Quantum calculation of the muon depolarization function: effect of spin dynamics in nuclear dipole systems

1992 J. Phys.: Condens. Matter 4 4533

(http://iopscience.iop.org/0953-8984/4/18/020)

J. Phys.: Condens. Matter 4 (1992) 4533–4556. Printed in the UK

Quantum calculation of the muon depolarization function: effect of spin dynamics in nuclear dipole systems

P Dalmas de Réotier† and A Yaouanc
Centre d'Etudes Nucléaires, DRFMC/SPSMS/LIH 85X, F-38041 Grenoble Cédex, France

Received 17 October 1991

Abstract. We have derived formal expressions for the positive muon depolarization functions using an iterative procedure both for the longitudinal and transverse experimental geometries. They are valid at least in the motional narrowing limit and at small time. We have used our formal expressions to study the depolarization functions for a muon diffusing in a lattice of nuclear dipoles neglecting the possibility of the muon returning to its original localization site. This work indicates that the effect of the spin dynamics on the depolarization functions is important at low fields for both geometries. In the case of the longitudinal geometry we have shown that our second-order iteration formula and the strong collision model with the numerically exact static function give the same result when the muon jumping rate is large enough. This has two important consequences: (i) when the muon jumping rate is sufficiently large it is possible to calculate the whole depolarization function with a modest numerical effort using our iterative formula; and (ii) the fact that the two methods give the same result in the fast diffusing limit shows that the strong collision model with the exact static function contains all the dynamics; the spin–lattice relaxation (T_1 process) is included.

1. Introduction

The diffusion of a light interstitial particle such as a positive muon, μ^+, is a fascinating subject in solid state physics. The quantum effects are important even at relatively high temperature (100 K). For aluminium, in the low-temperature region, the μ^+ diffusion constant is much larger than expected if only the lowest order of the muon–electron interaction is taken into account (Kehr *et al* 1982). However, as shown by Kondo (1986) and Yamada (1984), if the effect of the higher-order terms of this interaction is included, the agreement between theory and experiment is much better.

The information obtained on the μ^+ diffusion properties from positive muon spectroscopy (μSR) is extracted from the analysis of the measured μSR depolarization functions, $P_\alpha(t)$ (α refers to the direction of the measurement; see later), and is model dependent; see Chappert and Grynszpan (1984), Schenck (1985) and Chappert and Yaouanc (1986). Here we will only consider compounds for which the electronic magnetic moments have no effect on $P_\alpha(t)$ as most of the experimental work on diffusion has been performed on such compounds. In this case the depolarization of the μ^+ spin is due to the magnetic field produced at the μ^+ site by the nuclear dipoles of the lattice.

† Current address: Hahn–Meitner-Institut Berlin, D-1000 Berlin 39, Federal Republic of Germany.

0953-8984/92/184533+24$04.50

Usually a dynamical $P_\alpha(t)$ function is computed in terms of a depolarization function for a static μ^+, $P_\alpha^{(0)}(t)$, using the strong collision model (Kehr *et al* 1978, Hayano *et al* 1979). A common practice for the calculation of $P_\alpha^{(0)}(t)$ is to express it as an average of the μ^+ spin over the distribution function of the magnetic field at the μ^+ site. Whereas the μ^+ spin dynamics is described by the Larmor equation, the nuclear spin dynamics due to the magnetic field created by the μ^+ at the nuclear sites is neglected. The parameter (only one parameter for a μ^+ site of cubic symmetry) of the distribution function is estimated from the second moment of the interaction Hamiltonian between the μ^+ magnetic moment and the magnetic moment of the lattice nuclei (Van Vleck's method). Using quantum mechanics it has been shown numerically that this method does not describe $P_\alpha^{(0)}(t)$ in detail (Celio and Meier 1983, Celio 1986). It can even lead to a wrong estimate of the μ^+ diffusion constant. In this paper we present an iterative method to compute the $P_\alpha(t)$ functions which is valid at least in the motional narrowing limit. Although it is not only restricted to the case of depolarization due to nuclear dipole moments, in this paper we will only consider this source of depolarization. We will show for the longitudinal geometry, in the case of a μ^+ diffusing sufficiently rapidly, that this method leads to a $P_z(t)$ function which is the same as the one computed from the strong collision model with $P_z^{(0)}(t)$ calculated from quantum mechanics. Therefore the strong collision model with this $P_z^{(0)}(t)$ function contains all the spin dynamics. On the other hand the method which uses the strong collision model with $P_\alpha^{(0)}(t)$ deduced from the static distribution function method gives a bad approximation of $P_\alpha(t)$. Our work shows that it is important for a reliable description of $P_\alpha(t)$ in the two experimental geometries to take the spin dynamics into account.

Recently experiments designed to study in great detail the μ^+ diffusion properties have been reported for Al (Kadono *et al* 1990a) and Cu (Luke *et al* 1991). Kadono *et al* (1990b) have discussed results obtained on the weak ferromagnet MnSi for which the depolarization due to the ^{55}Mn nuclei is not negligible and should, therefore, be described properly if one wants to extract reliable information on the electronic spin dynamics in this compound. In all these cases our theoretical results should help to give a consistent picture of the μSR data.

The organization of this paper is as follows. In section 2 we briefly review the method of computation of the depolarization functions using the stochastic theory and discuss its validity. The formal quantum expressions for the functions, valid at least in the motional narrowing limit, are established in section 3. No hypothesis concerning the source of depolarization is made. In section 4 we discuss in some detail the case of a μ^+ diffusing in a lattice of nuclear dipoles. A comparison of the results from the second-order iteration with the strong collision model for the two types of static depolarization functions (classical and quantum mechanics) is made. Section 5 contains a summary, comments on possible extensions and the conclusions.

2. The depolarization functions from the stochastic theory

The stochastic theory of dynamical processes was first used to compute $P_\alpha(t)$ for the transverse and longitudinal experimental geometries by Kehr *et al* (1978) and Hayano *et al* (1979) respectively. Here we present a short review of their results to define the parameters and discuss the approximations made within this theory. The magnetic

field dynamics, which is due, in this theory, to the μ^+ diffusion and/or the interaction between the nuclear dipoles which are at the origin of the field distribution at the μ^+ site, is described by the 'strong collision' model (Kubo 1954). For clarity we will use the language of diffusion. A characteristic of this model is that the depolarization function for a static μ^+, $P_\alpha^{(0)}(t)$, is needed. This last function can be computed directly for simple cases using quantum mechanics (Celio 1986). But because of numerical difficulties, it is always useful to use a classical mechanics method with a static distribution. We first describe $P_\alpha^{(0)}(t)$ obtained from this method and then consider the case of a diffusing μ^+.

2.1. *Depolarization functions for a static muon*

By definition at the initial time, $t = 0$, the μ^+ spin is directed along the z-axis. The polarization at a later time in the α direction is given by

$$P_\alpha^{(0)}(t) = \int \frac{S_\alpha(t)}{S} D(\boldsymbol{B})\,\mathrm{d}\boldsymbol{B} \tag{1}$$

where S is the muon spin ($S = \frac{1}{2}$), $S_\alpha(t)$ its projection on the α direction at time t and $D(\boldsymbol{B})$ the static distribution function of the magnetic field $\boldsymbol{B}$ at the μ^+ site. The exponent (0) specifies that we are dealing with a depolarization function for a static μ^+. Its significance will be clarified when discussing the effect of diffusion. In a so-called transverse measurement, an external magnetic field B_{ext} is applied perpendicular to the z-axis (the x-axis for example) and $P_z^{(0)}(t)$ (or $P_y^{(0)}(t)$) is recorded. In a longitudinal experiment a field can be applied along the z-axis during the measurement of $P_z^{(0)}(t)$ but sometimes no field is applied.

For simplicity we suppose that the field distribution at the μ^+ site due to the lattice magnetic moments is isotropic (the second moments along the x-, y- and z-axes are the same). This is the case for a μ^+ in metallic copper and aluminium. A generalization for an anisotropic distribution can easily be made (Dalmas de Réotier 1990). In addition we will hypothesise that the distribution is Gaussian.

With these hypotheses, when no magnetic field is applied, the field distribution function is given by

$$D(\boldsymbol{B})\,\mathrm{d}\boldsymbol{B} = \left(\frac{\gamma_\mu}{\sqrt{2\pi}\Delta}\right)^3 \exp\left(-\frac{\gamma_\mu^2 B^2}{2\Delta^2}\right) B^2 \sin\theta\,\mathrm{d}B\,\mathrm{d}\theta\,\mathrm{d}\varphi \tag{2}$$

where γ_μ is the μ^+ gyromagnetic ratio, Δ^2/γ_μ^2 the second moment of the components of the magnetic field distribution, θ and φ the polar and azimuthal angles of the magnetic field vector at the μ^+ site. The $S_z(t)$ component of the μ^+ spin is obtained from the solution of the Larmor equation:

$$S_z(t) = S\left[\cos^2\theta + \sin^2\theta\cos\left(\gamma_\mu B t\right)\right]. \tag{3}$$

The Kubo–Toyabe function (Kubo and Toyabe 1966) is simply derived using equations (1), (2) and (3):

$$P_z^{(0)}(t) = \frac{1}{3} + \frac{2}{3}\left(1 - \Delta^2 t^2\right)\exp\left(\frac{-\Delta^2 t^2}{2}\right). \tag{4}$$

For this experimental geometry $P_x^{(0)}(t) = 0$ and $P_y^{(0)}(t) = 0$. Hayano *et al* (1979) give an expression for $P_z^{(0)}(t)$ in the case of an external field applied along the z-axis.

