_0^\infty C_0(t) dt = \sum_k c_k/\lambda_k,
\]

and the effective relaxation time \( \tau_1' \) given by

\[
\tau_1' = -1/\dot{\chi}_1(0) = \left( \sum_k c_k/\lambda_k \right)^{-1},
\]

[which yields precise information on the initial decay of \( C_0(t) \) in the time domain].

III. MATRIX CONTINUED FRACTION SOLUTION

We can calculate numerically the relaxation time \( \tau_1 \) and the dynamic susceptibility \( \chi_1(\omega) \) by using the matrix continued fraction approach developed in Ref. 14. According to this approach, the solution of the Gilbert-Langevin Eq. (1) for any anisotropy potential can be reduced to the solution of an infinite hierarchy of differential-recurrence equations for the statistical moments (equilibrium correlation functions)

\[
c_{lm}(t) = \langle \cos \theta(t) \rangle_0 Y_{lm} \left[ \theta(t), \varphi(t) \right]_0 \quad [\text{so that } c_{1,0}(t)/c_{1,0}(0) = C_0(t)]
\]

governing the dynamics of the magnetization. Here \( Y_{lm}(\theta, \varphi) \) is the spherical harmonic defined as

\[
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\]

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\[ Y_{l,m}(\vartheta, \varphi) = (-1)^n \sqrt{\frac{(2l + 1)(l-m)!}{4\pi(l+m)!}} e^{im\varphi} P^m_l(cos \vartheta), \]

where the \( P^m_l(x) \) are the associated Legendre functions. For the problem in question, one can obtain the differential-recurrence equations for \( c_{l,m}(t) \) using a general formula derived in Ref. 32 (see also Ref. 14, Chap. 7). Thus noting that the free energy density \( V \) from Eq. (4) is expressed in terms of \( Y_{l,m} \) as

\[
V = \sqrt{\frac{2\pi}{15}}(K_2 - K_1)[Y_{2,2}(\vartheta, \varphi) + Y_{2,-2}(\vartheta, \varphi)]
- \frac{4}{3} \sqrt{\frac{\pi}{5}}(K_3 + \frac{K_1-K_2}{2}) Y_{2,0}(\vartheta, \varphi) + \text{const},
\]

we have the 15th term differential-recurrence equation

\[
\tau_N \frac{d}{dt} c_{n,m}(t) = v_{n,m} c_{n-1,m}(t) + w_{n,m} c_{n+1,m}(t)
+ y_{n,m} c_{n+1,m+1}(t) + z_{n,m} c_{n-1,m+2}(t)
+ w_{n,m} c_{n-1,m+2}(t) + x_{n,m} c_{n+1,m+2}(t)
+ y_{n,m} c_{n+1,m+2}(t) + z_{n,m} c_{n-1,m+2}(t)
+ y_{n,m} c_{n+1,m+2}(t) + z_{n,m} c_{n+1,m+2}(t),
\]

where \( n \gg 1, -n \leq m \leq n \) and the coefficients \( v_{n,m}, w_{n,m}, \) etc. are defined in Appendix A. Equation (11) can be transformed into the three-term vector recurrence equation

\[
\tau_N \frac{d}{dt} C_n(t) = Q_n C_{n-1}(t) + Q_m C_n(t) + Q_{n+1} C_{n+1}(t), \quad (n \gg 1),
\]

where \( C_n(t) \) are the column vectors arranged in an appropriate way from \( c_{n,m}(t) \), viz.

\[
C_n(t) = \left( \begin{array}{c} c_{2n-2}(t) \\ c_{2n-2+1}(t) \\ \vdots \\ c_{2n-1}(t) \\ c_{2n-1+1}(t) \\ \vdots \\ c_{2n}(t) \end{array} \right), \quad (n \gg 1),
\]

and \( Q^-_n, Q_n, Q^+_n \) are the supermatrices given in Appendix A. The exact solution of Eq. (12) for the Laplace transform \( \tilde{C}_1(s) = \int_0^\infty C_1(t)e^{-st}dt \) can be given in terms of matrix continued fractions

\[
\tilde{C}_1(s) = \tau_N \Delta_1(s) \left( C_1(0) + \sum_{n=2}^\infty \prod_{k=2}^n Q_k^{-1} \Delta_k(s) \right) C_1(0),
\]

where the infinite matrix continued fraction \( \Delta_n(s) \) is defined as

\[
\Delta_n(s) = \frac{1}{\tau_N s I - Q_n - Q_n^+} \left( \begin{array}{c} I \\ -Q_n^+ \\ \vdots \\ -Q_{n+1}^+ \end{array} \right) \frac{1}{\tau_N s I - Q_{n+1} - Q_{n+1}^+} \left( \begin{array}{c} I \\ -Q_{n+2}^+ \vdots \\ -Q_{n+1}^+ \end{array} \right),
\]

