# Spin relaxation of positive muons due to dipolar interactions

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The spin dynamics of a polarized muon at an interstitial lattice site interacting with four or six neighboring nuclear spins is investigated. The difference from the results of the Kubo-Toyabe theory is pointed out and the range of validity of the secular approximation discussed.

Spin-relaxation processes in zero or weak external magnetic fields exhibit characteristic features which are very different from those in strong fields. This was first pointed out by Kubo and Toyabe.[1] Their theory, which is based on the approximation of random local fields, has recently been applied to muon spin rotation experiments. Hayano *et al.*[2] have shown that zero-field experiments are particularly suitable to investigate the diffusion properties of muons. For a static muon the Kubo-Toyabe theory predicts that the polarization in zero field decreases with a Gaussian behavior from its initial value of 1, passes through a minimum, and asymptotically reaches the value of $\frac{1}{3}$. This recovery to one-third is characteristic for a static situation. Even for a slowly diffusing muon the increase of the polarization to $\frac{1}{3}$ is significantly suppressed. Zero-field $\mu$SR experiments are currently being performed[3–6] to investigate the diffusion and trapping[7] of positive muons as a function of temperature and impurity concentrations in various metals.

In this Report we would like to present some results of numerical calculations of the muon polarization function which were obtained by solving the full dynamics of the spin system. The difference from the results of the Kubo-Toyabe theory, in which the influence of the surrounding nuclear spins is approximated by a random field, is pointed out. Furthermore, the range of validity of the secular approximation is investigated.

The spin Hamiltonian under consideration is given by the sum of the dipolar interactions between the muon ($I=\frac{1}{2}$) and $N$ neighboring spins ($J_j$) and of the Zeeman terms[8]:

$$H=\sum_{j=1}^{N} H_j + H_z \quad , \tag{1}$$

where

$$H_z=\hbar\gamma_\mu \vec{I}\cdot\vec{B}-\sum_{j=1}^{N}\hbar\gamma_J \vec{J}_j\cdot\vec{B} \tag{2}$$

and

$$H_j=\frac{\hbar^2\gamma_\mu\gamma_J}{r_j^3}[\vec{I}\cdot\vec{J}_j-3(\vec{I}\cdot\vec{n}_j)(\vec{J}_j\cdot\vec{n}_j)] \quad . \tag{3}$$

$\vec{n}_j$ is the unit vector in direction from the $\mu^+$ to the nucleus $j$ at distance $r_j$. We are interested in the time dependence of the muon polarization which is determined from

$$\vec{P}_\mu(t)=\mathrm{Tr}[\rho\exp(iHt/\hbar)\sigma_\mu\exp(-iHt/\hbar)] \quad , \tag{4}$$

where $\vec{\sigma}_\mu=2\vec{I}_\mu$ are the Pauli matrices. For a muon with initial polarization $\vec{P}_\mu(0)$ and for unpolarized nuclei, the density matrix $\rho$ is given by

$$\rho=\frac{1}{2(2J+1)^N}[1+\vec{P}_\mu(0)\,\vec{\sigma}_\mu] \quad . \tag{5}$$

Since the individual Hamiltonian operators $H_j$ do not commute, an analytical solution to Eq. (4) is, in general, not possible. Some exact statements, however, can be made in certain limiting cases.

In zero field the initial decay of the muon polarization is given by

$$\vec{P}_\mu(t)=\vec{P}_\mu(0)\,e^{-M_s t^2/2} \quad , \tag{6}$$

and $M_s$ can be calculated exactly. Using Eqs. (1), (3), and (4) one gets

$$M_s=\frac{1}{3}J(J+1)\sum_{j=1}^{N}\left(\frac{\hbar\gamma_\mu\gamma_J}{r_j^3}\right)^2(5-3\cos^2\theta_j) \quad , \tag{7}$$

where $\theta_j$ is the angle between the direction vector from the $\mu^+$ to nucleus $j$ and $\vec{P}_\mu(0)$. The polycrystalline average of $M_s$ is

$$\Delta_0^2=M_0=\frac{4}{3}J(J+1)\sum_{j=1}^{N}\left(\frac{\hbar\gamma_\mu\gamma_J}{r_j^3}\right)^2 \quad . \tag{8}$$

To simplify the calculation of the full time dependence of the muon spin polarization, the Kubo-Toyabe theory has been used. This is equivalent to approximating the spin operators $J_j$ of the surroundings nuclei by $c$ numbers:

$$\sum_{j=1}^{N} H_j=\hbar\gamma_\mu\vec{I}\cdot\vec{B}_D \quad . \tag{9}$$

 

The dipolar field $\vec{B}_D$ is assumed to be isotropic with random strength distributed according to a Gaussian function with width $\Delta$:

$$\Delta^2 = \tfrac{1}{3}\gamma_\mu^2 \langle \vec{B}_D^2 \rangle \ . \tag{10}$$

This leads to the Kubo-Toyabe function[2] in zero field:

$$P_\mu^{\rm KT}(t) = \tfrac{1}{3} + \tfrac{2}{3}(1-\Delta^2 t^2)\exp(-\tfrac{1}{2}\Delta^2 t^2) \ . \tag{11}$$

The question then arises in which cases the use of the Kubo-Toyabe theory can be justified. The approximation (9) seems to be adequate if the nuclei experience some other interaction (e.g., quadrupole effects) which dominates the dipolar term (3). With the interaction terms (1)–(3) alone, however, $P(t)$ behaves differently from Eq. (11).

