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A NEW METHOD TO CALCULATE THE MUON POLARIZATION FUNCTION

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Abstract

In this paper we report on a new technique, developed to study the spin dynamics of a static positive muon interacting with neighboring nuclear spins. The method is based on the Trotter formula and leads to exact results even when the dimension of the composite spin system is very large. First results are presented for the case of muons localized at the interstitial octahedral site in copper in an arbitrary magnetic field.

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For the last few years, muon spin-relaxation processes in zero- and low-field have been the focus of much theoretical and experimental work in the field of μSR ^[1-8]. Theoretically these processes exhibit features which are very different from those in strong fields, as was first pointed out by Kubo and Toyabe^[1]. Their theory, based on the approximation of random local fields, found wide applications in μSR experiments. In particular zero-field measurements have been recently performed^[6,7] to investigate the diffusion and trapping of positive muons as a function of temperature and impurity concentration in various metals.

In a more rigorous approach, however, the internal dynamics of the nuclear spin system interacting with the muon has to be taken into account. In previous papers^[3-4] we have presented some results for the muon polarization function $P(t)$ which were obtained from a numerical solution of the full Hamiltonian describing the interaction of a muon with four or six nuclear spins ($I=1/2$ or 1) arranged in various spatial configurations. The extension of the theory to realistic cases (more nuclear spins or larger J values) is straightforward, but the numerical evaluation is prohibited by the very large size of the Hamiltonian matrices which have to be diagonalized. Therefore one generally looks for approximations, which are relevant for a particular experiment^[5,8]. In this article we show that it is possible to solve this class of problems exactly. Our method is based on the Trotter formula for bounded operators^[9-10] and on a random phase assumption about the initial state of the host spin system. Though we will consider here only the case of an interstitial μ^+ (spin $1/2$), the technique can be easily generalized to the case of different interstitial or substitutional particles.

The spin Hamiltonian under consideration is given by the sum of the dipolar interactions between the muon ($I=1/2$) and N neighboring nuclear spins J , the quadrupolar

interactions of the nuclear spins due to the field gradient produced by the muon, and the Zeeman terms

$$H = \sum_{j=1}^N (H_j^D + H_j^Q) + H_z \quad (1a)$$

with

$$H_z = \hbar \gamma_\mu \mathbf{I} \cdot \mathbf{B} - \sum_{j=1}^N \hbar \gamma_j \mathbf{J}_j \cdot \mathbf{B} \quad (1b)$$

$$H_j^D = \frac{\hbar^2 \gamma_\mu \gamma_j}{r_j^3} [\mathbf{I} \cdot \mathbf{J}_j - 3(\mathbf{I} \cdot \mathbf{n}_j)(\mathbf{J}_j \cdot \mathbf{n}_j)] \quad (1c)$$

$$H_j^Q = \hbar \omega_Q [(\mathbf{n}_j \cdot \mathbf{J}_j)(\mathbf{n}_j \cdot \mathbf{J}_j) - J(J+1)/3] \quad (1d)$$

$\mathbf{n}_j$ is the unit vector in direction from the μ^+ to the nucleus j at distance r_j . The dipolar interaction among the nuclear spins $\mathbf{J}_j$ can be neglected since it affects the results only for larger times provided that $\gamma_j \leq 0.1 \gamma_\mu$. [4]

We are interested in the time dependence of the muon polarization which is determined from

$$P(t) = \text{Tr} (\rho \exp(i\hbar t/\hbar) \sigma_\mu \exp(-i\hbar t/\hbar)) \quad (2)$$

where $\sigma_\mu = 2 \mathbf{I}_\mu$ are the Pauli matrices.

For a muon initially polarized along the z -axis and for unpolarized nuclei, the density matrix ρ is given by

$$\rho = \frac{1}{2(2J+1)^N} (\mathbf{1} + \sigma_\mu^z) \quad (3)$$

Since the individual Hamilton operators H_j^D do not commute, an analytical solution to Eq.(2) is, in general, not possible. A numerical solution can be found by calculating the eigenvalues

and eigenvectors of H , however this procedure can only be performed for cases where the dimension d of the composite Hilbert space ($d=2(2J+1)^N$) is not too large.