(14)

\[
\Delta_1(s) = \frac{1}{\tau_1 s I - Q_1 - Q_1^+} \left( \begin{array}{c} I \\ -Q_1^+ \\ \vdots \\ -Q_{n+2}^+ \end{array} \right),
\]

(13)

(15)

\[
\tau_1 = \tilde{C}(0) = \tilde{c}_{1,0}(0)/c_{1,0}(0)
\]

as well as the spectrum of the correlation function \( \tilde{C}(\omega) = \tilde{c}_{1,0}(\omega)/c_{1,0}(0) \) and thus the complex susceptibility from Eq. (8). Moreover, by using matrix continued fractions, one can also evaluate the smallest nonvanishing eigenvalue \( \lambda_1 \).

The matrix continued fraction approach provides an effective method of computation of the susceptibility \( \chi_{\vartheta}(\omega) \) and correlation time \( \tau_1 \) (algorithms for calculating matrix continued fractions are discussed in Refs. 14 and 18). The advantage of the matrix continued fraction approach is that it applies to the case, where the magnetic anisotropy energy is comparable to the thermal energy \( kT \). Nevertheless, its application is rather limited since the dependence of \( \chi_{\vartheta}(\omega) \) and \( \tau_1 \) on the model parameters (damping coefficient, anisotropy constants) is not obvious by this method.

**IV. ASYMPTOTIC FORMULAS**

The qualitative behavior of \( \chi_{\vartheta}(\omega) \) and \( \tau_1 \) can readily be understood in the low temperature limit, where the magnetization relaxation is determined by the smallest nonvanishing eigenvalue \( \lambda_1 \). Indeed, according to Eq. (9), the correlation time \( \tau_1 \) contains contributions from *all* the eigenvalues \( \lambda_k \).
The smallest nonvanishing eigenvalue $\lambda_1$ is associated with the slowest overbarrier relaxation mode and so with the longtime behavior of $C_{\parallel}(t)$; the other eigenvalues $\lambda_k$ characterize high-frequency “intrawell” modes. In general, in order to evaluate $\tau_i$ numerically, a knowledge of all the $\lambda_k$ and $c_k$ is required. However, in the low temperature (high barrier) limit, $\lambda_1 \ll |\lambda_k|$ and $c_1 \simeq 1 \gg c_k$ ($k \neq 1$) provided the wells of the potential remain equivalent (as for the triaxial potential) so that

$$\tau_i \approx 1/\lambda_1. \quad (16)$$

In other words, the inverse of the smallest nonvanishing eigenvalue closely approximates the correlation time $\tau_i$ in the low temperature limit.

The smallest nonvanishing eigenvalue $\lambda_1$ may be estimated with the help of the Kramers escape rate theory as extended to the magnetic problem by Brown, Smith and de Rozario, Klik and Gunther, and Coffey et al. We recall that in order to estimate the characteristic time of reversal of the magnetic moment over the internal anisotropy potential barrier of a uniaxial particle, Brown adapted an ingenious method originally proposed by Kramers in connection with thermally activated escape of Brownian particles out of a high barrier. In general, in order to apply the method originally proposed by Kramers in connection with thermally activated escape of Brownian particles out of a high barrier, it is necessary to consider the escape rates as a function of the dissipation parameters of the Kramers method to magnetic problems see Refs. 14, 15, 27, and 35.

