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Local field muon transverse depolarization and nuclear free induction decay at low temperatures

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Abstract. The form of the transverse depolarization function of an I spin due to dipolar interaction with a system of S spins is analysed for local field interactions without using the high-temperature approximation. This analysis is valid in the absence of diffusion and in the presence of a large, static external magnetic field. It is relevant to muons implanted in metals and to the free induction decay of dilute I nuclei in a system of S nuclei, for which the local field model is valid for short times. An analytic expression for the transverse depolarization function is obtained using the Boltzmann distribution for the occupation of Zeeman states of the S spins at a temperature T. The behaviour of the depolarization function depends on the ratio of the external magnetic field B to the temperature T and three regimes are identified. For small B/T the familiar high-temperature result is independent of B/T and sample shape. For intermediate values of B/T the depolarization function depends on B/T and has an envelope that is independent of sample shape and a time-dependent phase that is dependent on sample shape. For larger values of B/T the depolarization function is again independent of B/T with the phase remaining dependent on sample shape. The results indicate the effects that could be observed at values of B/T larger than those corresponding to the high-temperature approximation and provide the values of B/T at which these would be apparent.

1. Introduction

The general problem of calculating the transverse magnetization free induction decay (FID) due to magnetic dipolar interactions remains unsolved for crystals. In the absence of diffusion and in the presence of a sufficiently large external magnetic field, the only relevant parts of the dipolar Hamiltonian, for like spins, are the secular contributions A and B where the A term corresponds to classical local field effects and the B term causes flip-flops between pairs of spins because each precessing nuclear moment produces a time-varying magnetic field at the resonance frequency of the other (Abragam 1961). The difficulty in obtaining a general solution for the FID arises because the A and B terms do not commute.

The local field model consists of retaining only the A terms and an analytic expression for the FID, in the form of an infinite product, can then be obtained in the high-temperature limit (Lowe and Norberg 1957, Gade and Lowe 1966, Lee *et al* 1967). This work also developed methods for obtaining series corrections to the local field FID to account for the B terms (see also Betsuyaka 1970).

In the case of unlike-spin dipolar interactions, only the A terms are secular and the B terms can be neglected. In a two-spin (I and S) component system where the I spins are dilute, or have small magnetic moments, the dipolar interaction between the I spins will be negligible. The calculation of the FID for the I spins need then only consider the local field A term for the I–S interaction but both the A and B terms still need to be retained

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for the indirect S–S interaction effect on the I spin FID. The flip-flop effect of the B term for the S–S interaction is, however, similar to motional narrowing (Abragam 1961, p 123) and does not affect the form of the FID at short times.

The local field model is therefore an excellent approximation for dilute or weakly interacting I spins in a system of S spins, at short times, in the absence of diffusion and in the presence of an external magnetic field very much larger than the dipolar magnetic fields experienced by the spins. These are the restrictions that are imposed throughout this paper. An important example of such a system is the transverse depolarization of positive muons in metals (Schenck 1985). The depolarization function of muons implanted in metals with a polarization transverse to the magnetic field is identical to the FID, the muons are extremely dilute and only the very-short-time behaviour of the depolarization function can be observed because of the 2.2 μs half-life of the muons. When the external field is comparable to the dipolar field, or when diffusion occurs, all terms in the dipolar Hamiltonian must be included in the calculation of the depolarization function and this has recently been considered by Dalmas de Réotier and Yaouanc (1992) using an iterative procedure.

The muon depolarization function, in the absence of diffusion and in the presence of a large external field, is often assumed to be a Gaussian but a detailed local field calculation (Cameron and Sholl 1991) has shown this can be a poor approximation in some cases. This work also considered the accuracy of the commonly used Abragam approximation (Abragam 1961) when slow or rapid muon diffusion occurs. The local field theory in the absence of diffusion has also been extended (Cameron and Sholl 1993) to include the effect of electric quadrupolar interactions between the quadrupole moment of the metal spins and the electric field gradient produced by the muon.

All the above theory has assumed the high-temperature approximation for which the populations of the Zeeman states of the S spins are equal. The purpose of the present work is to examine the local field theory, in the absence of diffusion and quadrupolar interactions, for low temperatures at which the Boltzmann distribution for the populations of the Zeeman states must be taken into account. The results for the depolarization function can therefore indicate the effects that could be observed at low temperatures and the magnetic fields and temperatures which would be required to observe deviations from the high-temperature behaviour.

Since the interaction between the I spins is assumed negligible, the system to be considered is a single I spin in a system of S spins where only the local field A terms of the dipolar I–S and S–S interactions are retained. The I spin is assumed to have an initial non-zero polarization (magnetization) transverse to an external magnetic field. For implanted muons in metals this is a result of the polarization of the incident beam and for nuclei in crystals could be produced by a radio-frequency pulse. The depolarization function for such systems is derived in section 2. The explicit form of it at arbitrary temperatures and some examples are given in section 3 and section 4. The implications for the experimental observation of low-temperature effects are discussed in section 5.