For the transverse geometry the spherical coordinates are not practical. In Cartesian coordinates, with an external field B_{ext} applied along the x-axis, the distribution can be written as

$$D(B)\,\mathrm{d}B = \left(\frac{\gamma_\mu}{\sqrt{2\pi}\Delta}\right)^3 \exp\left(-\frac{\gamma_\mu^2\,(B_{\text{ext}} - B_x)^2}{2\Delta^2}\right) \exp\left(-\frac{\gamma_\mu^2 B_y^2}{2\Delta^2}\right) \times \exp\left(-\frac{\gamma_\mu^2 B_z^2}{2\Delta^2}\right) \mathrm{d}B_x\,\mathrm{d}B_y\,\mathrm{d}B_z. \tag{5}$$

A closed form for $P_z^{(0)}(t)$ can be given if the intensity of B_{ext} is sufficiently large, $B_{\text{ext}} \gg \Delta/\gamma_\mu$. Then as the resultant magnetic field at the μ^+ site is, to a good approximation, directed only along the x-axis, the $S_x(t)$ component of the μ^+ spin deduced from the Larmor equation is

$$S_x(t) = S\cos\left(\gamma_\mu B_x t\right). \tag{6}$$

In this approximation the transverse depolarization function computed using equations (1), (5) and (6) is given by

$$P_z^{(0)}(t) = \exp\left(-\frac{\Delta^2 t^2}{2}\right)\cos\left(\gamma_\mu B_{\text{ext}} t\right). \tag{7}$$

The validity of the approximation made to derive equation (7), $B_{\text{ext}} \gg \Delta/\gamma_\mu$, has been investigated by comparing $P_z^{(0)}(t)$ integrated numerically with the function given in equation (7). The numerical integration is defined by equation (1) with $S_x(t)$ deduced from the Larmor equation (equation (3) in Cartesian coordinates) and $D(B)$ given by equation (5). Up to now the maximum measured Δ value for a nuclear dipole lattice in the transverse field geometry seems to be 0.40 MHz (Schenck 1985), i.e. $\Delta/\gamma_\mu = 4.7$ G. In practice the smallest transverse external magnetic field is about 10 G. This gives $B_{\text{ext}} = 2.1(\Delta/\gamma_\mu)$. In figure 1 we have drawn the two functions calculated with $B_{\text{ext}} = 5(\Delta/\gamma_\mu)$ and $B_{\text{ext}} = 2(\Delta/\gamma_\mu)$ respectively. The two plots show that while the analytical formula (equation (7)) is still a fair approximation at $B_{\text{ext}} = 5(\Delta/\gamma_\mu)$, it completely breaks down at lower fields. This is not surprising because at zero field $P_z^{(0)}(t)$ is given by the Kubo–Toyabe function.

Generally the second moment of the field distribution for the two experimental geometries is computed according to Van Vleck. This means that for the longitudinal and transverse case Δ^2 is taken as

$$\frac{2}{d\hbar^2}\,\mathrm{Tr}\left\{\left[\mathcal{H}'_{\text{dip}}, S_z\right]\left[S_z, \mathcal{H}'_{\text{dip}}\right]\right\} \qquad \text{and} \qquad \frac{1}{d\hbar^2}\,\mathrm{Tr}\left\{\left[\mathcal{H}'_{\text{dip}}, S_x\right]\left[S_x, \mathcal{H}'_{\text{dip}}\right]\right\}$$

respectively. $\mathcal{H}'_{\text{dip}}$ is the secular part of the dipolar Hamiltonian which describes the interaction between the μ^+ magnetic moment and the lattice nuclear magnetic moments. d is the dimension of the Hilbert space of the μ^+ sample system. The difference of a factor of two is related to the fact that in the longitudinal geometry the depolarization is produced by the two components of the field distribution perpendicular to the z-axis whereas for the transverse case only the component parallel to the direction of the applied field is responsible for the depolarization. In sections 4.1 and 4.2 we will show that Δ^2 can be obtained directly as a result of the iteration computation. Then the validity of the secular approximation can be gauged.

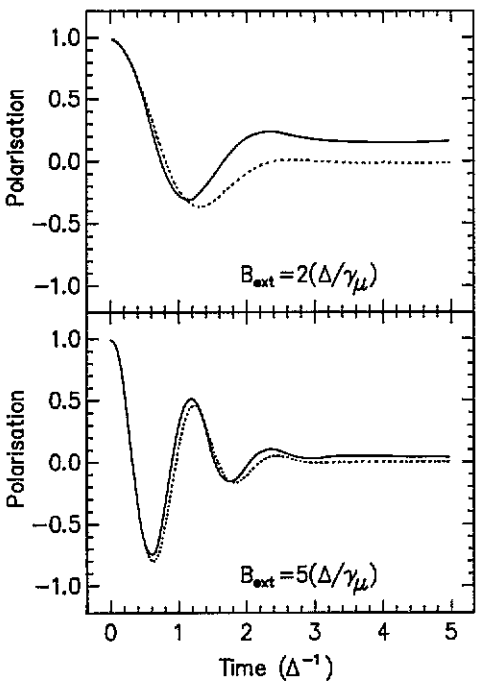

Figure 1. Comparison between two forms of the static transverse depolarization function deduced from the distribution method. For each figure, the full curve presents the result of the numerical integration (equations (1), (3) and (5)) and the dotted curve the approximate analytical function given in equation (7); see main text.

2.2. *Depolarization functions for a diffusing muon*

We now consider the case where the field distribution at the μ^+ is time-dependent. Following Kehr *et al* (1978) we describe it by the 'strong collision' model introduced by Kubo (1954) for nuclear magnetic resonance (NMR). It supposes that the local field at the μ^+ has a certain value for a given time interval. At a later time it takes a new value not correlated to the first one (Markov process). During its life in the sample, the μ^+ jumps l times and can see l different fields at its localization sites. Therefore the $P_\alpha(t)$ function which takes into account the μ^+ diffusion is given by

$$P_\alpha(t) = \sum_{l=0}^{+\infty} R_l(t)$$

where $R_l(t)$ is the product of the depolarization function in the case of l jumps by the probability that the μ^+ has jumped l times during the time interval $[0, t]$. For example we have $R_0(t) = P_\alpha^{(0)}(t)\exp(-\nu t)$ where ν is the jump frequency. The same model gives

$$R_1(t) = \left\langle \int_0^t \mathrm{d}t'\, P_{i\alpha}(t-t')\exp[-\nu(t-t')]\nu P_{j\alpha}(t')\exp(-\nu t') \right\rangle_{ij}$$

where $P_{i\alpha}(t)$ describes the evolution of the projection (given by the Larmor equation) of the μ^+ spin on the α direction in the time interval $[0, t]$. During this time the

μ^+ spin sees one field, B_i, which belongs to the field distribution. The hypothesis of the strong collision model allows the different terms of $R_1(t)$ to be decoupled and therefore to be written as

$$R_1(t) = \nu \int_0^t \mathrm{d}t' \, R_0(t-t') R_0(t').$$

Using the same method, an expression for $R_{l+1}(t)$ can be derived. It is then possible to deduce the following integral equation for $P_\alpha(t)$:

$$P_\alpha(t) = P_\alpha^{(0)}(t) \exp(-\nu t) + \nu \int_0^t \mathrm{d}t' \, P_\alpha(t-t') P_\alpha^{(0)}(t') \exp(-\nu t'). \tag{8}$$

The derivation of the equation for $P_\alpha(t)$ is independent of the shape of the $P_\alpha^{(0)}(t)$ function. It can be computed either from classical or quantum mechanics. This fact will be used in section 4. Therefore $P_\alpha(t)$ given by equation (8) may look quite general. But it should be realized that two physical hypotheses have been made. First we have neglected the possibility of the μ^+ returning to its original site after a jump. This is explicit from the jump frequency dependence we have taken, for example, for $R_0(t)$. The second hypothesis has been made when we have supposed that the fields seen by the μ^+ in the different sites are independent of each other. This approximation may not be valid if the μ^+ jumps between sites which share common neighbours.

Equation (8) has been derived using the language of the magnetic field distribution. Therefore one could think that it is not general because it may not take into account the whole spin dynamics (the spin–lattice relaxation in particular). This is not true if, for the computation of the static depolarization function, the effect of the spin dynamics is included. This fact is supported by the numerical comparison made in section 4 between the results of the strong collision model with the exact static depolarization function and our second-order iteration.

Using the Laplace transform of $P_\alpha(t)$, $P_\alpha(s)$, it is possible to study the asymptotic behaviour of $P_\alpha(t)$ valid when $\nu/\Delta \gg 1$. For the transverse geometry, when $B_{\mathrm{ext}} \gg \Delta/\gamma_\mu$, we get (we do not take into account the $\cos(\gamma_\mu B_{\mathrm{ext}} t)$ term because it is not modified by the μ^+ diffusion)

$$P_z(s) = \frac{1}{s}\left(1 + \frac{\Delta^2}{\nu^2}\right) - \frac{1}{s^2}\frac{\Delta^2}{\nu^2} - \frac{1}{s+\nu}\frac{\Delta^2}{\nu^2}.$$

The inverse Laplace transform of this latter function is

$$P_z(t) = 1 + \frac{\Delta^2}{\nu^2} - \frac{\Delta^2}{\nu} t - \frac{\Delta^2}{\nu} \exp(-\nu t).$$

These are the first two terms of the expansion of the Abragam formula

$$P_z(t) = \exp\left\{-\frac{\Delta^2}{\nu^2}[\cos(-\nu t) - 1 + \nu t]\right\} \cos(\gamma_\mu B_{\mathrm{ext}} t) \tag{9}$$

where we have included the $\cos(\gamma_\mu B_{\mathrm{ext}} t)$ for completeness. The Abragam formula is usually derived from a Gaussian–Markovian theory (Abragam 1961) whereas $P_\alpha(t)$

given by equation (8) is the result of a random-walk model. Kehr *et al* (1978) have shown numerically the remarkable result that $P_z(t)$ deduced from the Abragam formula and the integral equation with $P_z^{(0)}(t)$ given by equation (7) are very similar even when ν is small i.e. when the μ^+ is quasi-static.

A study of the asymptotic expansion of $P_z(t)$ in zero field by the same method shows that when the jump rate is sufficiently large we have

$$P_z(t) = \exp\left\{-2\frac{\Delta^2}{\nu^2}[\exp(-\nu t) - 1 + \nu t]\right\}. \tag{10}$$

This is the Abragam formula with the substitution $\Delta^2 \rightarrow 2\Delta^2$ and with $B_{ext} = 0$. The factor two is again related to the fact that two components of the magnetic field at the μ^+ site depolarize the μ^+ spin in the longitudinal geometry. This approximate form, which seems to have been overlooked up to now, is in fact excellent when the diffusion is sufficiently fast: the difference between the $P_z(t)$ functions calculated from equations (8) and (4) and the Abragam formula with the mentioned substitution is less than 1% if $\nu/\Delta > 3$. In the extreme motional narrowing limit the latter formula reduces to $\exp[-(2\Delta^2/\nu)t]$. We will see in the next section that the pre-asymptotic Abragam form (equation (10)) is very useful to understand the quantum mechanical results. In addition, when this form can be used, the computing time for $P_z(t)$ is notably reduced.