Using the approach of Coffey et al., the universal formula for the relaxation times (which is universal in the sense that it is valid for all values of damping including IHD and VLD regions) is given by

$$\lambda_i^{-1} \sim \tau_{\text{IHD}} A(8\alpha a^2 \sqrt{\delta}) / A^2(4\alpha a^2 \sqrt{\delta}). \quad (17)$$

where $\tau_{\text{IHD}}$ is the longest relaxation time of an orthorhombic crystal derived by Smith and de Rozario in the IHD limit (in our notation)

$$\tau_{\text{IHD}} = \frac{\pi e^{\sigma} (1 + \alpha^2)}{\alpha a^2 \sqrt{1 + 1/\delta} [1 - \delta + \sqrt{(1 + \delta^2) + 4 \delta^2 a^2}]. \quad (18)$$

where $\delta \equiv \Delta/\sigma$, $\Delta = \beta(K_2 - K_1) > 0$, and $\sigma = \beta(K_3 - K_2) > 0$ are the dimensionless anisotropy and barrier height parameters, respectively, $\tau_i = \tau_{\text{IHD}} a^2 / (1 + \alpha^2) = \beta M_s / (2\gamma)$ is an $\alpha$-dependent characteristic time, and

$$A(\alpha S) = \exp \left[ \frac{1}{\pi} \int_0^\infty \ln[1 - \exp(-\alpha S(x^2 + 1/4))] \times (x^2 + 1/4)^{-1} dx \right]. \quad (19)$$

Noting that $A(\alpha S)/\alpha \rightarrow S$ as $\alpha \rightarrow 0$, Eq. (18) yields

$$\frac{\tau_{\text{IHD}}}{\tau_0} \approx \frac{\pi e^{\sigma}}{4\alpha a^2 \sqrt{1 + \delta}}, \quad (19)$$

which is in agreement with estimations in the context of the Klik and Gunther theory.

In order to understand the qualitative behavior of the complex susceptibility $\chi_i(\omega)$, one can use a simple analytical equation derived in Refs. 37 and 38 (see also Ref. 14, Chaps. 7-9). According to Ref. 37, the correlation function $C_i(t)$ [which in general comprises an infinite number of decaying exponentials, see Eq. (6)] may be approximated in the IHD limit by two exponentials only, viz.

$$C_{\parallel}(t) = \Delta_1 e^{-\lambda_1 t} + (1 - \Delta_1) e^{-\lambda_2 t}, \quad (20)$$

where $\Delta_1$ and $\tau_0$ are expressed in terms of $\tau_{\text{IHD}}$, and $\lambda_1$ and $\lambda_2$ is given as

$$\chi_1(\omega) \sim \frac{\Delta_1}{1 + i\omega/\lambda_1} + \frac{1 - \Delta_1}{1 + i\omega/\lambda_2}. \quad (21)$$

The parameters $\Delta_1$ and $\tau_0$ in Eq. (21) are determined in such way as to guarantee the correct asymptotic behavior of $\chi_i(\omega)$ in the extreme cases of very low and very high frequencies

$$\chi_1(\omega) \sim \begin{cases} \frac{C_i(0)}{i\omega} + \cdots = - \frac{i}{\omega \tau_i} + \cdots, & \omega \rightarrow 0, \\ 1 - i\omega \int_0^\infty C_i(t)dt = 1 - i\omega \tau_i, & \omega \rightarrow \infty. \end{cases} \quad (22)$$

Equation (22) was derived and tested for particles with uniaxial (at all damping) and cubic anisotropy (at IHD values of damping, $\alpha \gg 1$). For $\alpha \ll 1$, where the interactions between the longitudinal and transverse modes cannot be ignored, Eq. (21) may be used at $\omega \tau_0 \ll 1$.

In practical calculations, Eq. (21) requires a knowledge of $\tau_{\text{IHD}}$, $\tau_i$, and $\lambda_1$. The smallest eigenvalue may readily be evaluated from Eq. (17). The effective relaxation time $\tau_i$ is given by an exact analytic equation

$$\tau_i^0 = 2\tau_{\text{IHD}}(\cos^2 \theta)_{\text{IHD}} (1 - (\cos^2 \theta)_{\text{IHD}})^{-1}, \quad (23)$$

where $(\cos^2 \theta)_{\text{IHD}}$ can be calculated from Eq. (7). Unfortunately, there is no simple equation for the correlation time $\tau_i$...
[Eq. (16) is unreliable here as it yields \( \tau_W = 0 \)]. However, one can overcome this problem\(^{38} \) by noting that the intrawell relaxation time \( \tau_W \) can be estimated in the low-temperature limit, \( \sigma \gg 1 \), from the deterministic Gilbert equation as

\[
\tau_W \sim \frac{1}{\omega_{\text{well}}} = \frac{\tau}{2\sigma \sqrt{1 + \delta}},
\]

where \( \omega_{\text{well}} = \gamma/M S^2 (\partial^2 V/\partial \phi^2)(\partial^2 V/\partial t^2) \) is the well angular frequency. Noting Eqs. (21), (22), and (24), one can evaluate \( \Delta_1 \) as

\[
\Delta_1 \approx 1 - \tau_W / \tau^f.
\]