2. Theory of transverse depolarization

A spin system consisting of one I spin and many identical S spins in a magnetic field $\boldsymbol{B}$ has the spin Hamiltonian

$$\mathcal{H} = \mathcal{H}_{ZI} + \mathcal{H}_{ZS} + \mathcal{H}_{d} = \hbar\omega_I I_z + \sum_j \hbar\omega_S S_{jz} + \mathcal{H}_{d} \tag{1}$$

where the first two terms are the Zeeman interactions of the I and S spins and $\mathcal{H}_d$ is the magnetic dipolar interaction between the I and S spins and between the S spins. The Larmor frequencies ω_I and ω_S are $\omega_I = -\gamma_I B$ and $\omega_S = -\gamma_S B$ where γ_I and γ_S are the respective gyromagnetic ratios.

The spin density operator ρ for this system has the equation of motion

$$\frac{d\rho}{dt} = \frac{i}{\hbar}[\rho, \mathcal{H}] \tag{2}$$

which has the solution

$$\rho(t) = \exp(-i\mathcal{H}t/\hbar)\rho(0)\exp(i\mathcal{H}t/\hbar) \tag{3}$$

when the Hamiltonian $\mathcal{H}$ is independent of time. The time development of the components of the polarization of the I spin transverse to B are the real and imaginary parts of the complex depolarization function $\langle I_+ \rangle$ given by

$$\langle I_+ \rangle = \mathrm{Tr}\{\rho(t)I_+\} = \mathrm{Tr}\{\exp(-i\mathcal{H}t/\hbar)\rho(0)\exp(i\mathcal{H}t/\hbar)I_+\} \tag{4}$$

where $I_+ = I_x + iI_y$ and the trace involves states for the I spin and all S spins. The density operator $\rho(0)$ may be written as $\rho_I(0)\rho_S$ where $\rho_I(0)$ depends on the way in which the initial non-equilibrium transverse polarization of the I spin is established. The ρ_S for equilibrium of the S spins in a magnetic field B and at a temperature T is

$$\rho_S = \exp[-\mathcal{H}_{ZS}/(kT)]/\mathcal{Z} = \prod_j \exp(-\alpha_S S_{jz})/\mathcal{Z}_j \tag{5}$$

where $\alpha_S = \hbar\omega_S/(kT)$ and $\mathcal{Z} = \prod_j \mathcal{Z}_j = \mathrm{Tr}_S\{\exp[-\mathcal{H}_{ZS}/(kT)]\}$ is the partition function with Tr_S denoting the trace over the S spins only. It will be seen that the value of the parameter α_S determines the magnetic field and temperature dependence of the depolarization function in the rotating frame. This dependence therefore occurs through the ratio B/T of the magnetic field and temperature.

The transformation to the frame of reference rotating at the Larmor frequency ω_I of the I spin is made by applying the transformation U^* of an operator U defined by

$$U^* = \exp(i\omega_I I_z t)U\exp(-i\omega_I I_z t) \tag{6}$$

to the operators ρ, $\mathcal{H}$ and I_+. Equation (4) then becomes

$$\langle I_+ \rangle = \mathrm{Tr}\{\rho^*(t)I_+^*\} = \exp(i\omega_I t)\langle I_+ \rangle^* \tag{7}$$

where the complex transverse depolarization in the rotating frame $\langle I_+ \rangle^*$ is

$$\langle I_+ \rangle^* = \mathrm{Tr}\{\rho^*(t)I_+\}. \tag{8}$$

The equation of motion of ρ^* is

$$\frac{d\rho^*}{dt} = \frac{i}{\hbar}[\rho^*, \mathcal{H}_d^*] \tag{9}$$

which has the solution

$$\rho^*(t) = \exp(-i\mathcal{H}_d^* t/\hbar)\rho(0)\exp(i\mathcal{H}_d^* t/\hbar) \tag{10}$$

if $\mathcal{H}_d^*$ is independent of time.

If the x and y axes are chosen so that the initial polarization along the y axis is zero, then at time $t = 0$, $\langle I_+ \rangle$ and $\langle I_+ \rangle^*$ are both given by $\mathrm{Tr}\{\rho(0)I_x\}$. The transverse depolarization functions, $F(t)$ in the laboratory frame and $F^*(t)$ in the rotating frame, are then

$$F(t) = \exp(i\omega_I t)F^*(t) \tag{11}$$

$$F^*(t) = \mathrm{Tr}\{\exp(-i\mathcal{H}_d^* t/\hbar)\rho(0)\exp(i\mathcal{H}_d^* t/\hbar)I_+\}/\mathrm{Tr}\{\rho(0)I_x\} \tag{12}$$

where $F(0) = F^*(0) = 1$. The real parts of $F(t)$ and $F^*(t)$ describe the depolarization of the component of the polarization in the x direction and the imaginary parts the depolarization of the component in the y direction in the laboratory and rotating frames, respectively. No approximations have been made in deriving these expressions.

The local field part $\mathcal{H}'_d$ of the dipolar Hamiltonian $\mathcal{H}_d$ in equation (1) is (Abragam 1961)

$$\mathcal{H}'_d = A_{IS} + A_{SS} = \sum_j a_j I_z S_{jz} + \frac{1}{2}\sum_{j,k}{}' a_{jk} S_{jz} S_{kz} \tag{13}$$

where the prime denotes the exclusion of the terms $j = k$ from the sum,

$$a_j = \gamma_I \gamma_S \hbar^2 (1 - 3\cos^2\theta_j)/r_j^3 \tag{14}$$

and $r_j = (r_j, \theta_j, \phi_j)$ is the vector from the I spin to the jth S spin with the polar angles defined relative to the direction of the magnetic field. The a_{jk} are the corresponding quantities for the jth and kth S spins.