3. Formal quantum expressions of the depolarization functions obtained from iteration

In this section we first discuss the relation between $P_\alpha(t)$ and the μ^+ spin operators. We then derive, using a second-order iterative procedure, a general expression for $P_\alpha(t)$. In the next subsections this expression is used to deduce formulae for the depolarization function for the longitudinal and transverse geometries.

For practical reasons it is useful to change the reference frame. In fact, from now on, we will use two reference frames, one for each experimental geometry. In figure 2 we define these two coordinate systems.

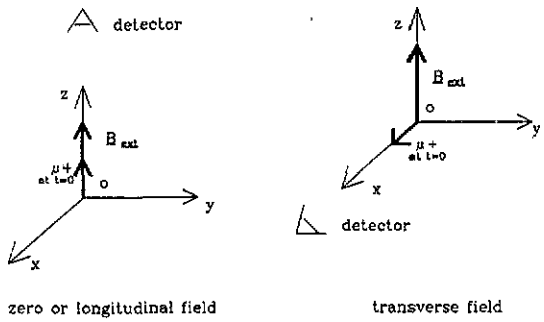

Figure 2. Reference frames used for the quantum computation (from section 3 on) of the depolarization functions in the longitudinal and transverse experimental geometries respectively.

3.1. Relation between $P_\alpha(t)$ and the μ^+ spin correlation function

From quantum mechanics we know that the depolarization function measured along the α direction, $P_\alpha(t)$, is the mean value of the time evolution of the Pauli operator σ_α:

$$P_\alpha(t) = \langle\psi|\sigma_\alpha(t)|\psi\rangle$$

where $|\psi\rangle$ is the wavefunction of the ensemble μ^+ sample at the initial time. Using the expansion of $|\psi\rangle$ on the Hilbert base of this ensemble, $|\psi\rangle = \sum_n C_n|n\rangle$, we can write

$$P_\alpha(t) = \sum_{n,m} C_m^* C_n \langle m|\sigma_\alpha(t)|n\rangle = \sum_n \langle n|\rho\sigma_\alpha(t)|n\rangle = \mathrm{Tr}\{\rho\sigma_\alpha(t)\}$$

where ρ is the density operator, $\rho_{n,m} = C_m^* C_n$. At the initial time the μ^+ and the sample are independent. Therefore $\rho = \rho_\mu\rho_s$ where ρ_μ and ρ_s are the density operators of the μ^+ and the sample respectively. As usual we have $\rho_s = \exp(-\beta\mathcal{H}_s)/\mathrm{Tr}\{\exp(-\beta\mathcal{H}_s)\}$ with $\beta = 1/k_B T$. $\mathcal{H}_s$ is the Hamiltonian operator of the sample, including the effects of the muon electric charge on its neighbours but excluding the effects of its spin. ρ_μ depends on the experimental geometry.

In a longitudinal experiment the μ^+ spin at the initial time is directed along the z-axis; see figure 2. This polarization can be described by the μ^+ density operator $\rho_\mu = \frac{1}{2}(\mathbf{1} + \sigma_z)$ where $\mathbf{1}$ is the identity operator. It follows that the depolarization function measured in this geometry is given by

$$P_z(t) = \mathrm{Tr}\{\rho_\mu\rho_s\sigma_z(t)\} = \mathrm{Tr}\{\rho_s\rho_\mu\sigma_z(t)\} = \tfrac{1}{2}\mathrm{Tr}\{\rho_s\sigma_z\sigma_z(t)\} \tag{11}$$

if $\mathrm{Tr}\{\rho_s\sigma_z(t)\} = 0$. This hypothesis is valid most of the time because $\mathrm{Tr}\{\rho_s\sigma_z(t)\}$ is proportional to the mean value of the μ^+ spin for an initially unpolarized μ^+ beam (in this case $\rho_\mu = \frac{1}{2} \times \mathbf{1}$ and $\mathrm{Tr}\{\rho\sigma_z(t)\} = \frac{1}{2}\mathrm{Tr}\{\rho_s\sigma_z(t)\}$. The intensity of the magnetic field usually applied is too small to modify the μ^+ beam polarization (a magnetic field of 1 T corresponds to a temperature of 6.5 mK).

Using the same method, it can be shown that the transverse depolarization function is given by

$$P_x(t) = \tfrac{1}{2}\mathrm{Tr}\{\rho_s\sigma_x\sigma_x(t)\}. \tag{12}$$

Equations (11) and (12) show that, for usual experimental conditions, $P_\alpha(t)$ is equal to the μ^+ spin correlation function.

3.2. General expression for $P_\alpha(t)$ obtained from iteration

The previous equations indicate that to compute $P_\alpha(t)$ we need an expression for $\sigma_\alpha(t)$. For most practical cases, $\sigma_\alpha(t)$ can only be calculated approximately. In this paper we use an iterative procedure. We suppose that the Hamiltonian $\mathcal{H}$ which describes the μ^+ sample system can be split into two parts: $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$. In this section we do not have to specify how the splitting is made. We use the intermediate representation and define the following operators in this representation:

$$\sigma_\alpha^*(t) = \exp(-i\mathcal{H}_0 t/\hbar)\,\sigma_\alpha(t)\exp(i\mathcal{H}_0 t/\hbar) \tag{13}$$

$$\mathcal{H}_1^*(t) = \exp(-i\mathcal{H}_0 t/\hbar)\,\mathcal{H}_1\exp(i\mathcal{H}_0 t/\hbar). \tag{14}$$

Using the Heisenberg equation which describes the time evolution of $\sigma_\alpha(t)$ and equations (13) and (14) we get

$$\frac{d\sigma_\alpha^*(t)}{dt} = \frac{i}{\hbar}[\mathcal{H}_1^*(t), \sigma_\alpha^*(t)].$$

The solution of this equation can be written as

$$\sigma_\alpha^*(t) = \sigma_\alpha^*(0) + \frac{i}{\hbar}\int_0^t dt' \, [\mathcal{H}_1^*(t'), \sigma_\alpha^*(0)] + \left(\frac{i}{\hbar}\right)^2 \int_0^t dt' \int_0^{t'} dt'' \, [\mathcal{H}_1^*(t'), [\mathcal{H}_1^*(t''), \sigma_\alpha^*(t'')]]. \tag{15}$$

The second and main hypothesis of the quantum computation of $P_\alpha(t)$ is (the first neglected the modification of the polarization of the μ^+ beam by the applied magnetic field) to set $\sigma_\alpha^*(t'') = \sigma_\alpha^*(0)$ to terminate the iteration. The validity of this approximation has been discussed in the past in connection with the Redfield theory (see, for example, Slichter (1963)). The results of this discussion are supported by our numerical work presented in section 4 which shows clearly that the approximation is quite good for describing either the initial part of a $P_\alpha(t)$ function (small time) in general or a $P_\alpha(t)$ function in the motional narrowing limit for all t values. We now use the expression for $\sigma_\alpha(t)$ deduced from equations (13)–(15) with $\sigma_\alpha^*(t'') = \sigma_\alpha^*(0)$ to get formulae for the depolarization functions.

3.3. Expression for the longitudinal depolarization function obtained from iteration

From the expressions given earlier, we can write

$$P_z(t) = P_z^{(0)}(t) + P_z^{(1)}(t) + P_z^{(2)}(t)$$

with:

$$P_z^{(0)}(t) = \frac{1}{2}\mathrm{Tr}\left\{\rho_s \sigma_z \exp\left(\frac{i\mathcal{H}_0 t}{\hbar}\right)\sigma_z \exp\left(\frac{-i\mathcal{H}_0 t}{\hbar}\right)\right\}$$

$$P_z^{(1)}(t) = \frac{1}{2}\mathrm{Tr}\left\{\rho_s \sigma_z \frac{i}{\hbar}\int_0^t dt' \exp\left(\frac{i\mathcal{H}_0 t}{\hbar}\right) \times \left[\exp\left(\frac{-i\mathcal{H}_0 t'}{\hbar}\right)\mathcal{H}_1 \exp\left(\frac{i\mathcal{H}_0 t'}{\hbar}\right), \sigma_z\right] \exp\left(\frac{-i\mathcal{H}_0 t}{\hbar}\right)\right\}$$

$$P_z^{(2)}(t) = \frac{1}{2}\mathrm{Tr}\left\{\rho_s \sigma_z \left(\frac{i}{\hbar}\right)^2 \int_0^t dt' \int_0^{t'} dt'' \exp\left(\frac{i\mathcal{H}_0 t}{\hbar}\right) A \exp\left(\frac{-i\mathcal{H}_0 t}{\hbar}\right)\right\}$$

and

$$A = \left[\exp\left(\frac{-i\mathcal{H}_0 t'}{\hbar}\right)\mathcal{H}_1 \exp\left(\frac{i\mathcal{H}_0 t'}{\hbar}\right), \left[\exp\left(\frac{-i\mathcal{H}_0 t''}{\hbar}\right)\mathcal{H}_1 \exp\left(\frac{i\mathcal{H}_0 t''}{\hbar}\right), \sigma_z\right]\right].$$

Notice that the exponent (0) does not have the same meaning as in section 2. For the computation of the traces we need to specify $\mathcal{H}_0$ and $\mathcal{H}_1$. We set $\mathcal{H}_0 = \mathcal{H}_s + \mathcal{H}_{\mu,z}$ where $\mathcal{H}_{\mu,z}$ is the μ^+ Zeeman Hamiltonian:

$$\mathcal{H}_{\mu,z} = -\tfrac{1}{2}\hbar\omega_\mu\sigma_z \qquad \omega_\mu = \gamma_\mu(B_{\text{ext}} + \langle B_z\rangle). \tag{16}$$

$\boldsymbol{B}$ is the effective magnetic field vector at the μ^+ due to the spins of the sample. We have $\langle A\rangle = \text{Tr}_s\{\rho_s A\}$. Notice that $\boldsymbol{B}$ does not contain any μ^+ spin operator. $\mathcal{H}_s$ has been defined previously. $\mathcal{H}_1$ which describes the magnetic interaction between the μ^+ spin and the spins in the sample can always be written as $\mathcal{H}_1 = -\frac{1}{2}\gamma_\mu\hbar\sigma\cdot\delta\boldsymbol{B}$ where $\delta\boldsymbol{B}$ is the fluctuating part of $\boldsymbol{B}$ ($\boldsymbol{B} = \delta\boldsymbol{B} + \langle\boldsymbol{B}\rangle$).

