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Zero-field spin relaxation of positive muons

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The spin dynamics of a muon interacting with neighboring nuclear spins in the presence of a strong quadrupolar coupling is investigated. An approximation for calculating the time-dependent muon polarization is presented which works equally well for integer and half-odd-integer spins J. In the particular case of equivalent nuclei with $J=\frac{3}{2}$, an analytic solution is given. The influence of nuclei in more-distant shells is discussed, and the change of the polarization with muon diffusion is calculated.

I. INTRODUCTION

Spin-relaxation processes in zero or weak external magnetic fields exhibit characteristic features quite different from those in strong fields. This was first pointed out by Kubo and Toyabe.[1] Their theory, based on the approximation of random local fields, found wide applications in muon-spin-rotation experiments. Hayano *et al.*[2] have shown that zero-field experiments are particularly suitable to investigate the diffusion properties of muons. Recently Clawson *et al.*[3] reported on zero-field measurements in copper. Analyzing their data in terms of the Kubo-Toyabe theory they found an increase of the muon hopping rate with decreasing temperature below 5 K.

In a more rigorous approach, however, the internal dynamics of the nuclear-spin system interacting with the muon (which cannot be described by local fields) has to be taken into account. In previous papers[4,5] 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 dipole interaction of a muon with four to six nuclear spins $J=\frac{1}{2}$ or 1. The results showed marked differences from those of the Kubo-Toyabe theory in the long-time behavior. Though the extension of the theory to more nuclear spins or larger J values is straightforward, the numerical evaluation is prohibited by the very large size of the Hamiltonian matrices to be diagonalized, and one must look for approximations relevant for experiments.

The presence of the muon induces a radial electrical field gradient leading to an interaction with the quadrupole moments of the nuclei. In the limit of strong quadrupole interaction, a common situation in experiments, Petzinger and Wei[6] gave an approximate treatment, valid for integer spins J. As shown earlier,[7] however, this method is not applicable for half-integer values of J, and it is the purpose of the present article to treat this case appropriately. In particular, we present numerical results for $J=\frac{3}{2}$ spins arranged in various geometries. To make the results available for analyzing experimental data, we fitted the calculated polarization $p(t)$ to a simple analytic function with few parameters.

II. THEORY

The spin Hamiltonian under consideration is given by the sum of the dipolar interactions between the muon $(I=\frac{1}{2})$ and N neighboring nuclear spins J and the quadrupole interactions of the nuclear spins due to field gradients produced by the muon,

$$H=\sum_{k=1}^{N} H_k \ , \tag{1}$$

with

$$H_k=H_k^D+H_k^Q \ , \tag{2}$$

where

$$H_k^D=\hbar\omega_k^D[\vec{I}\cdot\vec{J}_k-3(\vec{n}_k\cdot\vec{I}\,)(\vec{n}_k\cdot\vec{J}_k)] \tag{3}$$

and

$$H_k^Q=\hbar\omega_k^Q[(\vec{n}_k\cdot\vec{J}_k)(\vec{n}_k\cdot\vec{J}_k)-\tfrac{1}{3}J(J+1)] \ . \tag{4}$$

$\vec{n}_k$ is the unit vector in direction from the μ^+ to the nucleus k at distance r_k, and ω_k^D is given by[8]

$$\omega_k^D=\frac{\hbar\gamma_\mu\gamma_J}{r_k^3} \ . \tag{5}$$

In Eq. (4) it is assumed that the field gradient due to the muon is axially symmetric around $\vec{n}_k$ and that there are no other field gradients at the nuclei.

For the nuclear spins J_k we define the projection operators P_{k,m_k} which project out the $2J+1$ substates m_k in the following way:

$$P_{k,m_k} = | m_k \rangle \langle m_k | \ , \tag{6}$$

where

$$(\vec{n}_k \cdot \vec{J}_k) | m_k \rangle = m_k | m_k \rangle \ . \tag{7}$$

The contribution H_k to the Hamiltonian is now rewritten as

$$\begin{aligned} H_k &= \sum_{m_k} P_{k,m_k} H_k \sum_{m_k'} P_{k,m_k'} \\ &= \sum_{m_k} P_{k,m_k} H_k P_{k,m_k} + \sum_{\substack{(m_k \neq m_k') \\ m_k, m_k'}} P_{k,m_k} H_k^D P_{k,m_k'} \ . \end{aligned} \tag{8}$$

The quadrupole interaction, which is diagonal in the representation defined by Eq. (7), occurs in the first term only. The second term involves transitions between the levels of the quadrupole Hamiltonian induced by the dipolar interaction. If ω^Q is much larger than ω^D the corresponding off-diagonal matrix elements may be neglected when they connect quadrupole levels with different quadrupole energies. Since the dipolar part H_k^D has vanishing matrix elements between states with $| m_k - m_k' | > 1$ the neglect of the second term in Eq. (8) can be justified for all m_k except $m_k = \pm\frac{1}{2}$.