Since $\mathcal{H}'^*_d = \mathcal{H}'_d$, equation (12) for $F^*(t)$ becomes the local field expression $F^*_L(t)$ given by

$$F^*_L(t) = \mathrm{Tr}\{\exp(-\mathrm{i}A_{IS}t/\hbar)\rho(0)\exp(\mathrm{i}A_{IS}t/\hbar)I_+\}/\mathrm{Tr}\{\rho(0)I_x\} \tag{15}$$

using the result that both A_{IS} and ρ_S commute with A_{SS} and $\rho_I(0)$. Equation (15) does not involve the term A_{SS} describing the local field interaction between S spins.

Writing the trace for the I spin in a Zeeman representation and using the result that $\langle m'|I_+|m\rangle$ is zero unless $m' = m + 1$, it follows that

$$F^*_L(t) = \mathrm{Tr}_S\left\{\rho_S \exp\left(\mathrm{i}\sum_j a_j S_{jz} t/\hbar\right)\right\} \Big/ \mathrm{Tr}_S\{\rho_S\}. \tag{16}$$

This expression is independent of $\rho_I(0)$ and therefore $F^*_L(t)$ is independent of the manner in which the initial non-equilibrium polarization of the I spin is established. For example, the local field transverse depolarization is the same for both an implanted muon and an I spin acted on by a pulse to rotate the polarization away from the z direction.

Writing the trace for each S spin in a Zeeman representation, $F^*_L(t)$ becomes

$$F^*_L(t) = \prod_j \sum_m g(m)\exp(\mathrm{i}a_j m t/\hbar) \tag{17}$$

where $g(m) = \langle m|\rho_{S_j}|m\rangle$ is the probability of the jth S spin having the Zeeman quantum number m. The Fourier transform

$$f(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \exp(-\mathrm{i}\omega t)F^*_L(t)\,\mathrm{d}t \tag{18}$$

is the normalized distribution of frequencies, relative to ω_I, of the precession of the I spin due to the statistical distribution of magnetic moment orientations of the S spins. The local field complex depolarization function $F^*_L(t)$ in the rotating frame is the Fourier transform of this frequency distribution (Cohen and Reif 1957, Cameron and Sholl 1993).

Equation (17) provides a simple expression to evaluate the local field depolarization function for any distribution $g(m)$ at each S spin site j. For example, the familiar result in the high-temperature limit (Lee *et al* 1967, Gurevich *et al* 1972),

$$F^*_L(t) = \prod_j \frac{\sin[(2S+1)a_j t/(2\hbar)]}{(2S+1)\sin[a_j t/(2\hbar)]} \tag{19}$$

is obtained by using $g(m) = 1/(2S+1)$ in equation (17). Another example is the classical case, again in the high-temperature limit, for which $g(m) = 1/(2S)$ and the summation over m is replaced by an integral. The result is then

$$F_L^*(t) = \prod_j \frac{\sin(Sa_j t/\hbar)}{Sa_j t/\hbar}. \tag{20}$$

The value of S in this case simply scales the time, unlike the quantum case described by equation (19).

The moments M_n of the frequency distribution $f(\omega)$, defined by

$$M_n = \int_{-\infty}^{\infty} f(\omega)\omega^n \, d\omega \tag{21}$$

are related to the derivatives of $F_L^*(t)$ by (Abragam 1961)

$$M_n = (-i)^n \left(\frac{d^n F_L^*(t)}{dt^n} \right)_{t=0}. \tag{22}$$

Using equation (22), the moments for the expression (16) for $F_L^*(t)$ are

$$M_n = \frac{1}{\hbar^n} \mathrm{Tr}_S \left\{ \rho_S \left(\sum_j a_j S_{jz} \right)^n \right\} \Big/ \mathrm{Tr}_S\{\rho_S\}. \tag{23}$$

Writing the trace in a Zeeman representation gives the first two moments as

$$M_1 = \frac{1}{\hbar} \sum_j a_j \sum_m m\langle m|\rho_{S_j}|m\rangle / \sum_m \langle m|\rho_{S_j}|m\rangle \tag{24}$$

$$M_2 = \frac{1}{\hbar^2} \left[\sum_j a_j^2 \left(\frac{\sum_m m^2 \langle m|\rho_{S_j}|m\rangle}{\sum_m \langle m|\rho_{S_j}|m\rangle} \right) + \sum_{j,k}{}' a_j a_k \left(\frac{\sum_m m\langle m|\rho_{S_j}|m\rangle}{\sum_m \langle m|\rho_{S_j}|m\rangle} \right)^2 \right]. \tag{25}$$

In the high-temperature limit for which $\rho_{S_j} = 1/(2S+1)$, these become the well known Van Vleck (1948) results, $M_1 = 0$ and $M_2 = [S(S+1)/(3\hbar^2)] \sum_j a_j^2$. Equations (24) and (25) can also be calculated for the classical, high temperature case for which $\rho_{S_j} = 1/(2S)$ and the summations over m are replaced by integrals between $-S$ and S. The results are then $M_1 = 0$ and $M_2 = [S^2/(3\hbar^2)] \sum_j a_j^2$.

For arbitrary temperatures, for which ρ_S is given by the Boltzmann density operator of equation (5), equation (24) is identical to the expression for M_1 given by Abragam and Goldman (1981, p 285) for a system consisting of one I spin and many S spins. Since Abragam and Goldman do not make the local field restriction it can be concluded that M_1 for this system will always be local field in nature.