It is easily shown that $P_z^{(0)}(t) = 1$ and $P_z^{(1)}(t) = 0$. To get $P_z^{(2)}(t)$, the following identity, which can be proved, for a well defined f function, by integration by parts, is useful:

$$\int_0^t \mathrm{d}t' \int_0^{t'} \mathrm{d}t''\, f(t'-t'') = \int_0^t \mathrm{d}\tau\,(t-\tau) f(\tau). \tag{17}$$

After some algebra, using this identity, an expression for $P_z^{(2)}(t)$ can be derived:

$$P_z^{(2)}(t) = \frac{-\gamma_\mu^2}{2}\int_0^t \mathrm{d}\tau\,(t-\tau)\left[\exp(\mathrm{i}\omega_\mu\tau)\,\Phi_{+-}(\tau) + \exp(-\mathrm{i}\omega_\mu\tau)\,\Phi_{-+}(\tau)\right]$$

where $\Phi_{\alpha\beta}(\tau) = \frac{1}{2}[\langle\delta B_\alpha(\tau)\delta B_\beta\rangle + \langle\delta B_\beta\delta B_\alpha(\tau)\rangle]$ is a symmetrized correlation function of the magnetic field fluctuations at the μ^+ site. Notice that $\Phi_{\alpha\beta}(\tau) = \Phi_{\beta\alpha}(-\tau)$. We have

$$\delta B_\alpha(\tau) = \exp(\mathrm{i}\mathcal{H}_s\tau/\hbar)\,\delta B_\alpha \exp(-\mathrm{i}\mathcal{H}_s\tau/\hbar).$$

In section 2 it has been shown for the longitudinal relaxation function, from the strong collision model, that if the μ^+ diffuses sufficiently fast we can write

$$P_z(t) = \exp[-\psi_z(t)] \tag{18}$$

where $\psi_z(t)$ can be obtained from equation (10). If we postulate that this form for $P_z(t)$ is valid in general, $\psi_z(t)$ can be deduced from the quantum mechanical second-order iteration by identification of the expansion terms:

$$\psi_z(t) = \frac{\gamma_\mu^2}{2}\int_0^t \mathrm{d}\tau\,(t-\tau)\left[\exp(\mathrm{i}\omega_\mu\tau)\,\Phi_{+-}(\tau) + \exp(-\mathrm{i}\omega_\mu\tau)\,\Phi_{-+}(\tau)\right] \tag{19}$$

or

$$\psi_z(t) = \gamma_\mu^2\int_0^t \mathrm{d}\tau\,(t-\tau)\left\{\cos(\omega_\mu\tau)\left[\Phi_{xx}(\tau) + \Phi_{yy}(\tau)\right] + \sin(\omega_\mu\tau)\left[\Phi_{xy}(\tau) - \Phi_{yx}(\tau)\right]\right\}. \tag{20}$$

This result has already been given by McMullen and Zaremba (1978). As our derivation is systematic, it is easy to know where the approximations have been made. We have neglected the modification of the polarization of the μ^+ beam by the external magnetic field and the time evolution of $\sigma_\alpha^*(t'')$, $(\sigma_\alpha^*(t'') = \sigma_\alpha^*(0))$. In addition we have supposed that the quantum calculation gives the first two terms of the expansion of the exponential depolarization function (equation (18)). Notice that $P_z(t)$ given by equations (18)–(20) reduces to the NMR result if the characteristic time of the correlation functions, τ_c, is much shorter than t and $\omega_\mu\tau_c \ll 1$ (Moriya 1962). The next subsection shows that our method leads to a new expression for $P_x(t)$.

3.4. Expression for the transverse depolarization function obtained from iteration

The method used to derive $P_z(t)$ can be applied to the computation of the transverse depolarization function $P_x(t)$. The details are given in the appendix. As for $P_z(t)$, $P_x(t)$ can be written with the $\{+,-,z\}$ or $\{x,y,z\}$ coordinates. In the $\{+,-,z\}$ coordinates we have

$$P_x(t) = \mathrm{Re}\left\{\exp\left[\mathrm{i}\omega_\mu t - \Omega(t)\right]\right\} \tag{21}$$

with

$$\Omega(t) = \gamma_\mu^2 \int_0^t \mathrm{d}\tau\,(t-\tau)\Phi_{zz}(\tau) + \frac{\gamma_\mu^2}{2}\int_0^t \mathrm{d}\tau\,(t-\tau)\Phi_{-+}(\tau)\exp\left(-\mathrm{i}\omega_\mu\tau\right) + \mathrm{i}\frac{\gamma_\mu^2}{4\omega_\mu}\int_0^t \mathrm{d}\tau\,\left[\Phi_{++}(\tau)+\Phi_{--}(\tau)\right]\exp\left(-\mathrm{i}\omega_\mu\tau\right). \tag{22}$$

Using the $\{x,y,z\}$ coordinates we can write

$$P_x(t) = \exp\left[-\psi_x(t)\right]\cos\left[\omega_\mu t + \varphi(t)\right] \tag{23}$$

with

$$\psi_x(t) = \frac{\gamma_\mu^2}{2}\int_0^t \mathrm{d}\tau\,(t-\tau)\left\{2\Phi_{zz}(\tau) + \cos\left(\omega_\mu\tau\right)\left[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)\right] + \sin\left(\omega_\mu\tau\right)\left[\Phi_{xy}(\tau)-\Phi_{yx}(\tau)\right]\right\} - \frac{\gamma_\mu^2}{2\omega_\mu}\int_0^t \mathrm{d}\tau\,\sin\left(\omega_\mu\tau\right)\left[\Phi_{yy}(\tau)-\Phi_{xx}(\tau)\right]$$

$$\varphi(t) = \frac{\gamma_\mu^2}{2}\int_0^t \mathrm{d}\tau\,(t-\tau)\left\{\sin\left(\omega_\mu\tau\right)\left[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)\right] + \cos\left(\omega_\mu\tau\right)\left[\Phi_{yx}(\tau)-\Phi_{xy}(\tau)\right]\right\} + \frac{\gamma_\mu^2}{2\omega_\mu}\int_0^t \mathrm{d}\tau\,\cos\left(\omega_\mu\tau\right)\left[\Phi_{yy}(\tau)-\Phi_{xx}(\tau)\right]. \tag{24}$$

The expression for $\varphi(t)$ given above is valid when $\varphi(t) \ll 1$. It seems that these expressions for $P_x(t)$ have never been published so far. If the terms proportional to $1/\omega_\mu$ in the equations for $\psi_x(t)$ and $\varphi(t)$ are neglected (high-field approximation), we recover the NMR formulae (Moriya 1962, Heller 1976) and, as expected, $\psi_x(t)$ and $\psi_z(t)$ are related to each other:

$$\psi_x(t) = \tfrac{1}{2}\psi_z(t) + \gamma_\mu^2\int_0^t \mathrm{d}\tau\,(t-\tau)\Phi_{zz}(\tau).$$

In the motional narrowing limit and in the high-field approximation the phase term, $\varphi(t)$, is proportional to t and therefore leads to a frequency shift. We notice that this shift is influenced by spin–spin correlations (the $\Phi_{\alpha\beta}(\tau)$ terms).

The functional dependence of the μSR depolarization functions on the spin correlation functions is such that symmetry arguments can be used to simplify this dependence. Group theory should be useful for that purpose.

To understand the effect of the different terms in the depolarization functions, in the next section, we will discuss the case of the μ^+ diffusing in a lattice of nuclear dipoles.

4. Application to the case of nuclear dipole systems

The expressions for $P_z(t)$ and $P_x(t)$ given in the previous section are general and do not depend on the type of interactions included in the sample Hamiltonian, $\mathcal{H}_s$, and the coupling Hamiltonian, $\mathcal{H}_1$. The main hypothesis made in the course of the calculation of the functions is that the μ^+ diffusion is sufficiently rapid. This will be apparent when discussing the results which we are now going to derive. The μ^+ depolarization is due to the interaction between the μ^+ spin and the fluctuations of the electronic and nuclear spins of the lattice. Here we do not consider the electronic spins. In addition we neglect the dipolar interaction between the nuclear spins which is weak. Therefore $\mathcal{H}_1$ only includes the dipolar interaction between the μ^+ and the localized nuclear spins. We have

$$\mathcal{H}_1 = \mathcal{H}_{\text{dip}} = -\frac{\gamma_\mu \hbar}{2}\left[\sigma_z \delta B_z + \frac{1}{2}\left(\sigma_+ \delta B_- + \sigma_- \delta B_+\right)\right]$$

where

$$\delta B_z = \sum_{i=1}^{N}\left[\frac{A_i}{2}\exp\left(-\mathrm{i}\varphi_i\right) I_{i,+} + \frac{A_i}{2}\exp\left(\mathrm{i}\varphi_i\right) I_{i,-} - 2C_i I_{i,z}\right]$$

$$\delta B_+ = \sum_{i=1}^{N}\left[C_i I_{i,+} + B_i \exp\left(2\mathrm{i}\varphi_i\right) I_{i,-} + A_i \exp\left(\mathrm{i}\varphi_i\right) I_{i,z}\right]$$

$$\delta B_- = \left(\delta B_+\right)^* . \tag{25}$$

The previous equation is written taking into account the fact that for a nuclear system at a usual temperature, $I_{i,\alpha}(\tau) = \delta I_{i,\alpha}(\tau)$. The geometrical factors are

$$A_i = 3D_i \cos\theta_i \sin\theta_i \qquad B_i = \tfrac{3}{2} D_i \sin^2\theta_i$$

and

$$C_i = D_i \frac{1 - 3\cos^2\theta_i}{2} \qquad \text{with } D_i = \frac{\mu_0}{4\pi}\frac{\gamma_i \hbar}{r_i^3}.$$

r_i, θ_i and φ_i are the distance, polar and azimuthal angles of nucleus i relative to the μ^+. γ_i is the gyromagnetic ratio of I_i.