To demonstrate this we have solved Eq. (4) numerically for a muon interacting with spin $J=\frac{1}{2}$ nuclei[9] by calculating the $2\times(2J+1)^N$ eigenvalues and eigenvectors of the Hamiltonian operator $H$ and evaluating[10,11] the trace in the basis of eigenvectors of $H$. The result for the case of a muon located at the tetrahedral interstitial site in a fcc lattice, interacting with four nearest neighbors ($N=4$) is shown in Fig. 1 for the muon polarization along the $\langle 100\rangle$ direction. The long-time behavior is very different from the Kubo-Toyabe function. In particular, there is no recovery to $\frac{1}{3}$ but rather $P_\mu$ stays oscillating. This is due to the internal dynamics of the nuclear spins which is suppressed in the treatment of random local fields.

The same behavior is also found for a muon at the octahedral site with six nearest neighbors (Fig. 2) where in the $\langle 100\rangle$ direction $P_\mu$ even becomes negative. This direction is distinguished,[12] as can be seen by the dashed-dotted curve calculated for a polycrystalline average.

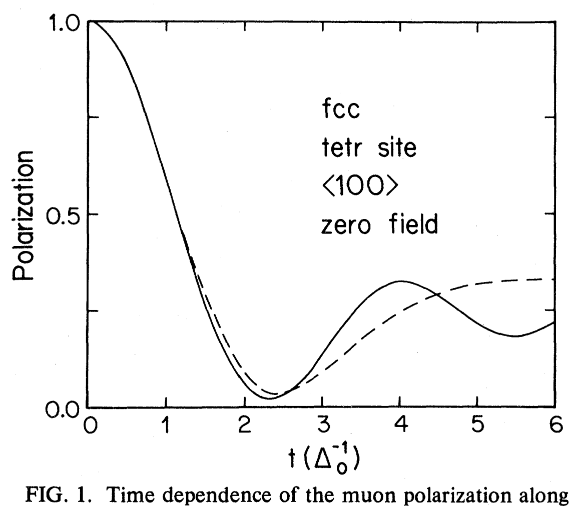


FIG. 1. Time dependence of the muon polarization along the $\langle 100\rangle$ direction for the tetrahedral site in a fcc lattice. The dashed line shows the Kubo-Toyabe function for the same second moment. The time is measured in units of $\Delta_0^{-1}$ [see Eq. (8)].

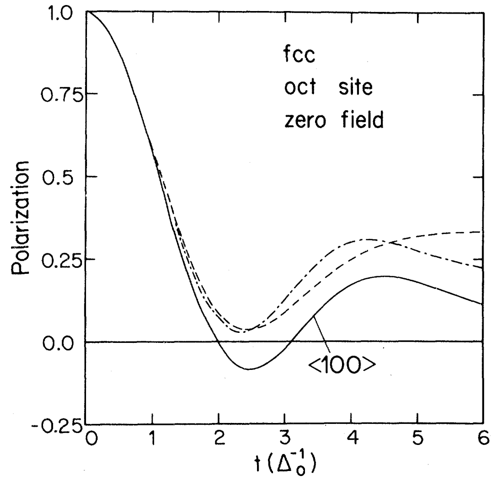


FIG. 2. Muon polarization along the $\langle 100\rangle$ direction (full line) for the octahedral site. The dashed-dotted line corresponds to the polycrystalline average and the dashed line is the Kubo-Toyabe function.

It is clear from these figures that the long-time behavior of the muon polarization in zero field can be different from the simple Kubo-Toyabe theory. This difference is only slightly changed if further shells are taken into account. Their influence can be estimated from Eq. (11) with the appropriate value of $\Delta$ and is found to be only a few percent for the investigated lattice configurations, provided $t\Delta_0 \lesssim 5$. For larger times they generally reduce the total value of $p_\mu$.

We also calculated the full spin dynamics in longitudinal and in transverse external fields. In the latter case, the numerical results can be compared to the predictions obtained from the secular approximation.

The secular approximation takes into account only those parts of $H_j$ which commute with the Zeeman interaction:

$$H_j^{\rm sec} = 2\hbar\lambda_j I^z J_j^z \ , \tag{12}$$

with

$$\lambda_j = \hbar\gamma_\mu\gamma_J[1-3(n_j^z)^2]/(2r_j^3) \ , \tag{13}$$

and where the $z$ axis has been chosen in the direction of the external field. The muon polarization perpendicular to the field is written as

$$P_\mu^x(t) = \cos(\gamma_\mu B t)\, G_x(t) \ . \tag{14}$$

The calculation of the relaxation function in the secular approximation is straightforward and leads to

$$G_x^{\rm sec}(t) = \prod_{j=1}^{N} \left[ \frac{\sin[(2J+1)\lambda_j t]}{(2J+1)\sin(\lambda_j t)} \right] \ . \tag{15}$$

A measure of the strength of the relaxation is the second moment $M_2$ which, in the present notation, can be defined as

$$M_2 = -\left.\frac{d^2 G_x^{\mathrm{sec}}}{dt^2}\right|_{t=0} = \sum_{j=1}^{N} \frac{4J(J+1)}{3}\lambda_j^2 \ . \qquad (16)$$

Its polycrystalline average is five times smaller than the zero-field quantity $M_0$.