Here we want to solve Eq.(2) by using the generalized Trotter formula for bounded operators. For a set of n bounded operators A_j ($j=1, \dots, n$), one can show that [9]

$$\begin{aligned} \exp(A_1 + A_2 + \dots + A_n) \\ = \lim_{k \rightarrow \infty} (\exp(A_1/k) \cdot \exp(A_2/k) \dots \exp(A_n/k))^k \end{aligned} \quad (4)$$

where the integer k must be chosen large enough to ensure convergence. This formula can be applied directly to the solution of Eq.(2) by choosing a convenient decomposition of the total Hamiltonian H . In our case we rewrite H as

$$\begin{aligned} H &= \sum_{j=1}^N H_j \\ &= \sum_{j=1}^N \frac{1}{N} \hbar \gamma_\mu \mathbf{I} \cdot \mathbf{B} - \hbar \gamma_j \mathbf{J}_j \cdot \mathbf{B} + H_j^D + H_j^Q \end{aligned} \quad (5)$$

At this point it is important to note that the total Hamiltonian is represented by a $(d \times d)$ matrix whereas the single Hamiltonians H_j all act in small subspaces of dimension $2(2J+1)$. [11] This means that instead of diagonalizing a single but very large matrix H , one has just to diagonalize N small matrices H_j and multiply the results according to Eq.(4). The number of necessary multiplications which have to be carried out (equal to k) depends on the norm of the operators H_j , i.e. on the particular problem which is studied.

The computing storage problem in the numerical evaluation of Eq.(2) can be drastically reduced by working in the Schroedinger-picture, where the time dependence is contained in the wave functions (spinors represented by vectors of d components), rather than in the

Heisenberg-picture, where one has to deal with time-dependent operators ($d \times d$ matrices). Thus we choose the representation where σ_μ^z is diagonal and we rewrite Eq.(2) as

$$P(i) = \sum_{n=1}^d w_n \langle \psi_n(i) | \sigma_\mu^z | \psi_n(i) \rangle \quad (6)$$

where

$$\begin{aligned} |\psi_n(i)\rangle &= \exp(-iH_1/\hbar) |\psi_n(0)\rangle \\ &= \lim_{k \rightarrow \infty} (\exp(-iH_1/\hbar k) \cdot \exp(-iH_2/\hbar k) \dots \exp(-iH_N/\hbar k))^k |\psi_n(0)\rangle \end{aligned} \quad (7)$$

The coefficients w_n describe the probability of finding the spin system in the pure state $|\psi_n(0)\rangle$ at time zero, and they are determined according to the density matrix ρ (Eq.(3))

$$\begin{aligned} w_n &= 2/d & \text{if } \sigma_\mu^z |\psi_n(0)\rangle &= + |\psi_n(0)\rangle \\ w_n &= 0 & \text{if } \sigma_\mu^z |\psi_n(0)\rangle &= - |\psi_n(0)\rangle \end{aligned} \quad (8)$$

Given an initial spinor $|\psi_n(0)\rangle$ it is straightforward to solve the Eq.(7) using the Trotter formula and to compute the matrix element $\langle \psi_n(i) | \sigma_\mu^z | \psi_n(i) \rangle$. With increasing d , however, it would require too much computing time to carry out this operation for all possible $d/2$ initial states. This final problem can be avoided by making use of a procedure which looks very similar to the *random phase approximation*.^[12] We define an initial state $|\phi(0)\rangle$ of the system as the following linear combination

$$|\phi(0)\rangle = \sum_{m=1}^{d/2} (2/d)^{1/2} e^{i\lambda_m} |\psi_m(0)\rangle \quad (9)$$

where the sum goes over the $d/2$ possible initial states (i.e. those states for which $w_m = 2/d$), and where the λ_m are phases chosen at random in the interval $(0, 2\pi)$.

Physically, $|\phi(0)\rangle$ describes a state with the muon spin polarized along the $+z$ axis and randomly polarized nuclear spins. The Schrodinger equation is linear, so at a later time t we have

$$|\phi(t)\rangle = \sum_{m=1}^{d/2} (2/d)^{1/2} e^{i\lambda_m} |\psi_m(t)\rangle \quad (10)$$

The matrix element of σ_μ is then found to be

$$\begin{aligned} \langle \phi(t) | \sigma_\mu | \phi(t) \rangle &= \sum_{m=1}^{d/2} (2/d) \langle \psi_m(t) | \sigma_\mu | \psi_m(t) \rangle \\ &+ \sum_{\substack{m,n=1 \\ (m \neq n)}}^{d/2} (2/d) e^{i(\lambda_m - \lambda_n)} \langle \psi_n(t) | \sigma_\mu | \psi_m(t) \rangle \end{aligned} \quad (11)$$