Therefore we define

$$Q_k = P_{k,1/2} + P_{k,-1/2} \ , \tag{9}$$

and rewrite Eq. (8) as

$$H_k = \sum_{m_k (\neq \pm 1/2)} P_{k,m_k} H_k P_{k,m_k} + Q_k H_k Q_k + R_k \ , \tag{10}$$

where the remainder is

$$\begin{aligned} R_k = &\sum_{m_k (\neq \pm 1/2)} P_{k,m_k} H_k^D Q_k + Q_k H_k^D \sum_{m_k (\neq \pm 1/2)} P_{k,m_k} \\ &+ \sum_{\substack{m_k, m_k' (\neq 1/2) \\ (m_k \neq m_k')}} P_{k,m_k} H_k^D P_{k,m_k'} \ . \end{aligned} \tag{11}$$

In the limit of strong quadrupole energy we can neglect R_k since it involves dipole transitions between quadrupole levels separated by energies of order ω^Q or is zero for the degenerate cases where $m_k = -m_k'$.

We denote the truncated Hamiltonian by $\overline{H}_k$,

$$\overline{H}_k = \sum_{m_k (\neq \pm 1/2)} P_{k,m_k} H_k P_{k,m_k} + Q_k H_k Q_k \ , \tag{12}$$

which can be written as

$$\overline{H}_k = \sum_{m_k (\neq \pm 1/2)} A_{k,m_k} + Q_k H_k Q_k \ , \tag{13}$$

with

$$\begin{aligned} A_{k,m_k} = \hbar \{ &-2\omega_k^D (\vec{n}_k \cdot \vec{I}\,) m_k \\ &+ \omega_k^Q [m_k^2 - \tfrac{1}{3} J(J+1)] \} P_{k,m_k} \ . \end{aligned} \tag{14}$$

In the case of a single nuclear spin ($N=1$) where an analytic solution is possible it has been shown[7] that this approximation leads to errors in the eigenvalues of the order of $(\omega^D/\omega^Q)^2$ which can be safely neglected in the calculation of the muon polarization function for the times of interest. To further check the approximation[9] we have compared the numerical solutions of the full and the truncated problem for $N=2$ and again found complete agreement for times t which fulfill $\omega^D t < 2$ if $\omega^Q > 20\omega^D$.

An inspection of Eqs. (13) and (14) shows that the truncated Hamiltonian is the direct sum of three parts which operate on spaces of dimensions 2×1, 2×1, and 2×2, each of which contains the quadrupole energy ω_k^Q as an additive constant, which therefore will be omitted in the following. For N nuclear spins the state space is thus reduced to invariant subspaces with the largest dimension being 2×2^N.

For the particular case of $J=\frac{3}{2}$ the problem can be further reduced. Noting that

$$Q_k (\vec{n}_k \cdot \vec{J}_k) Q_k = \tfrac{1}{2} \vec{n}_k \cdot \vec{\sigma}_k \tag{15}$$

and

$$Q_k (\vec{n}_k \times \vec{J}_k) Q_k = \vec{n}_k \times \vec{\sigma}_k \ , \tag{16}$$

where $\vec{\sigma}_k$ are Pauli matrices, we can write

$$Q_k H_k^D Q_k = \hbar\omega_k^D [\vec{I} \cdot \vec{\sigma}_k - 2(\vec{n}_k \cdot \vec{I}\,)(\vec{n}_k \cdot \vec{\sigma}_k)] \ . \tag{17}$$

By a unitary transformation of $\overline{H}_k$,

$$\widetilde{H}_k = (\vec{n}_k \cdot \vec{\sigma}_k) \overline{H}_k (\vec{n}_k \cdot \vec{\sigma}_k) \ , \tag{18}$$

we obtain

$$\widetilde{H}_k = \sum_{m_k (\neq \pm 1/2)} A_{k,m_k} - \hbar\omega_k^D \vec{I} \cdot \vec{\sigma}_k \ , \tag{19}$$

and are thus left with an isotropic interaction.