Equations (21) and (23)–(25) and allow one readily to calculate \( \chi(l) \).

V. RESULTS AND DISCUSSION

The greatest relaxation time predicted by the universal Eq. (17) and the correlation time \( \tau_l \) calculated numerically by the matrix continued fraction method for triaxial anisotropy are shown in Figs. 1 and 2 (as a function of the damping parameter \( \alpha \)), and 3 (as a function of the barrier height parameter \( \sigma \)). Apparently, at high barriers, \( \sigma \gg 5 \), the asymptotic Eq. (17) provides a good approximation of \( \tau_l \) for all values of \( \alpha \) (Figs. 1 and 2) and \( \Delta \gg 1 \) (Fig. 3). We emphasize that Eq. (17) is not valid for \( \delta = \Delta / \sigma \rightarrow 0 \) corresponding to uniaxial anisotropy. Here \( \tau_l \lambda_1 \) is given by Brown’s formula\(^{3,4} \)

\[
\tau_l \lambda_1 \sim 2\sigma^{3/2} e^{-\sigma / \sqrt{\pi}}.
\]

The uniaxial asymptote Eq. (26) is shown in Fig. 3 for comparison. It follows that the triaxial anisotropy causes the various damping regimes (IHD and VLD) of relaxation to appear unlike in an axially symmetric potential.

Results of the calculation from Eqs. (21) and (23)–(25), and those obtained using matrix continued fractions are compared in Figs. 4–6. Here the imaginary part of the normalized susceptibility \( \chi(l)(\omega) \) \((\beta M \mu = 1)\) is plotted for typical values of the model parameters \( \sigma, \Delta, \) and \( \alpha \). The results indicate that a marked dependence of \( \chi(l)(\omega) \) on \( \alpha \) exists and that three distinct dispersion bands appear in the spectrum. The characteristic frequency and half-width of the low-frequency band are completely determined by the smallest nonvanishing eigenvalue \( \lambda_1 \). Thus the low frequency behavior of \( \chi(l)(\omega) \) is dominated by the barrier crossing mode. In addition, a far weaker second relaxation peak appears at high frequencies. This high frequency relaxation band is due to the intrawell modes. The characteristic frequency of this band is \( \omega_{\text{well}} \sim \tau^f / (2\sigma \sqrt{1 + \delta}) \). The third ferromagnetic resonance (FMR)
peak due to excitation of transverse modes having the peak frequency close to the precession frequency \( \omega_p = \gamma H_0 / l \) of the magnetization appears only at low damping \( \alpha \ll 1 \) and strongly manifests itself in the high frequency region. As \( \alpha \) decreases, the FMR peak shifts to higher frequencies since \( \omega_p \sim \alpha^{-1} \) (see Fig. 4). As one can see in Figs. 4–6 the agreement between exact matrix continued fraction calculations and the approximate Eq. (21) is very good in the low-frequency region, \( \omega \tau_N \ll 1 \), for all values of damping because the low-frequency response is completely determined by the overbarrier relaxation mode. The approximate Eq. (21) yields a reasonable description of the high frequency relaxation band at IHV values of damping (\( \alpha \gg 1 \)), where one can ignore the interactions between the longitudinal and transverse modes. However, Eq. (21) does not allow one to describe the FMR peak which appears at very low damping, \( \alpha \ll 1 \).

