Magnetic dipole relaxation of the spin of the muon

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Muon polarization $P(t)$ due to the magnetic dipole interaction with integral and half-integral spins I of ambient nuclei in zero external magnetic field is calculated as a function of time. It is assumed that the electric quadrupole interaction between the muon and the nuclei (with $I > \frac{1}{2}$) has its maximum possible strength.

We calculate the spin relaxation for a positive muon interacting with the nuclei of ambient atoms via the dipole interaction between their magnetic moments. The characteristic properties of muon spin relaxation are particularly well defined in zero external magnetic field ($B = 0$). The first calculations[1] were performed on the basis of the Kubo-Toyabe theory[2] of spin relaxation with an approximately isotropic distribution of magnetic fields acting upon the particle. The relaxation function $P(t)$ obtained in Ref. 1, i.e., the muon polarization as function of time, has been very useful in studies of different processes leading to the relaxation of muon spin in matter, e.g., slow muon diffusion. Recent calculations[3–6] of $P(t)$ have taken into account both nuclear spin dynamics and the discrete disposition of the spins at the crystal lattice sites. These factors were found to have a significant effect on the form of $P(t)$. The function $P(t)$ determined in this way is in agreement with the measured relaxation of the spin of the muon localized in a diamagnetic metal[7] in which other muon spin relaxation processes are negligible.

At the same time, we recall the complexity of the method usually employed to calculate $P(t)$, based on the determination of the eigenfrequencies of the many-body Hamiltonian for interaction between the spin of the muon and the spins of the ambient nuclei. Mathematical difficulties restrict this method to the case of a small number of interacting nuclei and low nuclear spins. On the other hand, our method of calculating $P(t)$ enables us to extend the number of nuclei participating in the interaction and to consider the case of higher nuclear spins.

The relaxation function $P(t)$ is determined by the spin Hamiltonian for the interaction between the muon and the ambient nuclei, i.e.,

$$\hat{H}=\sum_{k=1}^{N}(\hat{H}_k^D+\hat{H}_k^Q), \tag{1}$$

which consists of the sum of the dipole $\hat{H}_k^D$ and quadrupole $\hat{H}_k^Q$ interactions:

$$\hat{H}_k^D=\hbar\omega_k^D[(\hat{\mathbf{S}}\hat{\mathbf{I}}_k)-3(\mathbf{n}_k\hat{\mathbf{S}})(\mathbf{n}_k\hat{\mathbf{I}}_k)], \tag{2}$$

$$\hat{H}_k^Q=\hbar\omega_k^Q[(\mathbf{n}_k\hat{\mathbf{I}}_k)-{}^1/_3I(I+1)],\quad \omega_k^D=\hbar\gamma_\mu\gamma_I/r_k^3, \tag{3}$$

where N is the number of nuclei interacting with the muon, $\mathbf{S}_k$ is the muon spin operator, $\hat{\mathbf{I}}_k$ is the nuclear spin operator, n_k is the unit vector in the direction between the muon and k th nucleus, ω_k^D and ω_k^Q are the frequencies of the dipole and quadrupole interactions, γ_μ and γ_I are the gyromagnetic ratios for the muon and the nucleus, and r_k is the distance between the muon and k th nucleus. As usual, we have neglected in the Hamiltonian (1) the relatively weak spin–spin interaction between the nuclei. All the nuclei are assumed to be identical, and the external magnetic field is $B = 0$.