The total Hamiltonian can now be written as

$$\widetilde{H} = -\hbar \vec{I} \cdot \sum_{k=1}^{N} (\vec{a}_{k,3/2} \oplus \vec{a}_{k,-3/2} \oplus \vec{\sigma}_k) \omega_k^D \ , \tag{20}$$

where the direct sum refers to the four-dimensional subspace of nucleus k. The quantities

$$\vec{a}_{k,m_k} = 2 m_k \vec{n}_k \tag{21}$$

can be considered as effective fields produced by the dipolar interactions from the nuclear-spin states $m_k = \pm\frac{3}{2}$, whereas the internal dynamics of the states $m_k = \pm\frac{1}{2}$ has been transformed into the effective spin-$\frac{1}{2}$ interaction described by the spin variables $\vec{\sigma}_k$.

In this way the Hamiltonian is partitioned into the direct sum of 3^N contributions,

$$\tilde{H} = \bigoplus_{j=1}^{3^N} \tilde{H}_j \;, \tag{22}$$

where the spin space on which $\tilde{H}_j$ acts has dimension 2×2^{h_j} where h_j ($h_j=0,1,\ldots,N$) denotes the number of $\vec{\sigma}_k$'s occurring in the partition j.

If all N nuclei have the same dipolar interaction frequency ω^D, each partial Hamiltonian is of the form

$$\tilde{H}_j = -\hbar\omega^D[\vec{\mathrm{I}}\cdot(\vec{\sigma}_{k_1}+\vec{\sigma}_{k_2}+\cdots+\vec{\sigma}_{k_h})+\vec{\mathrm{I}}\cdot\vec{\mathrm{B}}_j] \;, \tag{23}$$

where $\vec{\mathrm{B}}_j$ is an effective field produced by the $N-h_j$ vectors $\vec{\mathrm{a}}_{k,\pm 3/2}$ determined by the partition. By combining the h_j effective spins $\frac{1}{2}$ into a total spin

$$\vec{\mathrm{G}}_j = \tfrac{1}{2}(\vec{\sigma}_{k_1}+\vec{\sigma}_{k_2}+\cdots+\vec{\sigma}_{k_h}) \;, \tag{24}$$

a further reduction is possible,

$$\tilde{H}_j = \bigoplus_{F_j} g_{F_j}\tilde{H}_{j,F_j} \;, \tag{25}$$

since the 2^{h_j}-dimensional product representation induced by Eq. (24) is reducible. F_j takes the value $h_j/2$, $h_j/2-1,\ldots,0$ or $\frac{1}{2}$, and g_{F_j} are the multiplicities occurring in the decomposition. The Hamiltonians $\tilde{H}_{j,F_j}$ are given by

$$\tilde{H}_{j,F_j} = -2\hbar\omega^D\vec{\mathrm{I}}\cdot\vec{\mathrm{F}}_j - \hbar\omega^D\vec{\mathrm{I}}\cdot\vec{\mathrm{B}}_j \;. \tag{26}$$

The solution of this eigenvalue problem is straightforward. In the Appendix we give the eigenvalues together with the corresponding contribution $\vec{\mathrm{p}}_{j,F_j}(t)$ to the muon polarization $\vec{\mathrm{p}}(t)$.

According to Eqs. (18), (22), and (25) we thus write

$$\vec{\mathrm{p}}(t) = \sum_{j=1}^{3^N}\sum_{F_j} g_{F_j}\vec{\mathrm{p}}_{j,F_j}(t) \;, \tag{27}$$

where

$$\vec{\mathrm{p}}_{j,F_j}(t) = \mathrm{Tr}_{F_j}(\rho e^{i\tilde{H}_{j,F_j}t}\,2\vec{\mathrm{I}}\,e^{-i\tilde{H}_{j,F_j}t}) \;. \tag{28}$$

The trace is taken over the subspace of the spin operators $\vec{\mathrm{I}}$ and $\vec{\mathrm{F}}_j$, and ρ is the density matrix,

$$\rho = \frac{1}{2(2J+1)^N}[1+2\vec{\mathrm{I}}\cdot\vec{\mathrm{p}}(0)] \;. \tag{29}$$

We thus have obtained an analytic solution for the muon polarization in the case of N equivalent spin-$\frac{3}{2}$ nuclei valid in the limit of strong quadrupole interaction.