One may conclude that the longitudinal magnetic susceptibility \( \chi(\omega) \) and relaxation time \( \tau_1 \) of systems of single domain particles with triaxial anisotropy may be evaluated exactly in terms of matrix continued fractions (for all values of model parameters) as well as in terms of simple analytic equations (in the low-temperature limit). In contrast to uniaxial particles, where the damping only enters in the diffusion time \( \tau_N \), for the particles with triaxial anisotropy, there is an inherent geometric dependence of \( \chi(\omega) \) and \( \tau_1 / \tau_N \) on the value of the damping parameter \( \alpha \) arising from coupling of the longitudinal and transverse relaxation modes. In the derivation of the above results, it was supposed that all particles are identical. In order to take into account the polydispersity of the particles of a real sample one must also average the susceptibility over appropriate distribution functions (e.g., over that of particle volumes). The approach developed can be applied with small modifications to the evaluation of the transverse and nonlinear responses of orthorhombic crystals, to the estimation of the effect of an external magnetic field on the relaxation behavior of the magnetization of such crystals, etc.

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APPENDIX A: MATRICES \( Q_n, Q_n^+, Q_n^- \)

The matrices \( Q_n, Q_n^+, Q_n^- \) are defined as

\[
Q_n = \begin{pmatrix} X_{2n} & W_{2n} \\ Y_{2n-1} & X_{2n-1} \end{pmatrix}, \quad Q_n^+ = \begin{pmatrix} Z_{2n} & Y_{2n} \\ 0 & Z_{2n-1} \end{pmatrix}, \quad Q_n^- = \begin{pmatrix} V_{2n} & 0 \\ W_{2n-1} & V_{2n-1} \end{pmatrix}.
\]

The dimensions of the matrices \( Q_n, Q_n^+, Q_n^- \) are accordingly equal to \( 8n \times 8n, 8n \times 8(n+1), \) and \( 8n \times 8(n-1) \). In turn, the matrices \( Q_n, Q_n^+, Q_n^- \) consist of submatrices. There are five distinct types of three diagonal submatrices \( V_l, W_l, X_l, Y_l, \) and \( Z_l \) which have the dimensions \( (2l+1) \times (2l+1), (2l+1) \times (2l+1), (2l+1) \times (2l+3), \) and \( (2l+1) \times (2l+5), \) respectively. The elements of these submatrices are given by

\[
(V_l)_{n,m} = \delta_l n m v_{l,-l+m+3} + \delta_{n-2m} v_{l,-l+m+1} + \delta_{n,m} v_{l,-l+m-1},
\]

\[
(W_l)_{n,m} = \delta_{n-3m} w_{l,-l+m+2} + \delta_{n-1m} w_{l,-l+m} + \delta_{n+1m} w_{l,-l+m-2},
\]

\[
(X_l)_{n,m} = \delta_{n-2m} x_{l,-l+m+1} + \delta_{n,m} x_{l,-l+m-1} + \delta_{n+2m} x_{l,-l+m-3},
\]

\[
(Y_l)_{n,m} = \delta_{n-1m} y_{l,-l+m} + \delta_{n+1m} y_{l,-l+m-2} + \delta_{n+3m} y_{l,-l+m-4},
\]

\[
(Z_l)_{n,m} = \delta_{n-2m} z_{l,-l+m+1} + \delta_{n+2m} z_{l,-l+m-3} + \delta_{n+4m} z_{l,-l+m-5},
\]

where
\[ v_{n,m} = \left( \sigma + \frac{\Delta}{2} \right) \left( \frac{n+1}{2(n-1)} \right) \sqrt{\frac{(n-1)^2 - \ell^2}{(n+1)(n-3)}}, \]

\[ \bar{v}_{n,m} = v_{n,m} = -\frac{\Delta (n+1)}{4(2n-1)} \times \sqrt{\frac{(n+m-3)(n+m-2)(n+m-1)(n+m)}{(2n+1)(2n-3)}}, \]

\[ w_{n,m} = -i \frac{\Delta (n+1)}{4(2n-1)} \times \sqrt{\frac{(n+m-3)(n+m-2)(n+m-1)(n+m)}{(2n+1)(2n-3)}}, \]

\[ \bar{w}_{n,m} = -w_{n,m} = -i \frac{\Delta (n+1)}{4(2n-1)} \times \sqrt{\frac{(n+m-3)(n+m-2)(n+m-1)(n+m)}{(2n+1)(2n-3)}}, \]

\[ x_{n,m} = \frac{n(n+1)}{2} + \left( \frac{\Delta}{2} + \sigma \right) \frac{n(n+1)-3m^2}{n(n+1)(2n+3)}, \]