The high-temperature expressions (19) and (20) are real expressions for $F_L^*(t)$ and have odd moments which are zero because $g(m)$ is an even function. If the polarization along the y axis in the rotating frame is initially zero, it remains zero. These statements are not true for arbitrary temperatures considered in the next section.

3. Results for arbitrary temperatures

The form of the local field depolarization function $F_L^*(t)$ when the high-temperature approximation is not made is obtained by substituting the density operator for the Boltzmann

distribution given by equation (5) into equation (17). The result is (Cameron 1992)

$$F_L^*(t) = \prod_j \frac{\sum_m \exp[(-\alpha_S + ia_j t/\hbar)m]}{\sum_m \exp(-\alpha_S m)}$$

$$= \prod_j \frac{[A_j(t) - iB_j(t)]\sinh(\alpha_S/2)}{[\cos(a_j t/\hbar) - \cosh(\alpha_S)]\sinh[(2S+1)\alpha_S/2]} \tag{26}$$

where $\alpha_S = \hbar\omega_S/(kT)$ and $A_j(t)$ and $B_j(t)$ are given by

$$A_j(t) = \cos[(S+1)a_j t/\hbar]\cosh(S\alpha_S) - \cos(Sa_j t/\hbar)\cosh[(S+1)\alpha_S] \tag{27}$$

$$B_j(t) = \sin[(S+1)a_j t/\hbar]\sinh(S\alpha_S) - \sin(Sa_j t/\hbar)\sinh[(S+1)\alpha_S]. \tag{28}$$

The result for the classical case under conditions of arbitrary temperature, for which the probability distribution of allowed orientations of the S spin to the z axis is given by a continuous Boltzmann function, may be evaluated in a similar way and the result is

$$F_L^*(t) = \prod_j \frac{[C_j(t) - iD_j(t)]\alpha_S}{[(a_j t/\hbar)^2 + \alpha_S^2]\sinh(S\alpha_S)} \tag{29}$$

where $C_j(t)$ and $D_j(t)$ are given by

$$C_j(t) = \alpha_S \cos(Sa_j t/\hbar)\sinh(S\alpha_S) + (a_j t/\hbar)\sin(Sa_j t/\hbar)\cosh(S\alpha_S) \tag{30}$$

$$D_j(t) = \alpha_S \sin(Sa_j t/\hbar)\cosh(S\alpha_S) - (a_j t/\hbar)\cos(Sa_j t/\hbar)\sinh(S\alpha_S). \tag{31}$$

The expressions (26) and (29) for $F_L^*(t)$ are complex and, as shown below, M_1 is non-zero because the probability $g(m)$ of an S spin being in the Zeeman state m is not an even function. The polarization along the y axis in the rotating frame is non-zero, except at time zero, and the frequency distribution $f(\omega)$ is not symmetric. A complete picture of the depolarization function in the rotating frame can be obtained by considering functions $G(t)$ and $\phi(t)$ defined by

$$F_L^*(t) = G(t)\exp(i\phi(t)) \tag{32}$$

where $G(t)$ is the magnitude of the complex polarization and $\phi(t)$ describes its motion with respect to the rotating frame. In the high-temperature case, the depolarization function measured in the laboratory frame is a decaying, oscillatory function with oscillations of constant frequency ω_I whereas, if the high-temperature approximation is not made, the amplitude function describing the decay will differ from the high-temperature function and the presence of $\phi(t)$ indicates that there is a time-dependent phase shift of the sinusoidal oscillations.

In the limit $\alpha_S \to 0$, equations (26) and (29) for $F_L^*(t)$ reduce to their high temperature counterparts, equations (19) and (20). A limiting form of both equations (26) and (29) also exists for very low temperatures (or very high fields) corresponding to $\alpha_S \to \infty$. In this limit it can be shown that $G(t) \to 1$ and, by considering the form of $\mathrm{Re}\{F_L^*(t)\} = G(t)\cos\phi(t)$, that $\phi(t) \to S\sum_j a_j t/\hbar$. The form of $F_L^*(t)$ as $\alpha_S \to \infty$ is then

$$F_L^*(t) \to \exp\left(iS\sum_j a_j t/\hbar\right). \tag{33}$$

The first and second local field moments M_1 and M_2 can be obtained either by direct evaluation of the expressions (24) and (25) using the Boltzmann density operator of equation (5) or by expanding equations (26) and (29) as a series in t and using equation (22). The

results for both the quantum case described by equation (26) and for the classical case described by equation (29) may be expressed in the form

$$M_1 = \frac{Q_1}{\hbar}\sum_j a_j \tag{34}$$

$$M_2 = \frac{1}{\hbar^2}\left(Q_2\sum_j a_j^2 + Q_1^2{\sum_{j,k}}' a_j a_k\right) = \frac{1}{\hbar^2}(Q_2 - Q_1^2)\sum_j a_j^2 + M_1^2 \tag{35}$$

where Q_1 and Q_2 are functions of S and α_S. For the quantum case Q_1 and Q_2 are given by

$$Q_1 = \frac{S\sinh[(S+1)\alpha_S] - (S+1)\sinh(S\alpha_S)}{\cosh(S\alpha_S) - \cosh[(S+1)\alpha_S]} \tag{36}$$

$$Q_2 = \frac{(S+1)^2\cosh(S\alpha_S) - S^2\cosh[(S+1)\alpha_S]}{\cosh(S\alpha_S) - \cosh[(S+1)\alpha_S]} - \frac{1}{1-\cosh(\alpha_S)}. \tag{37}$$