III. RESULTS

In this way we have calculated the muon polarization function for a μ^+ interacting with $N=4$, 6, and 8 nuclei with $J=\frac{3}{2}$ in a tetrahedral, octahedral, and cubic arrangement, respectively. The results are shown in Fig. 1. It is seen that with increasing N the oscillations are reduced and the minimum is shifted to shorter times. The latter effect is simply a consequence of an increasing second moment M which can be calculated exactly (see, e.g., Ref. 5). For the tetrahedral, octahedral, and cubic arrangement one obtains $M=16\times\omega_D^2$, $24\times\omega_D^2$, and $32\times\omega_D^2$, respectively.

In experiments the muon is sitting at an interstitial lattice site, and we therefore have to consider the effect of the nuclear spins beyond the first shell. Our theory can easily be applied to several shells. Considering N_1 nuclei with spin $J=\frac{3}{2}$ on a first shell with dipolar frequency ω_1 and N_2 on a second shell with ω_2, we obtain[10] a partitioning into $3^{N_1+N_2}$ Hamiltonians $\tilde{H}_j$ of the form

$$\tilde{H}_j = -\hbar\omega_1(2\vec{\mathrm{I}}\cdot\vec{\mathrm{G}}_1+\vec{\mathrm{I}}\cdot\vec{\mathrm{B}}_1)-\hbar\omega_2(2\vec{\mathrm{I}}\cdot\vec{\mathrm{G}}_2+\vec{\mathrm{I}}\cdot\vec{\mathrm{B}}_2) \;, \tag{30}$$

where the maximum values of G_1 and G_2 are $N_1/2$ and $N_2/2$. This three-spin problem, however, is no longer analytically solvable.

Since the contribution of further shells to the second moment, which is additive, is small we expect them to cause an appreciable change of $p(t)$ only at large times. This justifies the use of an additional approximation in treating the further shells. An analytic solution is still possible by truncating completely, for further shells, the second term in Eq. (8), which involves transitions between $m_k \neq m_k'$ levels. The Hamiltonian can then be reduced to expressions of the form of Eq. (26) with modified effective fields.

We have calculated $p(t)$ in this way for a muon at the octahedral site in a fcc lattice interacting with nuclei of spin $J=\frac{3}{2}$ on two shells. An estimate of the error induced by this further approximation can be obtained by comparing the correct second moments of the first shell, $M_1=24\omega_1^2$, of the second shell, $M_2=(96/81)\omega_1^2$, and the value for the approximation $M_2'=(80/81)\omega_1^2$.

The additional fields produced by the eight nuclei on the second shell increases the number N_f of different frequencies drastically ($N_f\sim 4\times 10^4$). A histogram of the amplitudes as a function of frequency is shown in Fig. 2. This may be compared to the frequency distribution

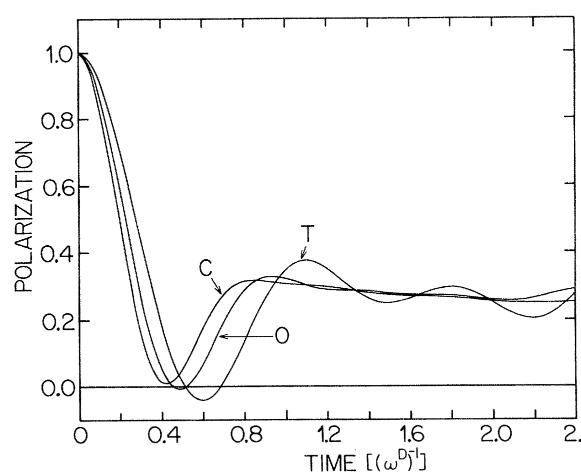

FIG. 1. Polarization function for a muon interacting with $N=4$, 6, or 8 nuclei with $J=\frac{3}{2}$ in a tetrahedral (T), octahedral (O), or cubic (C) arrangement, respectively. Time in units of $1/\omega^D$.