\[ \bar{x}_{n,m} = x_{n,m} = 3\Delta \frac{\sqrt{(n+1)^2 - (m-1)^2}}{4} \frac{(n+1)^2 -(n-1)^2}{(2n+1)(2n+3)}, \]

\[ y_{n,m} = -i \left( \frac{\Delta}{2} + \sigma \right) \frac{m}{4} \sqrt{\frac{(n+1)^2 - m^2}{(2n+1)(2n+3)}}, \]

\[ \bar{y}_{n,m} = -y_{n,m} = \frac{\Delta}{4} \sqrt{\frac{(n+1)^2 - (m-1)^2}{(2n+1)(2n+3)}}, \]

\[ z_{n,m} = -\frac{\Delta}{2} + \sigma \frac{n}{2(n+3)} \sqrt{\frac{(n+1)^2 - m^2}{(2n+1)(2n+5)}}, \]

\[ \bar{z}_{n,m} = z_{n,m} = \frac{\Delta}{4} \frac{n}{2(n+3)}, \]

\[ \bar{z}_{n,m} = z_{n,m} = \frac{\Delta}{4} \frac{n}{2(n+3)}, \]

\[ \times \sqrt{\frac{(n+m+4)(n+m+3)(n+m+2)(n+m+1)}{(2n+1)(2n+5)}}. \]

**APPENDIX B: CALCULATION OF \( C_n(0) \)**

The initial condition vectors \( C_n(0) \) in Eq. (13) may be evaluated by noting that the equilibrium averages \( \langle Y_{i,m} \rangle_0 \) satisfy the following recurrence equation [cf. Eq. (11)]:

\[ v_{n,m}(Y_{n-2,m}) + w_{n,m}(Y_{n-1,m}) + x_{n,m}(Y_{n,m}) + y_{n,m}(Y_{n+1,m}) + z_{n,m}(Y_{n+2,m}) = 0. \]

\[ + x_{n,m}(Y_{n+2,m}) + y_{n,m}(Y_{n+1,m}) + z_{n,m}(Y_{n+2,m}) \]

\[ + v_{n,m}(Y_{n-2,m}) + w_{n,m}(Y_{n-1,m}) + x_{n,m}(Y_{n,m}) + y_{n,m}(Y_{n+1,m}) + z_{n,m}(Y_{n+2,m}) = 0. \]

Thus, one can transform the above equation into the tridiagonal vector recurrence equation

\[ Q_n R_{n-1} + Q_n R_n + Q_{n+1} R_{n+1} = 0, \quad (n \geq 1), \quad (B1) \]

where \( R_0 = 1/\sqrt{4\pi} \) and

\[ R_n = \begin{pmatrix} \langle Y_{2n-2m} \rangle_0 \\ \vdots \\ \langle Y_{2n-2m+1} \rangle_0 \\ \langle Y_{2n-2m+2} \rangle_0 \end{pmatrix}, \quad n \geq 1. \]

Equation (B1) has a solution \[14\]

\[ R_n = S_n R_{n-1} = S_n S_{n-1} \cdots S_2 S_1 / \sqrt{4\pi}, \]

where \( S_2 = -\Delta_1(0) Q_2 \). Using the identity \( c_{n,m}(0) \equiv d_{n+1,m}(Y_{n+1,m}) + d_{n,m}(Y_{n,m}) \), where \( d_{n,m} = \sqrt{(n^2 - m^2)(4n^2 - 1)} \), the initial condition vectors \( C_n(0) \) are given by

\[ C_n(0) = \begin{bmatrix} 0 & 0 & \cdots & 0 \\ D_{2n-1} & 0 \\ \vdots \\ D_{2n} & 0 \\ 0 & 0 \end{bmatrix} R_n \]

\[ + \begin{bmatrix} D_{2n} & 0 \\ 0 & 0 \end{bmatrix} R_{n+1}. \]

Here the superscript \( T \) denotes matrix transposition. The dimension of the matrix \( D_1 \) is \((2l+1) \times (2l+1)\) and its elements are given by \( (D_1)_{n,m} = \delta_{n-1,m} d_{l-i, l+i} \).
33 H. A. Kramers, Physica (Utrecht) 7, 284 (1940).