These have the limiting values 0 and $S(S+1)/3$ as $\alpha_S \to 0$, and $-S$ and S^2 as $\alpha_S \to \infty$, respectively. For the classical case the results are

$$Q_1 = [1 - S\alpha_S\coth(S\alpha_S)]/\alpha_S \tag{38}$$

$$Q_2 = \{2[1 - S\alpha_S\coth(S\alpha_S)] + (S\alpha_S)^2\}/\alpha_S^2 \tag{39}$$

which have the limiting values 0 and $S^2/3$ as $\alpha_S \to 0$, and $-S$ and S^2 as $\alpha_S \to \infty$, respectively. In the limit $\alpha_S \to 0$, the expressions (34) and (35) for M_1 and M_2 reduce to the high-temperature expressions. As $\alpha_S \to \infty$, $M_2 = M_1^2$ for both of the above cases and therefore, from equation (33), $F_L^*(t) \to \exp(-iM_1 t)$.

The above discussion shows that both for sufficiently small and sufficiently large α_S, $F_L^*(t)$, M_1 and M_2 are independent of the temperature and the magnitude of the external magnetic field, although the assumption that the external field is sufficiently large remains. It is of interest to examine the range of values of α_S between the $\alpha_S \to 0$ (high-temperature) and $\alpha_S \to \infty$ (low-temperature or very-high-field) limits for which effects which are dependent on temperature (and on the strength of the external field) could be observed. An indication of this region can be obtained by calculating Q_1 and Q_2 as functions of α_S. The quantum and classical forms of Q_1 and Q_2 as given by equations (36)–(39) are shown in figure 1 for $S = \frac{1}{2}$, $\frac{3}{2}$ and $\frac{5}{2}$. The greatest dependence on α_S occurs for larger values of S and for $S = \frac{1}{2}$ there is no α_S dependence of Q_2 for the quantum case. The figure suggests that the range of values of α_S over which temperature-dependent effects could be observed is ~ 0.1–5. In the case of copper ($S = \frac{3}{2}$), for example, this corresponds approximately to B/T between 200 and 10^4 T K^{-1}.

The summation $\sum_j a_j \propto \sum_j (1 - 3\cos^2\theta_j)/r_j^3$ is conditionally convergent and for this reason the values of the moments, which contain terms such as $\sum_j a_j$ and ${\sum_{j,k}}' a_j a_k$, can depend on the shape of the sample. Terms such as $\sum_j a_j^2$ are absolutely convergent and are independent of the sample shape (Abragam and Goldman 1981, ch 5). Although $F_L^*(t)$ can be expanded as an infinite series in t with coefficients depending on the moments M_n and may therefore be dependent on the sample shape, it can be shown that the amplitude $G(t)$ is independent of the sample shape. This result follows from expanding $G^2(t) = |F_L^*(t)|^2$ as a power series in t and noting that the coefficients involve only absolutely convergent summations of the form $\sum_j (a_j^2)^n$. The moments and the phase function $\phi(t)$ are, however, shape dependent. This shape dependence still exists in the very-low-temperature (or very-high-field) limit because conditionally convergent summations are present in the $\alpha_S \to \infty$

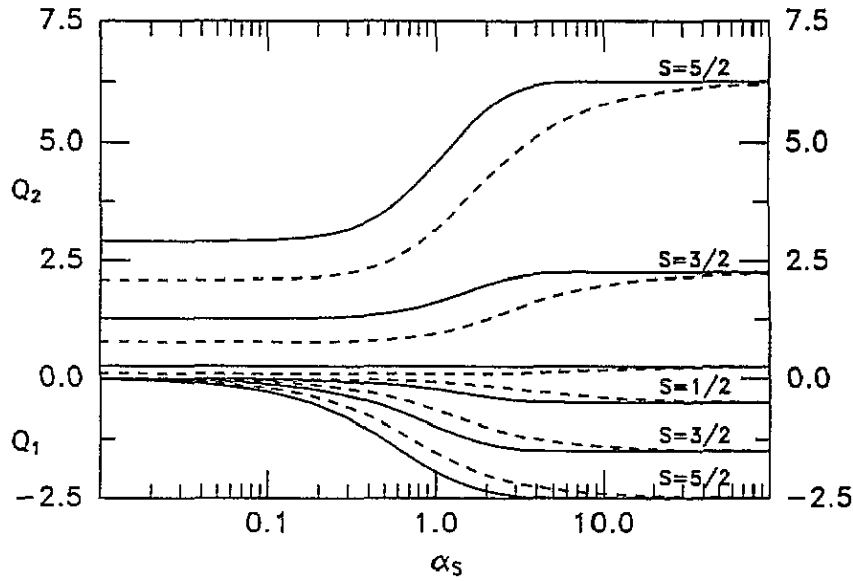

Figure 1. The variation of the parameters Q_1 and Q_2, as defined by equations (36)–(39), as a function of $\alpha_S = \hbar\omega_S/(kT)$ for $S = \frac{1}{2}, \frac{3}{2}$ and $\frac{5}{2}$. The values of Q_2 are positive and the values of Q_1 are negative. The solid curves are the results for the quantum case and the dashed curves are the results for the classical case.

forms of these quantities, even though no shape dependence occurs in the high-temperature limit $\alpha_S \to 0$.