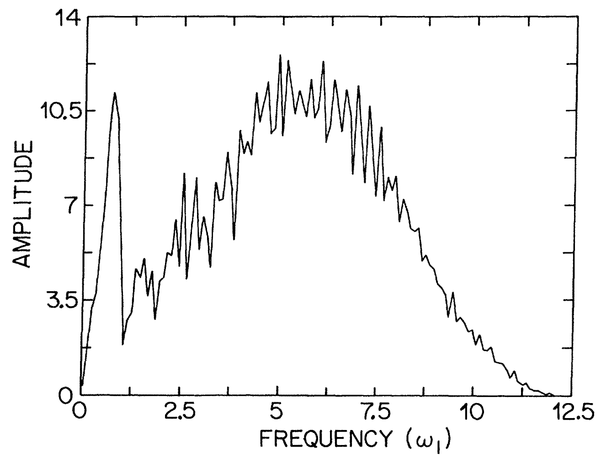

FIG. 2. Histogram of the unnormalized amplitudes vs frequency for the polarization of a muon at the octahedral site in a fcc lattice interacting with $J=\frac{3}{2}$ nuclei on the first- and second-neighbor shells. Frequency in units of ω_1 and bin size is 0.1.

$$A(\omega)\propto\omega^2\exp\left[-\frac{\omega^2}{2\Delta^2}\right], \tag{31}$$

which was assumed by Kubo and Toyabe[1] (KT), and which leads to the polarization function

$$p_{\rm KT}(t)=\frac{1}{3}+\frac{2}{3}(1-\Delta^2t^2)e^{-\Delta^2t^2/2}, \tag{32}$$

where Δ^2 is half of the second moment.

In Fig. 3 we compare the polarization functions calculated for the first- and second-nearest-neighbor shells. It is seen that the changes in $p(t)$ introduced by the additional interaction of the second shell are small. The change for $t<0.7/\omega_1$ is due to the increase of the second moment by about 4% and the characteristic maximum at $t\sim0.92/\omega_1$ is barely affected. For larger times the small oscillations are reduced. A significant deviation is seen for times larger than $2.1/\omega_1$, but for this time regime anyway, we expect that corrections from the dipole interaction among the nuclei and from the quadrupole levels $m_k=\pm\frac{3}{2}$ will show up.

TABLE I. Parameter values for Eq. (33). B and Ω_i in units of ω_1.

A_1	0.24121	Ω_1	7.60957
A_2	0.37634	Ω_2	4.90900
A_3	0.11859	Ω_3	0.81610
B	1.65	C	0.26459

To make the results available for further applications we found it convenient to fit $p(t)$ to the following analytic function:

$$p(t)=C+\sum_{i=1}^{3}A_i\cos(\Omega_it)e^{-B^2t^2/2}. \tag{33}$$

The eight parameters for this equation are given in Table I. This expression fits the calculated function for $t<2/\omega_1$ with a root-mean-square deviation of 1.3×10^{-3} and a maximal deviation of 3×10^{-3} occurring at large times.

In Fig. 4 we compare our results for a muon at the octahedral site in a fcc lattice with the Kubo-Toyabe function, Eq. (32), with Δ chosen to produce the same second moment. Clearly, $p_{\rm KT}(t)$ is a monotonically increasing function for $t>0.5/\omega_1$ and does not exhibit the characteristic maximum at $0.92/\omega_1$. This difference may be relevant when diffusion of the muon is considered.

In the strong-collision model[2] the dynamic muon polarization function $p(\nu,t)$ is determined by the integral equation