The μ^+ diffusion can be introduced by proper terms in the μ^+ sample system Hamiltonian. This quantum approach, which can describe the $P_\alpha(t)$ functions when the μ^+ diffuses coherently (McMullen and Zaremba 1978, Kondo 1986), leads to a complicated formalism. In most practical cases a semiclassical approach is sufficient. In this latter approach, as in NMR (Abragam 1961), the μ^+ coordinates are considered to be time-dependent and an ensemble average is performed over these coordinates.

In the previous section we have shown that the $P_\alpha(t)$ functions depend on magnetic field correlation functions and therefore, using equation (25), on terms such as

$$q_\alpha\left(r_i\right) q_\beta\left(r_j\right) \left\langle I_{i,\alpha}(\tau) I_{j,\beta}(0)\right\rangle \tag{26}$$

where the q_α and q_β functions, which describe the geometry around the μ^+, can be deduced by identification using equation (25). We will neglect the case where a nuclear spin is nearest neighbour to two μ^+ localization sites. Thus we will take $r_j = r_i$ in equation (26). As a result of the μ^+ diffusion, r_i is a stochastic function, $r_i(\tau)$. Therefore we substitute the product function $q_\alpha(r_i)\,q_\beta(r_i)$ in equation (26) by its average over the stochastic diffusion process, $\overline{q_{i,\alpha}(\tau)q_{i,\beta}(0)}$. Here to proceed further we write $\overline{q_{i,\alpha}(\tau)q_{i,\beta}(0)} = q_{i,\alpha}(0)q_{i,\beta}(0)f_\nu(\tau)$ and assume that the correlation function $f_\nu(\tau)$ takes the form

$$f_\nu(\tau) = \exp(-\nu|\tau|) \tag{27}$$

where ν is the μ^+ jump frequency. McMullen and Zaremba (1978) have shown that this form is a good approximation for a μ^+ hopping on a simple cubic lattice. Basically with this simple form the possibility for the μ^+ to return to its original site is neglected. Therefore this approximation should break down if the number of nearest-neighbour interstitial sites is small. This could be the case for a μ^+ diffusing between octahedral or tetrahedral interstitial sites of a BCC lattice.

Notice that, since at normal experimental temperatures $\langle\beta\mathcal{H}_s\rangle \ll 1$, the density operator of the nuclear spin system is very simple:

$$\rho_s = \mathbf{1}/\prod_{i=1}^{N}(2I_i+1)\,.$$

We first discuss the longitudinal depolarization function.

4.1. Study of the longitudinal depolarization function

The $P_z(t)$ function can be computed from equations (18) and (19) and an expression for the field correlation function $\Phi_{-+}(\tau) = \Phi^*_{+-}(\tau)$. Using the definition of this function, the expression of the dipolar magnetic field at the μ^+ site and our prescription for taking the μ^+ diffusion into account, some algebra gives

$$\Phi_{-+}(\tau) = f_\nu(\tau)\sum_{i=1}^{N} p_i(\tau)$$

with

$$\begin{aligned} p_i(\tau) = {} & A_i^2\Lambda_{ii}^{zz}(\tau) + B_i^2\Lambda_{ii}^{+-}(\tau) + C_i^2\Lambda_{ii}^{-+}(\tau) \\ & + B_iC_i\left[\exp(-2\varphi_i)\,\Lambda_{ii}^{++}(\tau) + \exp(2i\varphi_i)\,\Lambda_{ii}^{--}(\tau)\right] \\ & + A_iC_i\left[\exp(-i\varphi_i)\,\Lambda_{ii}^{z+}(\tau) + \exp(i\varphi_i)\,\Lambda_{ii}^{-z}(\tau)\right] \\ & + A_iB_i\left[\exp(i\varphi_i)\,\Lambda_{ii}^{z-}(\tau) + \exp(-i\varphi_i)\,\Lambda_{ii}^{+z}(\tau)\right]. \end{aligned} \tag{28}$$

We have used the symmetrized spin–spin correlation function $\Lambda_{ij}^{\alpha\beta}(\tau)$:

$$\Lambda_{ij}^{\alpha\beta}(\tau) = \tfrac{1}{2}\left[\langle I_{i,\alpha}(\tau)I_{j,\beta}\rangle + \langle I_{j,\beta}I_{i,\alpha}(\tau)\rangle\right]. \tag{29}$$

Because of the approximation discussed after equation (26) ($r_j = r_i$) we take $i = j$. As we neglect the interaction between the nuclear spins, we have

$$I_{i,\alpha}(\tau) = \exp\left(i\mathcal{H}_{i,s}\tau/\hbar\right) I_{i,\alpha} \exp\left(-i\mathcal{H}_{i,s}\tau/\hbar\right)$$

where $\mathcal{H}_{i,s}$ is the Hamiltonian of nuclear spin i.

In order to understand the physical meaning of our result we first assume that the nuclear spins feel only an external magnetic field. Therefore we have $\mathcal{H}_{i,s} = -\hbar\omega_{i,z} I_{i,z}$ with $\omega_{i,z} = \gamma_i(1 + K_i)B_{\text{ext}}$ where γ_i is the gyromagnetic ratio and K_i the Knight shift. With this simple Hamiltonian, $I_{i,\alpha}(\tau)$ can be computed analytically using the identities $\exp(ia I_z) I_\pm \exp(-ia I_z) = \exp(\pm ia) I_\pm$. After some algebra we get

$$\psi_z(t) = \sum_{i=1}^{N} \omega_{i,d}^2 I_i (I_i + 1) \times \left[\tfrac{1}{6}\left(1 - 3\cos^2\theta_i\right)^2 g_\nu^{(c)}(\omega_\mu - \omega_{i,z}, t) + 3\sin^2\theta_i \cos^2\theta_i g_\nu^{(c)}(\omega_\mu, t) + \tfrac{3}{2}\sin^4\theta_i g_\nu^{(c)}(\omega_\mu + \omega_{i,z}, t)\right]. \quad (30)$$

$\omega_{i,d} = (\mu_0/4\pi)\gamma_\mu\gamma_i\hbar/r_i^3$ is the dipolar pulsation for I_i. We have set

$$g_\nu^{(c)}(\omega, t) = \int_0^t d\tau\,(t - \tau) f_\nu(\tau) \cos(\omega\tau).$$

Notice we have $g_\nu^{(c)}(\omega, t) = [1 - \cos(\omega t)]/\omega^2$ in the quasi-static limit and $g_\nu^{(c)}(\omega, t) = (\nu t)/(\nu^2 + \omega^2)$ in the motional narrowing limit. Equation (30) has already been published by Hayano *et al* (1979). In zero field, $\omega_{i,z} = \omega_\mu = 0$, we recover the function given in equation (10) with

$$\Delta^2 = \tfrac{1}{6}\sum_{i=1}^{N} \omega_{i,d}^2 I_i (I_i + 1)\left(5 - 3\cos^2\theta_i\right). \quad (31)$$

The fact that the quantum calculation gives the Abragam formula (with $2\Delta^2$ instead of Δ^2) does not seem to have been noticed before.

The nuclear spins around the μ^+ larger than $\frac{1}{2}$ feel an electric field gradient which is produced by the μ^+ electric charge (Schenck 1985) and the lattice electric charges for nuclear spins with local symmetry less than cubic. To describe the effect of this electric field gradient, we add a new term to $\mathcal{H}_{i,s}$ which, for the case of a gradient due only to the μ^+, is simply written (Hartmann 1977) $\mathcal{H}_{i,q} = \hbar\omega_{i,q}[(n_i \cdot I_i)(n_i \cdot I_i) - I_i(I_i + 1)/3]$. n_i is the unit vector linking the μ^+ localization site to the lattice site of nucleus i.

In order to appreciate the quantum effects on $P_z(t)$, in figure 3 we present two examples (zero field case and $B_{\text{ext}} = 30$ G) of this function computed from three different models. The functions are calculated either from our second iteration formula or from the strong collision model. The static functions of the latter model are either taken from Kubo–Toyabe theory or are computed exactly using Celio's method (1986). The result of the first and third models cannot be distinguished on the figure. We have supposed that the μ^+ is diffusing between the octahedral sites of metallic copper. We have taken the values given by Luke *et al* (1991). We have

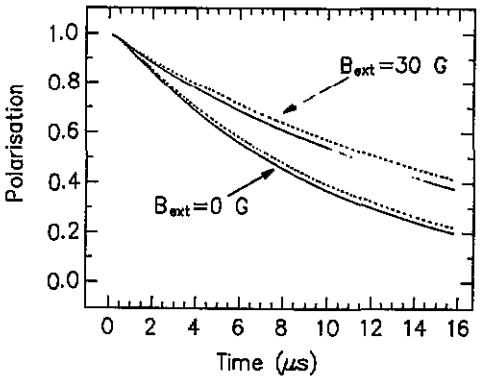

Figure 3. Examples of the effect of the spin dynamics on the longitudinal relaxation function in the case of the μ^+ in metallic copper. The examples are given for two values of the external magnetic field, B_{ext}. The full curves present these functions given by our second-order iteration or the strong collision model with the exact static depolarization functions computed from quantum mechanics (the two types of curves cannot be distinguished) and the dotted curves the results of the strong collision model with the static functions given by the Kubo–Toyabe model. The parameters of the curves are given in the main text.