Abragam and Goldman (1981) have discussed the shape dependence of M_1. The summation $\sum_j a_j$ for the system of one I spin and many crystallographically equivalent S spins is zero for a spherical sample, and for an ellipsoid does not depend on the position of the I spin within the ellipsoid. For a cubic structure of S spins, the summation taken over a cube is also zero. It is then possible, as discussed by Abragam and Goldman, to treat the problem of calculating $\sum_j a_j$ by separating the sample into an internal region and an external region, choosing the internal region to be a sphere centred on the I spin so that the contribution to $\sum_j a_j$ from the spherical internal region is zero and the external contribution then gives the value of $\sum_j a_j$. The result will converge for a particular shape but the convergence is very slow and the converged value will depend on the shape chosen. The convergence of $F_L^*(t)$, M_1 and M_2 under arbitrary temperature conditions is therefore quite slow compared to the rate of convergence of these quantities in the high-temperature case.

4. Examples

The transverse depolarization functions $F_L(t)$ in the laboratory frame and $F_L^*(t)$ in the rotating frame are related by equation (11) and $F_L^*(t)$ is specified by its magnitude $G(t)$ and phase factor $\phi(t)$ as defined in equation (32). To illustrate the type of behaviour that can occur, some results of calculations of the amplitude function $G(t)$ and the phase function $\phi(t)$ are presented in figures 2, 3 and 4. These results are for the I spin at an octahedral site in copper (FCC lattice, $S = \frac{3}{2}$, $\gamma_S = 2\pi \times 1.1533 \times 10^7$ Hz T^{-1}) for which the parameter $\alpha_S = \hbar\omega_S/(kT) = (-\hbar\gamma_S/k)(B/T)$ has magnitude $(5.535 \times 10^{-4}$ K $T^{-1})(B/T)$. The sign of α_S is determined by the sign of γ_S. The function $G(t)$ and the

second moment M_2 are even functions of α_S while the function $\phi(t)$ and the first moment M_1 are odd functions of α_S. The results in figures 2–5 are shown for positive α_S. The units of time are chosen, by considering the high-temperature expression for M_2, to be α where $\alpha^2 = 3a^6/[\gamma_I^2\gamma_S^2 S(S+1)\hbar^2]$ and a is the lattice parameter. For muons in copper, $\alpha = 64.9$ μs.

In the high-temperature limit the amplitude function $G(t)$ is of Gaussian form at short times but it can show significant deviation from a Gaussian at longer times (Cameron and Sholl 1991). This is also the case for $G(t)$ at low temperatures. The major effect as B/T increases is that $G(t)$ decays more slowly as shown in figure 2 for two directions of the magnetic field. The limiting form as $B/T \to \infty$ is $G(t) = 1$ as discussed in the previous section. These results do not depend on the shape of the sample.

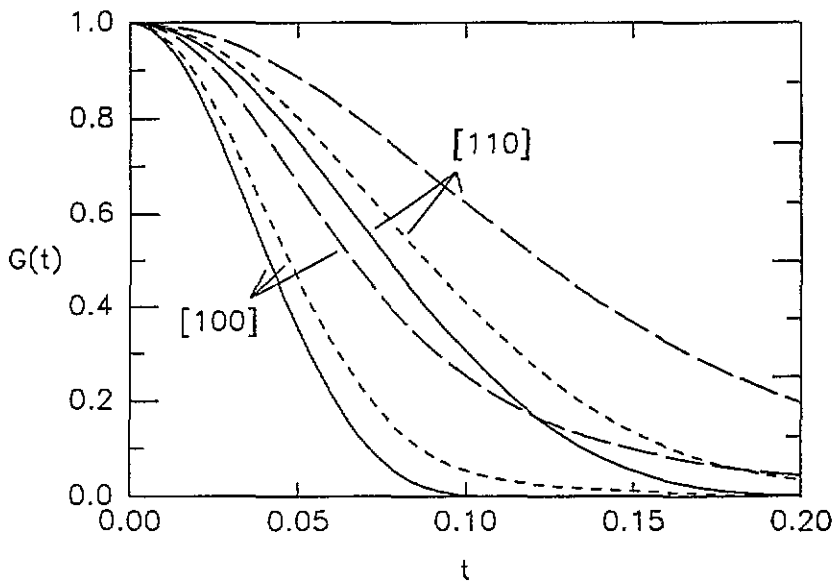

Figure 2. The amplitude function $G(t)$ as defined by equation (32) for the I spin at an octahedral site in a copper crystal and the magnetic field in the [100] and [110] directions. The short-dashed curves are the results for $B/T = 1000$ T K^{-1} ($\alpha_S = 0.5535$) and the long-dashed curves are for $B/T = 2000$ T K^{-1} ($\alpha_S = 1.107$) and, for comparison, the high-temperature ($\alpha_S \to 0$) result is shown for each field direction by the solid curves. The amplitude function is independent of the shape of the sample. The time t is in the dimensionless units defined in section 4.