$$p(\nu,t)=e^{-\nu t}p(t)+\nu\int_0^t dt'p(\nu,t-t')e^{-\nu t'}p(t'), \tag{34}$$

where ν is the average hopping rate and $p(t)$ is the static

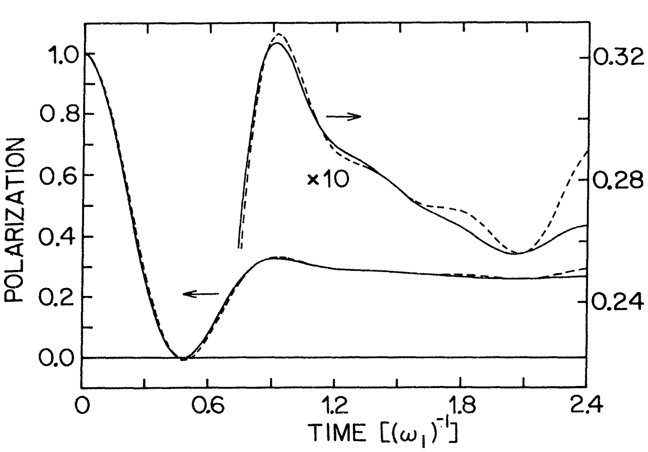

FIG. 3. Polarization functions for a muon at the octahedral site in a fcc lattice interacting with $J=\frac{3}{2}$ nuclei on the first- (dashed line) and the first- and second- (solid line) nearest-neighbor shells. For $t>0.8$ the curves are shown in the upper part of an enlarged scale. Time in units of $1/\omega_1$.

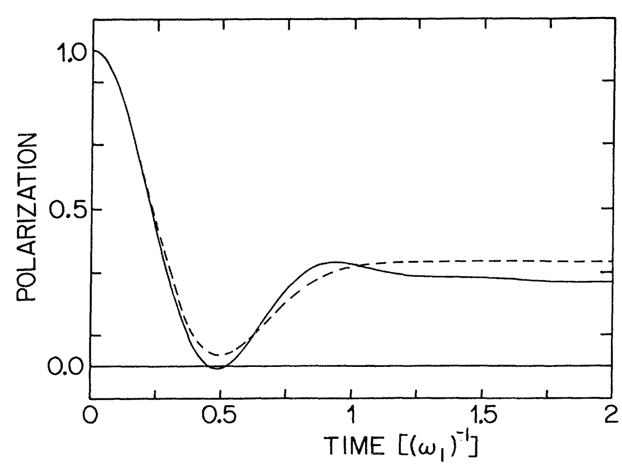

FIG. 4. Comparison of the Kubo-Toyabe function (dashed line) with the polarization of a muon at the octahedral site in a fcc lattice interacting with $J=\frac{3}{2}$ nuclei on the first- and second-neighbor shells. Time units of $1/\omega_1$. Parameter Δ in Eq. (32) has been chosen such that both curves have the same second moment, $M=24+80/81$.

function. Although this Volterra equation of the second kind could be solved by Laplace transformation, the awkward inverse transformation makes a direct numerical solution by discretization much more convenient.

In Fig. 5, $p(\nu,t)$ is shown for several values of ν as calculated from Eq. (34) using the parametrized form (33) for the static function $p(t)$. It is see that the maximum at around $t=0.9/\omega_1$ is gradually reduced with increasing ν. This is in contrast to the dynamic Kubo-Toyabe theory[2] where a maximum is built up for small but nonzero ν and does not show up in the static limit.

In conclusion we have presented an approximation for calculating the muon polarization function in zero external field which gives good results in the experimentally relevant case of strong quadrupole interaction. For N equivalent nuclei with $J=\frac{3}{2}$ an analytic solution is possible which shows characteristic oscillations depending on the geometrical arrangement on the first shell. Inclusion of further shells influences the results only slightly for times of experimental relevance. There is no universal function like that proposed by Kubo and Toyabe which accurately describes the muon polarization in zero field. This should be considered when zero-field data are used to determine the muon diffusion rate.

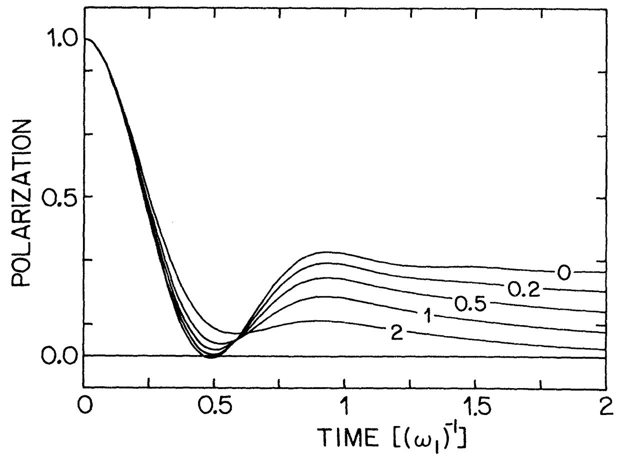

FIG. 5. Dynamic polarization functions obtained from the solution of the integral equation (34) for hopping rates $\nu=0.2$, 0.5, 1, or $2\times\omega_1$ as indicated. Curve $\nu=0$ is the static function given by Eq. (33). Time in units of $1/\omega_1$.