$\gamma_{Cu} = 72.51$ Mrad s^{-1} T^{-1}, $Q_{Cu} = -0.205$ barn, $V_{zz} = 0.441 \times 10^{-21}$ V m^{-2} and $r = 1.81$ Å. Q_{Cu} is the nuclear quadrupole moment of Cu, V_{zz} the zz component of the electric field gradient tensor at one of the six Cu nuclei and r the distance between the μ^+ and a Cu nucleus. We consider only the six nearest-neighbour Cu nuclei to the μ^+ because the exact computation of the static $P_z(t)$ functions can only be performed for this restricted number of nuclei. This leads to $\omega_q = -3.200$ μs^{-1}, $\omega_d = 0.110$ μs^{-1} and $\Delta = \omega_d\sqrt{12} = 0.381$ MHz (the effect of the electric field gradient on Δ is taken into account approximately; see Schenck (1985)). We have chosen $\nu = 3$ MHz which should be roughly the value of the μ^+ jump frequency at room temperature. While the $P_z(t)$ functions computed either from the strong collision model with the exact static functions or from our second iteration formula cannot be distinguished on the figure, the functions obtained from the strong collision model with the static functions computed according to Kubo–Toyabe are clearly different. The functions deduced from this latter method decrease more slowly. This can be understood intuitively. For example let us consider the structure of the iteration formula for the zero-field case. Although the Zeeman and the quadrupolar Hamiltonians do not have the same structure, it is certain that when the μ^+ diffusion is sufficiently fast, terms such as

$$\frac{\nu}{\nu^2 + \omega^2} = \frac{1}{\nu}\left(\frac{1}{1 + \omega^2/\nu^2}\right) \tag{32}$$

are present; see equation (30). Thus $P_z(t)$ contains flipping terms which describe the spin dynamics. We find, for example, a term with the factor $\nu/[\nu^2 + (\omega_\mu - \omega_{i,z})^2]$ in the motional narrowing limit which, following well known NMR theory (see for example Slichter (1963)), describes the mutual spin flip of the μ^+ and I_i spins. For Cu as the ratio ω^2/ν^2 is not negligible when compared with one, $\omega_q^2/\nu^2 = 1.22$; this leads to an increase in the depolarization. Whereas the zero-field curve shown in figure 3 computed from the iteration formula (or the strong collision model with the

exact static function) can be described by the strong collision model with the static function calculated according to Kubo–Toyabe if we take $\nu = 2.8$ MHz, it is not possible to fit the $B_{ext} = 30$ G curve by this latter theory with $\Delta = 0.381$ MHz. The closest curve obtained with this Δ value gives $\nu = 2.4$ MHz. Therefore, at the best, an experimental spectrum analysed with this model leads to an under-estimation of the jump frequency. In addition the analysis of the zero- and longitudinal-field data are not consistent.

A depolarization function computed from the iteration formula must be a good approximation at least when the μ^+ jumping rate is large enough. The fact that the strong collision model with the exact static function and the iteration formula give the same result at $\nu = 3$ MHz means that all the spin dynamics is already contained in the exact static function. The diffusion described by the strong collision model does not add a new spin dynamics mechanism. It only modifies the properties of it. This can be understood because the physical hypothesis which we have made to compute $P_z(t)$ from the iteration equation are the same as the ones leading to the integral equation of the strong collision model. The Kubo–Toyabe method does not describe the physics in detail: it neglects the nuclear spin dynamics which is essential. The strong collision model gives a good description (within its own hypothesis; basically the correlations between the nuclear spins are neglected) if the right static function is used.

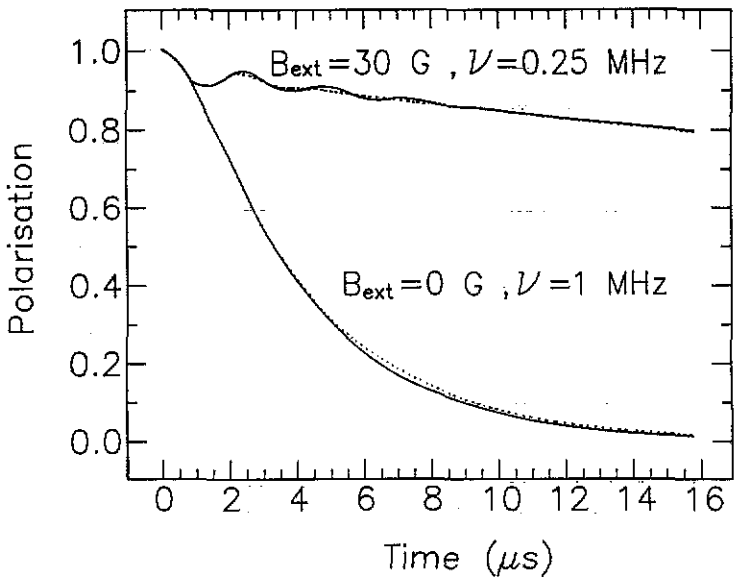

Figure 4. Study of the validity of the second-order iteration. The curves are drawn for the case of a μ^+ in metallic copper. The full curves present the results of our second-order iteration. The dotted curves show the prediction of the strong collision model with the static functions computed from quantum mechanics. ν is the μ^+ jump rate.

Up to now we do not know what is the smallest value of the jumping rate for which the iteration formula can be used to describe the μ^+ diffusion in Cu. In figure 4 we present $P_z(t)$ computed either from the strong collision model with the exact static function or from the second-order iteration. It is quite clear that even for $\nu = 1$ MHz at zero field the iteration is a reasonable approximation. This fact

is interesting because it means that our iteration formula can be used to describe $P_z(t)$ even for a relatively slowly diffusing μ^+. Thus, for most practical cases, the function can be obtained with a modest computing time. The structure of the iteration formula (see equation (30) for an example) clearly indicates that the spin dynamics of the nuclear spins is important. This probably explains why the exact static depolarization function is different from the prediction of Kubo and Toyabe for the longitudinal geometry.

The results just described could lead to the idea that the second-order iteration gives reliable results only in the motional narrowing limit. This is not so. It gives a good picture of the initial part of $P_z(t)$ in the whole range of ν. For example, when an electric field gradient and/or a magnetic field acts on the lattice nuclei, the iteration result has a less restricted time range validity compared with the usual parabola obtained from Van Vleck's method because of the effect of the non-secular terms. This is clearly seen if one looks at the initial part of the numerically exact $P_z^{(0)}(t)$ function for the case of a static μ^+ in Cu at zero field: whereas the parabola is a good approximation only up to 0.7 μs, the iteration formula gives a fair approximation up to 2 μs. As has already been mentioned, this is in contrast to what is seen for the case of negligible electric field gradients and external field. This is probably related to the fact that the longitudinal Abragam formula (equation (10) derived using the distribution method) is a good approximation (if ν is large enough) of the quantum result for the zero-field case only if no gradient acts on the nuclei.

4.2. Study of the transverse depolarization function

The $P_x(t)$ function can be computed from equations (21) and (22) with expressions for $\Phi_{-+}(\tau) = \Phi_{+-}^*(\tau)$, $\Phi_{zz}(\tau)$ and $\Phi_{++}(\tau) = \Phi_{--}^*(\tau)$. $\Phi_{-+}(\tau)$ is given by equation (28). The other two field correlation functions can be determined using the expression of the dipolar magnetic field at the μ^+ site and our prescription for taking the μ^+ diffusion into account. We have

$$\Phi_{zz}(\tau) = f_\nu(\tau) \sum_{i=1}^{N} s_i(\tau)$$

with

$$\begin{aligned} s_i(\tau) = {} & \frac{A_i^2}{4} \exp(-2i\varphi_i)\, \Lambda_{ii}^{++}(\tau) + \frac{A_i^2}{4} \exp(2i\varphi_i)\, \Lambda_{ii}^{--}(\tau) + 4C_i^2 \Lambda_{ii}^{zz}(\tau) \\ & - A_i C_i \exp(i\varphi_i) \left[\Lambda_{ii}^{-z}(\tau) + \Lambda_{ii}^{z-}(\tau)\right] - A_i C_i \exp(-i\varphi_i) \\ & \times \left[\Lambda_{ii}^{+z}(\tau) + \Lambda_{ii}^{z+}(\tau)\right] + \frac{A_i^2}{4} \left[\Lambda_{ii}^{+-}(\tau) + \Lambda_{ii}^{-+}(\tau)\right] \end{aligned} \tag{33}$$

and

$$\Phi_{++}(\tau) = f_\nu(\tau) \sum_{i=1}^{N} u_i(\tau)$$

with

$$\begin{aligned} u_i(\tau) = {} & C_i^2 \Lambda_{ii}^{++}(\tau) + B_i^2 \exp(4i\varphi_i)\, \Lambda_{ii}^{--}(\tau) + A_i^2 \exp(2i\varphi_i)\, \Lambda_{ii}^{zz}(\tau) \\ & + C_i B_i \exp(2i\varphi_i) \left[\Lambda_{ii}^{+-}(\tau) + \Lambda_{ii}^{-+}(\tau)\right] + C_i A_i \exp(i\varphi_i) \\ & \times \left[\Lambda_{ii}^{+z}(\tau) + \Lambda_{ii}^{z+}(\tau)\right] + B_i A_i \exp(3i\varphi_i) \left[\Lambda_{ii}^{-z}(\tau) + \Lambda_{ii}^{z-}(\tau)\right]. \end{aligned} \tag{34}$$

In order to understand our result we first assume that the nuclear spins feel only an external magnetic field. After some algebra we get

$$\psi_x(t) = \tfrac{1}{2}\psi_z(t) + \sum_{i=1}^{N} \omega_{i,\mathrm{d}}^2 I_i (I_i + 1)\, v_i (\omega_\mu, \omega_{i,z}, t)$$

where

$$\begin{aligned} v_i (\omega_\mu, \omega_{i,z}, t) = {} & \tfrac{1}{3}\left(1 - 3\cos^2\theta_i\right)^2 g_\nu^{(\mathrm{c})}(0,t) + 3\sin^2\theta_i \cos^2\theta_i g_\nu^{(\mathrm{c})}(\omega_{i,z}, t) \\ & + \cos(2\varphi_i)\,\frac{3\sin^4\theta_i - 2\sin^2\theta_i}{2\omega_\mu} h_{\nu,\omega_\mu}^{(\mathrm{s})}(\omega_{i,z}, t) \\ & + \frac{3\cos(2\varphi_i)\cos^2\theta_i \sin^2\theta_i}{2\omega_\mu} k_\nu^{(\mathrm{s})}(\omega_\mu, t) \end{aligned}$$

and

$$\varphi(t) = \sum_{i=1}^{N} \omega_{i,\mathrm{d}}^2 I_i (I_i + 1)\, w_i (\omega_\mu, \omega_{i,z}, t)$$

where

$$\begin{aligned} 2w_i (\omega_\mu, \omega_{i,z}, t) = {} & \tfrac{1}{6}\left(1 - 3\cos^2\theta_i\right)^2 g_\nu^{(\mathrm{s})}(\omega_\mu - \omega_{i,z}, t) \\ & + 3\sin^2\theta_i \cos^2\theta_i g_\nu^{(\mathrm{s})}(\omega_\mu, t) + \tfrac{3}{2}\sin^4\theta_i g_\nu^{(\mathrm{s})}(\omega_\mu + \omega_{i,z}, t) \\ & - \cos(2\varphi_i)\,\frac{3\sin^4\theta_i - 2\sin^2\theta_i}{\omega_\mu} h_{\nu,\omega_\mu}^{(\mathrm{c})}(\omega_{i,z}, t) \\ & - \frac{3\cos(2\varphi_i)\cos^2\theta_i \sin^2\theta_i}{\omega_\mu} k_\nu^{(\mathrm{c})}(\omega_\mu, t). \end{aligned} \tag{35}$$