The phase function $\phi(t)$ is shown in figures 3 and 4 for the same system. This function is zero in the high-temperature limit but is non-zero at low temperatures and depends on the sample shape. Figure 3 shows some results for a spherical sample and figure 4 for a sphere and an ellipsoid. The ellipsoid has axes in the ratio 1:2:3 and two different orientations of it in the magnetic field are considered. One case, denoted 'e_1', has the shortest and longest axes along the x and z directions, respectively, and the other case, denoted 'e_2', has the shortest and longest axes in the z and x directions, respectively. It can be seen that the phase changes in a non-linear manner as a function of time in all cases and shows significant dependence on sample shape. The phase shift is not appreciable at short times and data at sufficiently long times would be required to observe the effect.

The moments M_1 and M_2 are also shown for the same system in figure 5. These are consistent with the results in figures 2, 3 and 4 but show the dependence on α_S across the

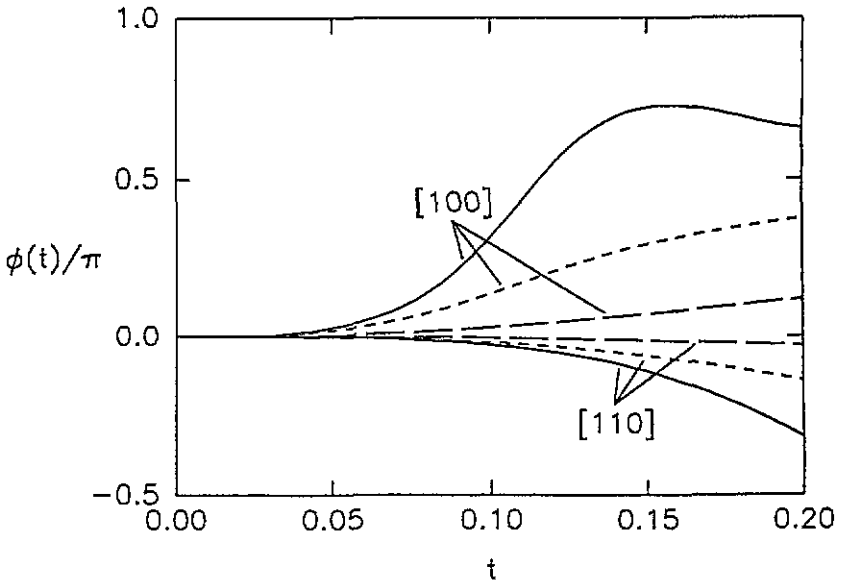

Figure 3. The phase function $\phi(t)$ as defined by equation (32) for the I spin at an octahedral site in a spherical sample of crystalline copper and the magnetic field in the [100] and [110] directions. The solid curves are the results for $B/T = 1000$ T K^{-1} ($\alpha_S = 0.5535$), the short-dashed curves are for $B/T = 2000$ T K^{-1} ($\alpha_S = 1.107$) and the long-dashed curves for $B/T = 4000$ T K^{-1} ($\alpha_S = 2.214$). In the high-temperature limit ($\alpha_S \to 0$), $\phi(t) = 0$. The curves illustrate the dependence of the phase function on magnetic field and temperature for a given sample shape. The time t is in the dimensionless units defined in section 4.

region where the behaviour changes from the high-temperature limit to the low-temperature limit. As in figure 1 it is again apparent from these results that the transition regime is $\alpha_S \sim 0.1 - 5$.

In the high-temperature limit M_2 and $G(t)$ are independent of the sample shape, $G(t) \simeq 1 - M_2 t^2/2$ for small times and $M_1 = 0$. At low temperatures $M_1 (\neq 0)$ and M_2 are shape dependent but $G(t)$ is not. The small-t behaviour of $G(t)$ in this case is $G(t) \simeq |1 + iM_1 t - M_2 t^2/2| \simeq 1 - (M_2 - M_1^2)t^2/2$ and $(M_2 - M_1^2)$ is shape independent (see equation (35)) even though M_1 and M_2 are shape dependent.

5. Discussion

The transverse depolarization function is the same for the FID of dilute nuclear spins in a crystal and for polarized muons implanted in metals after correction for the radioactive decay of the muon. This work describes the local field depolarization function for such systems, which is valid only for short times in the absence of diffusion and in the presence of a sufficiently large external magnetic field. There are few models for which the depolarization function can be found exactly and the low-temperature, high-field model considered here provides an additional exact result. The dependence of the function on magnetic field B and temperature T arises through the parameter $\alpha_S = (-\hbar\gamma_S/k)(B/T)$. In the high-temperature limit, $\alpha_S \lesssim 0.1$, the local field depolarization function in the laboratory frame, $F_L(t)$, is a decaying function $G(t)$, which is not necessarily of Gaussian form, modulated by $\cos(\omega_I t)$, where $\omega_I = -\gamma_I B$. The function $G(t)$ is independent of B, T and sample shape.