We shall now consider a situation that is frequently encountered in muon spin relaxation in metals, namely,

$$\omega_k^Q\gg\omega_k^D. \tag{4}$$

Since muon spin relaxation due to dipole interactions occurs in a time $t\sim(\omega_k^D)^{-1}$, where ω_k^D corresponds to minimum distances r_k, the inequality given by (4) allows us to average over time intervals

$$(\omega_k^Q)^{-1}\ll\delta t\ll(\omega_k^D)^{-1} \tag{5}$$

when we evaluate $P(t)$. The interval defined by (5) corresponds to averaging the dipole Hamiltonian $\hat{H}_k^D$ over the quadrupole interactions:

$$\langle\hat{H}_k^D\rangle=\langle\exp(i\hat{H}_k^Qt/\hbar)\hat{H}_k^D\exp(-i\hat{H}_k^Qt/\hbar)\rangle. \tag{6}$$

When the average defined by (6) is evaluated, the interaction between the muon and each individual nucleus is considered in its own "radial" coordinate frame (x_k, y_k, z_k) in which the quantization axis z_k is parallel to the vector $\mathbf{n}_k$. In the radial coordinate frame, the quadrupole operator (3) has the form

$$\hat{H}_k^Q=\hbar\omega_k^Q[\hat{I}_{zk}^2-{}^1/_3I(I+1)], \tag{7}$$

and the operator $\exp(i\hat{H}_k^Qt/\hbar)$ in (6) can be transformed so that

$$\exp(i\hat{H}_k^Qt/\hbar)=\exp(-iA_k)\sum_m\hat{E}_m\exp(i\omega_k^Qm^2t), \tag{8}$$

where $m=\frac{1}{2},\frac{3}{2},\dots,I$ for half-integral I and $m = 1,2,\dots,I$ for integral I. In these expressions, $A_k=\frac{1}{3}\omega_k^QI(I+1)$, and $\hat{E}_m$ is a diagonal operator with two elements equal to unity, i.e.,

$$(\hat{E}_m)_{m,\,m}=(\hat{E}_m)_{-m,\,-m}=1. \tag{9}$$

Substituting (8) in (6), we obtain

$$\langle\hat{H}_k^D\rangle=\sum_{m,m'}\langle\hat{E}_m\exp(i\omega_k^Qm^2t)\hat{H}_k^D\hat{E}_{m'}\exp(-i\omega_k^Qm'^2t)\rangle.$$

Using (5) and recalling that $\omega_k^Q\gg\omega_k^D$, we have

$$\langle\hat{H}_k^D\rangle=\sum_m\hat{E}_m\hat{H}_k^D\hat{E}_m. \tag{10}$$

It follows from (8) and (10) that, for $I=\frac{1}{2}$ we have $\langle\hat{H}_k^D\rangle=\hat{H}_k^D$, which should be the case in the absence of the

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quadrupole interaction between nuclei with spin $I=\frac{1}{2}$.

Replacing $\hat{H}_k^D$ with the expression given by (2), and recalled that

$$\sum_m \hat{E}_m \hat{I}_{z_k} \hat{E}_m = \hat{I}_{z_k},$$

we find that

$$\langle \hat{H}_k{}^D \rangle = \hbar\omega_k{}^D \left[\hat{\mathrm{S}} \left(\sum_m \hat{E}_m \hat{\mathbf{I}}_k \hat{E}_m \right) - 3\hat{S}_{z_k} \left(\sum_m \hat{E}_m \hat{I}_{z_k} \hat{E}_m \right) \right]$$
$$= \hbar\omega_k{}^D \left[\hat{S}_{x_k} \left(\sum_m \hat{E}_m \hat{I}_{x_k} \hat{E}_m \right) + \hat{S}_{y_k} \left(\sum_m \hat{E}_m \hat{I}_{y_k} \hat{E}_m \right) \right.$$
$$\left. - 2\hat{S}_{z_k} \hat{I}_{z_k} \right] = \hbar\omega_k^D [\hat{S}_{x_k} \hat{F}_{x_k} + \hat{S}_{y_k} \hat{F}_{y_k} - 2\hat{S}_{z_k} \hat{I}_{z_k}]. \tag{11}$$

The cases of integral and half-integral spin I, for which the operators $\hat{F}$ are determined in different ways, will now be considered separately.