The first term on the right side of Eq.(11) is equal to the exact solution (Eq.(6)), and the second term can be considered as a small random correction which disappears in the limit of very large d , where neighbouring terms interfere destructively.^[12] In order to really check the magnitude of this term, one has just to calculate the matrix element (11) for various random initial states; the average over these results will approach the exact solution, whereas the differences between the single results will be a measure of the error associated with using an arbitrary initial state for the host spins. A major advantage of this procedure is that the number of simulations decreases with increasing d ; for example we found 4 simulations sufficient in the case $d=8192$.

One can imagine a wide variety of problems which can be solved using the method described above. In this paper we present some results for the case of a muon in copper,

because so much experimental work has been done on this material in the last few years^[6,7]. We assume that the muon is located at an octahedral interstitial site in the copper lattice and interacts with 6 neighboring nuclear spins $J=3/2$ according to the Hamiltonian of Eq.(1). For a μ^+ and a Cu nucleus at a distance of half the lattice constant one has $\omega_D = \hbar^2 \gamma_\mu \gamma_I / r^3 = 0.11 \mu s^{-1}$, and $\omega_Q = 3.2 \mu s^{-1}$ ^[13]. Following the procedure described in the first part of this letter we calculated the muon polarization function $P(t)$ in zero external field and for various applied longitudinal magnetic fields.

The results are shown in Figs.1 and 2. The ZF function of Fig.1 can be compared with both the Kubo-Toyabe theory and with the ZF function obtained by Holzschuh and Meier^[5] in the limit of very strong quadrupolar coupling. The difference between the two quantum-mechanical calculations is negligible, whereas both these results are found to deviate strongly from the Kubo-Toyabe behavior. In particular there is no recovery of the polarization to the asymptotic value $1/3$ but rather $P(t)$ stays oscillating. Our result was obtained by neglecting the effect of the nuclear spins beyond the first shell. As shown before^[5], however, this effect is practically negligible for $t < 15 \mu s$. In Fig.1, together with the ZF result, the muon polarization function $P(t)$ is shown for the case of an external magnetic field of 10 G and 30 G applied in the direction parallel to the initial muon polarization (which here was assumed to be along the $\langle 111 \rangle$ crystal axis). These results clearly show the decoupling effect caused by the external field which progressively reduces the local field fluctuations seen by the muon.

Recently a new type of μSR experiment suggested by Abragam^[14], the level-crossing-resonance (LCR) technique, was performed in copper.^[15] Here one employs the longitudinal-field configuration and, by adjusting the field strength, matches the energy splitting of the muon's Zeeman levels to the magnetic levels of the copper nuclei, which are determined mainly by their quadrupolar energies. This resonance condition is roughly given by

$\gamma_\mu B \approx 2 \omega_Q$; in terms of magnetic fields this means $B_{RES} \approx 2 \omega_Q / \gamma_\mu \approx 80$ G. Fig. 2 shows some results obtained using our technique in the LCR region. In particular we calculated $P(t)$ for applied longitudinal fields of 70 G, 80 G and 100 G along the $\langle 111 \rangle$ crystal orientation. These results give specific information on the first n.n. shell because the effect of the further shells is completely negligible at the LCR. Further we point out that all these results (except the ZF-function) are slightly orientation dependent. The introduction of this new method will greatly facilitate the analysis of ZF-, LF- and μLCR -results in terms of fundamental microscopic parameters instead of phenomenological quantities. The generalization of the static results to the dynamical case is well-known and can be carried out for example by solving the strong collision model^[2].

In conclusion, we have presented a powerful method which allows one to exactly calculate the time evolution of the muon polarization function in a crystal. This approach should lead to a better understanding of the physics of positive muons, and might be generally applicable to describe different interstitial or substitutional particles in solids.

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Figure captions

Fig. 1. Polarization function for a muon at the octahedral site in copper in ZF and for longitudinal fields of 10 G and 30 G applied along the $\langle 111 \rangle$ axis.

Fig. 2. Polarization function for a muon at the octahedral site in copper for longitudinal fields of 70 G, 80 G and 100 G applied along the $\langle 111 \rangle$ axis. For comparison the ZF-function is shown again as a dashed line. The curve for 80 G shows clearly the relaxation due to the level-crossing resonance.

Fig. 1

Fig. 2