We have defined

$$g_\nu^{(\mathrm{s})}(\omega, t) = \int_0^t \mathrm{d}\tau\,(t-\tau) f_\nu(\tau) \sin(\omega\tau)$$

$$h_{\nu,\omega_\mu}^{(\mathrm{s})}(\omega, t) = \int_0^t \mathrm{d}\tau\, \sin(\omega_\mu\tau) \cos(\omega\tau) f_\nu(\tau)$$

$$h_{\nu,\omega_\mu}^{(\mathrm{c})}(\omega, t) = \int_0^t \mathrm{d}\tau\, \cos(\omega_\mu\tau) \cos(\omega\tau) f_\tau(\tau)$$

$$k_\nu^{(\mathrm{s})}(\omega, t) = \int_0^t \mathrm{d}\tau\, \sin(\omega\tau) f_\nu(\tau)$$

$$k_\nu^{(\mathrm{c})}(\omega, t) = \int_0^t \mathrm{d}\tau\, \cos(\omega\tau) f_\tau(\tau).$$

These formulae have never been published. The expression for $\psi_x(t)$ given by Hayano *et al* (1979) is identical to our expression if our two terms proportional to

$1/\omega_\mu$ are neglected. In the high-field approximation the $g_\nu^{(c)}(\omega,t)$ and $g_\nu^{(s)}(\omega,t)$ functions go to zero. Thus we recover, as expected, the transverse Abragam formula (equation (9)) with

$$\Delta^2 = \sum_{i=1}^{N} \omega_{i,d}^2 I_i (I_i + 1) \tfrac{1}{3} \left(1 - 3\cos^2\theta_i\right)^2 .$$

In order to test the effect of the spin dynamics and the terms proportional to $1/\omega_\mu$ on $P_x(t)$, we present in figure 5 an example of this function computed with three different approximations. One curve (full) has been calculated using our complete second-order iteration formula. A computation of $P_x(t)$ has been made with this formula but omitting the $1/\omega_\mu$ terms. The results of these two computations cannot be distinguished in figure 5. Therefore the $1/\omega_\mu$ terms have no effect for the case considered. A numerical study shows that the $1/\omega_\mu$ terms have a small effect if the μ^+ diffusion is not too fast ($\nu = 1$ MHz). The second curve (dotted) is the result of the transverse Abragam formula (equation (9)) with $\Delta = 0.234$ MHz. This value has been calculated using the expression for Δ just given. We have supposed that the μ^+ feels the effect of only two Cu nuclear magnetic moments. One is located at 1.5 Å on the x-axis and the other at 2 Å on the y-axis. Such a non-symmetric environment has been chosen in order to detect the effect of the $1/\omega_\mu$ terms. When the μ^+ is in a highly symmetric environment, these terms do not contribute to $P_x(t)$. This can be seen from equation (24). A field of 20 G ($B_{ext} > 5(\Delta/\gamma_\mu) = 13.7$ G) is applied along the z-axis. We take $\nu = 3$ MHz and suppose that the nuclei do not feel any electric field gradient. Figure 5 clearly shows that whereas the terms proportional to $1/\omega_\mu$ have no effect, the spin dynamics (which is described by the terms which contain $\omega_{i,z}$ and ω_μ) has a strong influence on the depolarization function. Notice that a frequency shift is present.

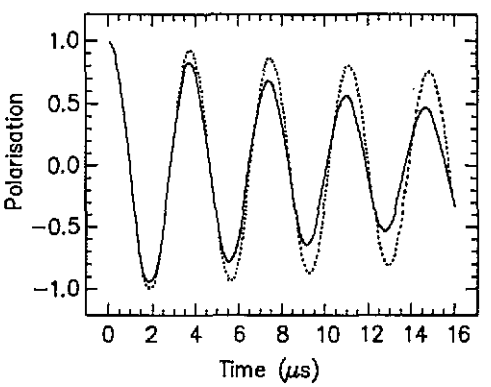

Figure 5. Example of the effect of the spin dynamics on the transverse depolarization function for a small external magnetic field. The full curve gives the result of our second-order iteration. The dotted curve shows the prediction of the Abragam formula. The external applied magnetic field is $B_{ext} = 20$ G. The other parameters of the curves are given in the main text.

In the case of the longitudinal depolarization function we have indicated that the second-order iteration is a good approximation if the μ^+ diffusion is sufficiently rapid (and in general if one is only interested in the initial part of the function). We may

expect for the transverse depolarization function that this approximation is tolerable even for a static μ^+ if one is not interested in too large a time. This hypothesis is supported by two facts. First the $P_x(t)$ functions described by the Markov process with the static function given by equation (7) (the case of a static μ^+ is included in the framework of the stochastic model) and the transverse Abragam formula are never very different (Kehr *et al* 1978). Second when $P_x(t)$ can be easily computed numerically using equation (12) as, for example, in the case of a static μ^+ interacting with only one nucleus, the data can be reasonably described by our iteration formula if $t \lesssim 9$ μs. However more work is needed to check the validity of this hypothesis.

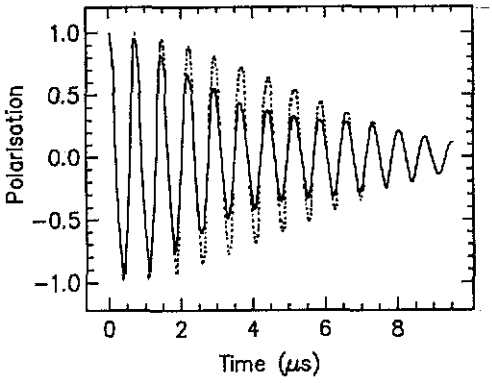

Figure 6. Effect of the spin dynamics on the transverse depolarization function in the case of a static μ^+ in metallic copper. To avoid the possible effect of the level crossing we neglect the electric field gradient due to the μ^+. The full curve gives the result of our second-order iteration. The dotted curve shows the prediction of equation (7). The external applied magnetic field is $B_{ext} = 100$ G.

Before finishing this section we would like to point out an interesting result which should be checked out by an exact calculation of $P_x(t)$. In figure 6 we present $P_x(t)$ computed with our iteration formula (full curve) for the case of a static μ^+ in Cu neglecting the electric field gradient due to the μ^+. An external field of 100 G is applied along the [110] direction. The dotted curve is the result of equation (7). The two curves are clearly different. The curve computed from the second-order iteration formula does not have a Gaussian envelope. Thus for this case the spin dynamics still influences the depolarization function at $B_{ext} = 100$ G. We point out that even at 300 G there is still a difference between the classical and quantum curves. On the other hand, the same computation when B_{ext} is along the [100] direction gives a $P_x(t)$ function which cannot be distinguished from the result of equation (7). A close look at equation (35) shows that this is expected because θ_i is either 0, $\pi/2$ or π. Thus the terms proportional to $\sin^2\theta_i \cos^2\theta_i$ do not contribute to $\psi_x(t)$. In addition the other terms, except the one leading to the Abragam formula (first term of the v_i function), are negligible because B_{ext} is large. Therefore the influence of the spin dynamics on the depolarization functions depends strongly on the geometrical conditions.

5. Summary, possible extensions and conclusions

In this paper we have discussed in some detail the stochastic model (with the static depolarization functions given by the distribution method) which is usually used to

describe the μSR data. We have noticed that a renormalized Abragam formula is a good approximation of the longitudinal depolarization function, $P_z(t)$, when the μ^+ diffusion is sufficiently rapid. We have determined at which value of the external field the simple Gaussian approximation to the static transverse depolarization function is valid.

The main concern of this work has been the quantum computation of the depolarization functions using a second-order iterative method. We have found formulae which are extensions of the ones used in NMR.

We have applied these formulae to compute the depolarization functions for a μ^+ diffusing in a nuclear dipole lattice. Our results are an extension of the ones given by Hayano *et al* (1979). Using our second-order formula, we have computed the depolarization functions for the two experimental geometries, in different physical situations. Our results show that it is necessary to take into account the spin dynamics to describe the depolarization functions at low fields properly.

We have shown that the exact static depolarization function contains all the spin dynamics. The diffusion does not add a new spin dynamics mechanism. This fact shows that it is important to describe the spin dynamics for a static μ^+ properly before trying to include the effect of diffusion on the depolarization function.

Our results represent the first step towards a full description of the depolarization functions for the case of a μ^+ diffusing sufficiently fast in a nuclear dipole system. In order to reach a completely satisfactory description, the two hypotheses which we have made should be studied. We have neglected the nuclear spin correlations (equation (26)) which could be important if the μ^+ jumps between sites which have common nuclear spins as nearest neighbours. In addition we have chosen a phenomenological form for the correlation function, $f_\nu(\tau)$, which takes into account the μ^+ diffusion (equation (27)). $f_\nu(\tau)$ depends on the lattice structure and the μ^+ localization site. A numerical calculation of this function should be possible for each physical case. The result could be introduced in equations (28), (33) and (34) to compute the depolarization functions.

Within the hypotheses of the strong collision model a μSR depolarization function can be computed in a reliable way if the related exact static function is used. This always requires a large amount of CPU (central processing unit) time. For many physically interesting cases the calculation cannot even be performed (Dalmas de Réotier *et al* 1990) because this time would be too long. We now suggest a new method which, as in Celio's method (1986), uses the Trotter formula but in a different way. Instead of decomposing the total Hamiltonian, $\mathcal{H}$, as the sum of Hamiltonians for each μ^+ spin–nuclear spin pair, a more fruitful method could be to write $\mathcal{H}t = N(\mathcal{H}\tau)$ with $t = N\tau$. This sliding of the time should lead with the help of the Trotter formula to a path representation of the depolarization function. The advantage of the proposed method over the one used by Celio could be the possibility of finding a semiclassical approach for the depolarization function. (Up to now only two methods exist. We have either the fully quantum or the classical distribution method.)