For the external field along the $\langle 111 \rangle$ direction the relaxation function $G_x^{\mathrm{sec}}(t)$ is shown in Fig. 3 for the tetrahedral site with $N=4$ (solid line). The exact numerical calculations perfectly agree with the secular approximation (solid line) for field values which exceed 300 times the strength of the dipolar field $B_D$. A deviation can be seen (dashed-dotted line) for $B_{\mathrm{ext}} \approx 60 B_D$. The dashed line represents the Gaussian-decay approximation of the transverse relaxation function given by the second moment $M_2$ [Eq. (16)].

Within the secular approximation the influence of the further neighbor shells can be easily calculated from Eq. (15) and its contribution to $G_x^{\mathrm{sec}}(t)$ is found to be small[13] for the lattice configurations under consideration.

An inspection of Eq. (15) shows that the relaxation function is oscillating. Its first zero at $t_0$ is determined by the largest value of the $\lambda_j$'s, which is obtained for $n_j^z = \pm 1$. $G_x^{\mathrm{sec}}(t)$ is then non-negative if the $\mu^+$ is sitting at a site of inversion symmetry, otherwise it will be negative for times slightly larger than $t_0$.[14]

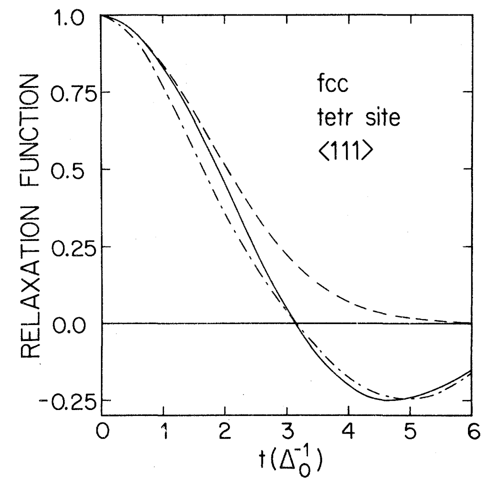


FIG. 3. Relaxation function in transverse fields along the $\langle 111 \rangle$ direction in a fcc lattice. The dashed line shows the Gaussian decay described by $M_2$ [Eq. (16)]. The full line is the secular approximation [Eq. (15)] which agrees with the numerical solution [Eq. (4)] for $B_{\mathrm{ext}} \gtrsim 300 B_D$ but differs from the exact behavior for $B_{\mathrm{ext}} = 60 B_D$ (dashed-dotted curve).

In conclusion, the exact calculation of the spin dynamics of a polarized muon at an interstitial site with four or six nearest-neighbor nuclei with spin $\frac{1}{2}$ shows marked differences from the Kubo-Toyabe theory in the long-time behavior, especially for some specific symmetry directions and in the long-time regime. In analyzing experiments which aim at demonstrating the static behavior of muons by zero-field measurements, one should therefore take into account the sometimes strong dependence of the polarization decay on the crystal orientation.


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[8] The dipolar interaction among the nuclear spins $J_j$ is neglected. Its inclusion in the numerical treatment is straightforward. Furthermore, we do not account for a quadrupolar interaction due to a field gradient induced by the muon at the neighboring nuclei.

[9] We are presently trying to extend the calculations to higher spin nuclei and to include quadrupole effects. The large dimensions of the matrices, however, require special programming and computational efforts.

[10] The method of calculation is the same as has been used to evaluate $p_\mu(t)$ for anomalous muonium. The corresponding equations may be found in the Appendix of Ref. 11.

[11] P. F. Meier, in *Exotic Atoms 79,* edited by K. Crowe *et al.* (Plenum, New York, 1980).

[12] $\vec{p}_\mu(t)$ also becomes negative for the $\langle 111 \rangle$ direction at the tetrahedral site. This direction and the $\langle 100 \rangle$ at the octahedral site are distinguished by the fact that $\vec{p}_\mu$ is pointing towards one of the nearest neighbors.

[13] An exceptional case occurs for the field along the $\langle 100 \rangle$ direction at the tetrahedral site where $M_2$ from the first-neighbor shell is vanishing.

[14] For a $\mu^+$ at the octahedral site in Cu one obtains $t_0 = 7.3\ \mu$s for $\langle 100 \rangle$, whereas for the tetrahedral site and $\langle 111 \rangle$ $G_x$ becomes negative at $t_0 = 4.7\ \mu$s.