At larger values of B/T, in the range $0.1 \lesssim \alpha_S \lesssim 5$, the function $G(t)$ decays more

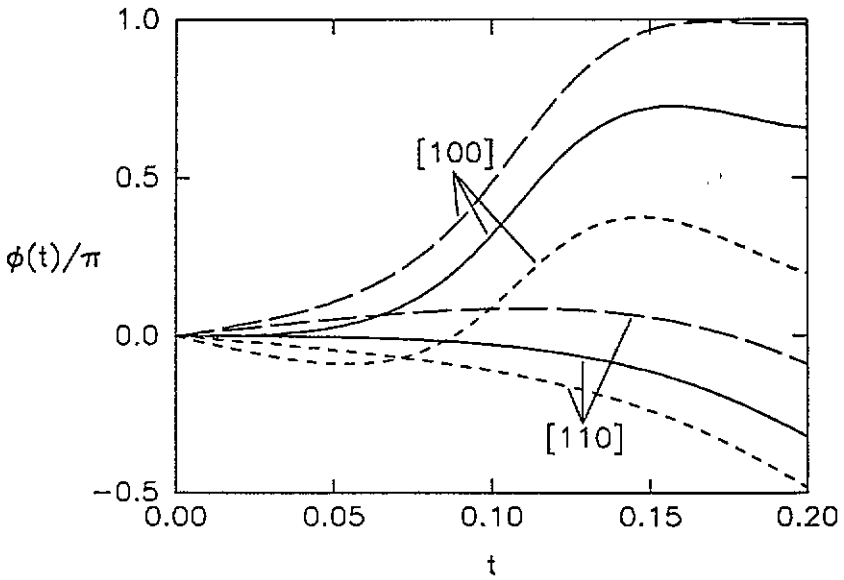

Figure 4. The phase function $\phi(t)$ as defined by equation (32) for the I spin at an octahedral site in a copper crystal and the magnetic field in the [100] and [110] directions. Results are shown for $B/T = 1000$ T K^{-1} ($\alpha_S = 0.5535$) and for the three sample shapes described in the text. The solid curves are the results for the sphere and the short-dashed and long-dashed curves are those for the ellipsoids 'e_1' and 'e_2', respectively. The curves illustrate the dependence of the phase function on the sample shape for a given magnetic field and temperature. The time t is in the dimensionless units defined in section 4.

slowly and depends on B/T but is still independent of sample shape. The oscillating modulation of this function becomes $\cos[\omega_I t + \phi(t)]$, where $\phi(t)$ is dependent on B/T and sample shape, for the component of the polarization in the x direction (direction of the initial polarization). The effect of the low temperatures could therefore be observed in the dependence of $G(t)$ on B/T and also in the time-dependent phase shifts of the oscillations.

At still larger values of B/T, for which $\alpha_S \gtrsim 5$, the function $G(t) \rightarrow 1$ and the modulating function becomes $\cos[(\omega_I + M_1)t]$. The value of the first moment M_1 in this limit is independent of B/T but does depend on the shape of the sample.

The results show that the high-temperature approximation is valid unless extreme conditions of low temperature and high magnetic fields (by current standards) exist. In cases where the high-temperature approximation is not valid, there are new features in the depolarization function: a time-dependent phase function $\phi(t)$, shape dependence of some parameters and non-zero values of the first moment M_1.

Expressions have been derived for the depolarization function in all the above cases retaining only the local field (A) terms in the dipolar Hamiltonian for both the I–S and S–S interactions. The flip-flop (B) terms are correctly omitted for the I–S interactions since they are non-secular but they should be included in the indirect effect of the S–S interactions on the depolarization of the I spin. The effect of these terms will only be significant at long times since it can be shown that they do not contribute to the moments M_1 and M_2 and therefore only contribute to order t^3 in a Taylor expansion of the depolarization function. Further, the contribution of the B terms to higher moments is of order γ_S/γ_I so their effect will be small if $\gamma_S \ll \gamma_I$. This will be the case for muons because of the large muon magnetic moment.

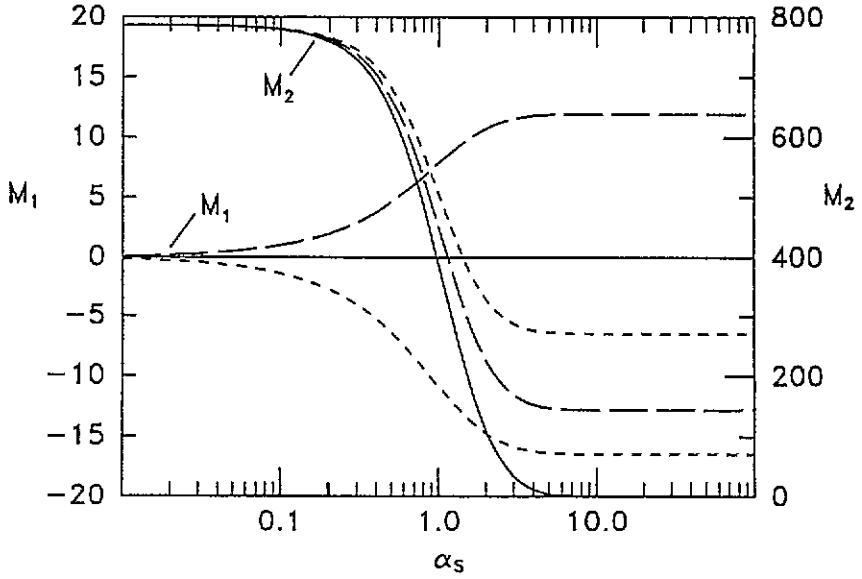

Figure 5. The first and second moments M_1 (in units of $1/\alpha$) and M_2 (in units of $1/\alpha^2$) as functions of α_S for an I spin occupying an octahedral site in a copper crystal. The results are for the magnetic field in the [100] direction and for the three sample shapes described in the text. The solid curves are the results for the sphere and the short-dashed and long-dashed curves are those for the ellipsoids 'e_1' and 'e_2', respectively.

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