In this paper we have considered that only one source of depolarization is present. But when two sources act to depolarize the μ^+ as in MnSi (Kadono *et al* 1990b), the resultant depolarization function is the product of the function describing the fluctuations of the electronic magnetism by the function taking into account the depolarization due to the ^{55}Mn magnetic moments. Then to deduce reliable information on the electronic fluctuations from the experimental data, it is important to describe the second source of depolarization properly. The application of our results could

help to resolve part of the present inconsistency in the analysis of the data.

In conclusion we have shown that to get a consistent analysis of the μ^+ diffusion data in a nuclear spin lattice, it is important to take into account the spin dynamics. A numerical comparison between the results of the strong collision model with the exact static function and our formula for the longitudinal depolarization in the case of Cu has allowed us to study the validity of the iteration. We have found that it is surprisingly good. The advantage of using our formula over the strong collision model is while the computing time needed to calculate a depolarization function using our formula is only about 1 min, the computation with the second model requires, to get a reliable result, many hours of CPU (on a VAX 8800). Our result suggests that the difference between a numerically exact static depolarization function and the related function deduced from the distribution method is due to the neglect of the nuclear spin dynamics by the latter method. Clearly more work is needed to understand the effect of the spin dynamics.

Acknowledgments

We would like to thank P C M Gubbens for his help in drawing the figures for this paper. One of us (Dalmas de Réotier) is supported by the Alexander von Humboldt Stiftung.

Note added in proof. Recently it has been shown (Dalmas de Réotier P, Yaouanc A and Meshkov S V 1992 *Phys. Lett.* **162A** 206) that it is possible to compute exactly (better than 1%) a static $P_z(t)$ function using a Monte Carlo method with classical mechanics including the effect of the spin dynamics if the dimension of the Hilbert space is large enough. So far, this method is limited to the case where no electric field gradient acts on the nuclei. The CPU time needed is important. Thus our iterative procedure is still valuable. In addition it can be used even if the Hilbert space is small.

Appendix. Sketch of the derivation of $P_x(t)$

We follow the method presented in section 3.1 for the derivation of $P_z(t)$. The computation of $P_x(t)$ up to the second order in the iteration gives this function as the sum of three terms. The first two terms are easily computed. We have

$$P_x^{(0)}(t) = \cos(\omega_\mu t) \qquad P_x^{(1)}(t) = 0. \tag{A1}$$

The computation of $P_x^{(2)}(t)$ is more involved. The iteration gives

$$P_x^{(2)}(t) = \frac{1}{2}\left(\frac{\mathrm{i}}{\hbar}\right)^2 \int_0^t \mathrm{d}t' \int_0^{t'} \mathrm{d}t'' \ \mathrm{Tr}\left\{\rho_s \sigma_x \exp(\mathrm{i}\mathcal{H}_0 t/\hbar)\, A \exp(-\mathrm{i}\mathcal{H}_0 t/\hbar)\right\}$$

with

$$A = [\exp(-\mathrm{i}\mathcal{H}_0 t'/\hbar)\,\mathcal{H}_1 \exp(\mathrm{i}\mathcal{H}_0 t'/\hbar)\,, [\exp(-\mathrm{i}\mathcal{H}_0 t''/\hbar)\,\mathcal{H}_1 \exp(\mathrm{i}\mathcal{H}_0 t''/\hbar)\,, \sigma_x]]\,.$$

A tedious but straightforward evaluation of the trace leads to

$$P_x^{(2)}(t) = -\gamma_\mu^2 \cos(\omega_\mu t) \int_0^t d\tau\,(t-\tau)\Phi_{zz}(\tau) - \frac{\gamma_\mu^2}{2}\cos(\omega_\mu t)\int_0^t d\tau\,(t-\tau)$$
$$\times \{\cos(\omega_\mu\tau)[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)] + \sin(\omega_\mu\tau)[\Phi_{xy}(\tau)-\Phi_{yx}(\tau)]\}$$
$$-\frac{\gamma_\mu^2}{2}\cos(\omega_\mu t)\int_0^t dt'\int_0^{t'} dt''\,k_c(t',t'') - \frac{\gamma_\mu^2}{2}\sin(\omega_\mu t)\int_0^t d\tau\,(t-\tau)$$
$$\times \{\sin(\omega_\mu\tau)[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)] + \cos(\omega_\mu\tau)[\Phi_{yx}(\tau)-\Phi_{xy}(\tau)]\}$$
$$-\frac{\gamma_\mu^2}{2}\sin(\omega_\mu t)\int_0^t dt'\int_0^{t'} dt''\,k_s(t',t'')$$

where

$$k_c(t',t'') = \cos[\omega_\mu(t'+t'')][\Phi_{yy}(t'-t'') - \Phi_{xx}(t'-t'')]$$
$$-\sin[\omega_\mu(t'+t'')][\Phi_{xy}(t'-t'') + \Phi_{yx}(t'-t'')]\,.$$

The k_s function is the k_c function with the substitution of the cos and sin functions by the sin and $-$cos functions respectively. The identity given by equation (17) has been used. The expression given here can be simplified with the help of the following identity

$$\int_0^t dt'\int_0^{t'} dt''\exp[i\omega_\mu(t'+t'')]\,\Phi(t'-t'')$$
$$= \frac{1}{2i\omega_\mu}\int_0^t \{\exp[i\omega_\mu(2t-\tau)] - \exp(i\omega_\mu\tau)\}\,\Phi(\tau)\,d\tau$$

which can be proved by integration by parts. In addition taking into account the fact that $\Phi_{\alpha\beta}(t)$ is a real function when $\{\alpha,\beta\} = \{x,y,z\}$, we finally obtain

$$P_x^{(2)}(t) = -\gamma_\mu^2\cos(\omega_\mu t)\int_0^t d\tau\,(t-\tau)\Phi_{zz}(\tau) - \frac{\gamma_\mu^2}{2}\cos(\omega_\mu t)\int_0^t d\tau\,(t-\tau)$$
$$\times \{\cos(\omega_\mu\tau)[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)] + \sin(\omega_\mu\tau)[\Phi_{xy}(\tau)-\Phi_{yx}(\tau)]\}$$
$$+\frac{\gamma_\mu^2}{2\omega_\mu}\cos(\omega_\mu t)\int_0^t d\tau\,\sin(\omega_\mu\tau)[\Phi_{yy}(\tau)-\Phi_{xx}(\tau)]$$
$$-\frac{\gamma_\mu^2}{2}\sin(\omega_\mu t)\int_0^t d\tau\,(t-\tau)$$
$$\times \{\sin(\omega_\mu\tau)[\Phi_{xx}(\tau)+\Phi_{yy}(\tau)] + \cos(\omega_\mu\tau)[\Phi_{yx}(\tau)-\Phi_{xy}(\tau)]\}$$
$$-\frac{\gamma_\mu^2}{2\omega_\mu}\sin(\omega_\mu t)\int_0^t d\tau\,\cos(\omega_\mu\tau)[\Phi_{yy}(\tau)-\Phi_{xx}(\tau)]\,. \qquad \text{(A2)}$$

We suppose that an expression of $P_x(t)$ valid in general is

$$P_x(t) = \exp[-\psi_x(t)]\cos[\omega_\mu t + \varphi(t)]\,.$$

Keeping only the first terms of its expansion we write

$$P_x(t) = [1 - \psi_x(t)] \cos(\omega_\mu t) - \varphi(t) \sin(\omega_\mu t) . \tag{A3}$$

This formula is valid if $\varphi(t) \ll 1$. The identification term by term of equation (A3) with equations (A1) and (A2) leads to the expressions for $\Omega(t)$, $\psi_x(t)$ and $\varphi(t)$ given in the main text.

References

Abragam A 1961 *The Principle of Nuclear Magnetism* (London: Oxford University Press)

Celio M 1986 *Phys. Rev. Lett.* **56** 2720

Celio M and Meier P F 1983 *Phys. Rev.* B **27** 1908

Chappert J and Grynszpan R I (eds) 1984 *Muon and Pions in Materials Research* (Amsterdam: North-Holland)

Chappert J and Yaouanc A 1986 *Microscopic Methods in Metals (Topics in Current Physics 40)* (Berlin: Springer)

Dalmas de Réotier P 1990 *PhD Thesis* Institut National Polytechnique de Grenoble unpublished

Dalmas de Réotier P, Yaouanc A and Boucher J P 1990 *Hyperfine Interact.* **65** 1121

Hartmann O 1977 *Phys. Rev. Lett.* **39** 832

Hayano R S, Uemura Y J, Imazato J, Nishida N, Yamazaki T and Kubo R 1979 *Phys. Rev.* B **20** 850

Heller P 1976 *Proc. Int. School of Physics Enrico Fermi, Course LIX* ed K A Müller and A Rigamonti (Amsterdam: North-Holland)

Kadono R, Kiefl R F, Kreitzman S R, Li Q, Pfiz T, Riseman T M, Zhou H, Wäppling R, Harris S W, Hartmann O, Karlsson E, Hempelmann R, Richter D, Niinikoski T O, Le L P, Luke G M, Sternlieb B and Ansaldo E J 1990a *Hyperfine Interact.* **64** 737

Kadono R, Matsuzaki T, Yamazaki T, Kreitzman S R and Brewer J H 1990b *Phys. Rev.* B **42** 6515

Kehr K W, Honig G and Richter D 1978 *Z. Phys.* B **32** 49

Kehr K W, Richter D, Welter J M, Hartmann O, Karlsson E, Norlin L O, Niinikoski T O and Yaouanc A 1982 *Phys. Rev.* B **26** 567

Kondo J 1986 *Hyperfine Interact.* **31** 117

Kubo R 1954 *J. Phys. Soc. Japan* **9** 935

Kubo R and Toyabe T 1966 *Magnetic Resonance and Relaxation* ed R Blinc (Amsterdam: North-Holland)

Luke G M, Brewer J H , Kreitzman S R, Noakes D R, Celio M, Kadono R and Ansaldo E J 1991 *Phys. Rev.* B **43** 3284

McMullen T and Zaremba E 1978 *Phys. Rev.* B **18** 3026

Moriya T 1962 *Prog. Theor. Phys. (Kyoto)* **28** 371

Schenck A 1985 *Muon Spin Rotation Spectroscopy* (Bristol: Adam Hilger)

Slichter C P 1963 *Principles of Magnetic Resonance* (New York: Harper and Row)

Yamada K 1984 *Prog. Theor. Phys.* **72** 195