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APPENDIX

The eigenvalues of the Hamiltonian

$$H=\omega(\vec{\mathrm{I}}\cdot\vec{\mathrm{F}}+\vec{\mathrm{I}}\cdot\vec{\mathrm{B}}) \qquad \text{(A1)}$$

are given by

$$\lambda_m^{\pm}=-\frac{\omega}{4}\pm\frac{\omega}{2}[B^2+B(2m-1)+F(F+1)+\tfrac{1}{4}]^{1/2}\ , \qquad \text{(A2)}$$

where $m=-F+1,\ldots,+F$ and by

$$\lambda_{F+1}^{+}=(\omega/2)(F+B)\ , \qquad \text{(A3)}$$

$$\lambda_{-F}^{-}=(\omega/2)(F-B)\ . \qquad \text{(A4)}$$

It is convenient to introduce the angles α_m $(0\le\alpha_m<\pi/2)$ for $m=-F,\ldots,F+1$ by

$$\tan(2\alpha_m)=\frac{[F(F+1)-m(m-1)]^{1/2}}{m-\frac{1}{2}-B}\ . \qquad \text{(A5)}$$

The muon polarization parallel to $\vec{\mathrm{B}}$ is then given by[11]

$$p^{\parallel}(t)=\frac{1}{2F+1}\left\{1+\sum_{m=-F+1}^{F}[\cos^2(2\alpha_m)+\sin^2(2\alpha_m)\cos(\lambda_m^{+}-\lambda_m^{-})t]\right\}\ , \qquad \text{(A6)}$$

whereas for the perpendicular case one has

$$p^{\perp}(t)=\frac{1}{2F+1}\sum_{m=-F}^{F}[\cos^2\alpha_{m+1}\sin^2\alpha_m\cos(\lambda_{m+1}^{+}-\lambda_m^{+})t+\cos^2\alpha_{m+1}\cos^2\alpha_m\cos(\lambda_{m+1}^{+}-\lambda_m^{-})t$$
$$+\sin^2\alpha_{m+1}\sin^2\alpha_m\cos(\lambda_{m+1}^{-}-\lambda_m^{+})t+\sin^2\alpha_{m+1}\cos^2\alpha_m\cos(\lambda_{m+1}^{-}-\lambda_m^{-})t]\ . \qquad \text{(A7)}$$

For a muon initially polarized in direction $\vec{\mathrm{m}}$ one thus obtains the polariation along $\vec{\mathrm{m}}$ from

$$p^m(t)=\cos^2\theta\, p^{\parallel}(t)+\sin^2\theta\, p^{\perp}(t)\ , \qquad \text{(A8)}$$

where θ is the angle between $\vec{\mathrm{m}}$ and $\vec{\mathrm{B}}$.

The normalization of p^m has been chosen such that $p^m(0)=1$. Since each Hamiltonian $\tilde{H}_{j,F_j}$ in Eq. (26) has the form of Eq. (A1), the contributions $p_{j,F_j}^m(t)$ [see Eq. (27)] are given by

$$p_{j,F_j}^m=\frac{2F+1}{2(2J+1)^N}p^m(t)\ . \qquad \text{(A9)}$$

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[6]K. G. Petzinger and S. H. Wei, in *Proceedings of the Yamada Conference,* Ref. 5.

[7]P. F. Meier, in *Proceedings of the Yamada Conference,* Ref. 5.

[8]For a muon and a Cu nucleus at a distance of half the lattice constant (octahedral site) one has $\omega^D=0.11\ \mu s^{-1}$, whereas $\omega^Q=3.2\ \mu s^{-1}$.

[9]Preliminary results of an exact calculation for $N=4$ (tetrahedral case) further confirm the validity of the approximation [M. Schillaci and P. F. Meier (unpublished)].

[10]It is assumed that the quadrupole interaction exceeds the dipolar one also for the second shell.

[11]Knowing the eigenvalues and eigenfunctions of the Hamiltonian [Eq. (A1)] the calculation of the muon polarization functions for the parallel and perpendicular cases is straightforward. See Ref. 7 where a similar problem has been solved.