For integral I, we have $\hat{F}_{x_k} = \hat{F}_{y_k} = 0$ and it follows from (11) that the average dipole Hamiltonian is

$$\langle \hat{H}_k{}^D \rangle = -2\hat{S}_{z_k} \hat{I}_{z_k}. \tag{12}$$

It is clear from this expression that the dynamics of the muon spin S is determined exculsively by the radial projection I_{z_k} of the nuclear spin, which is conserved because of the strong quadrupole interaction. The function $P(t)$ can therefore be determined in this case by classical methods by calculating the magnetic field that acts on the muon and is due to the radial projections of the magnetic moments of the ambient nuclei. Of course, for integral I, the functions $P(t)$ can also be calculated for half integral I by the quantum mechanical method described below [see (14)].

For half integral spin I, the operators $\hat{F}_{x_k}$ and $\hat{F}_{y_k}$ in (11) are determined by the two nonzero elements $(\hat{F})_{1/2,-1/2}$ and $(\hat{F})_{-1/2,1/2}$ and can be symbolically written in the form

$$\hat{F}_{x_k} = {}^1\!/_2 ({}^1\!/_2 + I) \hat{\sigma}_{x_k}, \qquad \hat{F}_{y_k} = {}^1\!/_2 ({}^1\!/_2 + I) \hat{\sigma}_{y_k}. \tag{13}$$

These expressions are meant to indicate that the Pauli matrices $\hat{\sigma}_{x_k}$ and $\hat{\sigma}_{y_k} = 0$ are augmented with zeros in all the peripheral rows and columns of the operators $\hat{F}_{x_k}$ and $\hat{F}_{y_k}$. The relaxation function for this case is given by

$$P(t) = \frac{2}{(2I+1)^N} \operatorname{Sp} (\hat{S}_z \exp(i\hat{\mathscr{H}}t/\hbar) \hat{S}_z \exp(-i\hat{\mathscr{H}}t/\hbar) \tag{14}$$

where $\hat{S}_z$ is the projection of the muon spin on to the intial polarization of the muon spin in the laboratory frame (x,y,z) for $t=0$ and $\hat{\mathscr{H}}$ is the spin Hamiltonian of the muon and the ambient nuclei, averaged over the electric quadrupole interaction between the muon and the nuclei. The Hamiltonian $\hat{\mathscr{H}}$ is the sum of N averaged Hamiltonians (11) that describes the interaction between the muon and the spins of the individual nuclei. The operators $\hat{S}_{x_k}, \hat{S}_{y_k}, \hat{S}_{z_k}$ that appear in (11) in the radial coordinate frames (x_k, y_k, z_k) must be transformed so that they act on the components of the muon wave function ψ specified in the laboratory frame (x,y,z):

$$\hat{\mathscr{H}} = \sum_{k=1}^N \hbar\omega_k{}^D [(\hat{M}_k{}' \hat{S}_{x_k} \hat{M}_k) \hat{F}_{x_k} + (\hat{M}_k{}' \hat{S}_{y_k} \hat{M}_k) \hat{F}_{y_k}$$
$$- 2(\hat{M}_k{}' \hat{S}_{z_k} \hat{M}_k) \hat{I}_{z_k}]. \tag{15}$$

The two-dimensional operator $\hat{M}_k$ in this expression transforms the muon wave function ψ from the laboratory frame of coordinates to the kth radial frame, and the operator $\hat{M}'_k$ executes the reverse transformation

$$\psi' = \hat{M}_k \psi, \quad \psi = \hat{M}_k{}' \psi'.$$

The operators $\hat{M}_k$ and $\hat{M}'_k$ can be expressed in terms of the Euler angles defining rotations of the coordinate axes of the laboratory frame (x,y,z) under transformation to the kth radial frame and back again. The result is

$$\hat{\mathscr{H}} = \sum_{k=1}^N \hbar\omega_k{}^D [(\hat{S}_x \cos\alpha_k + \hat{S}_y \sin\alpha_k) \hat{F}_{x_k} + (-\hat{S}_x \sin\alpha_k \cos\beta_k$$
$$+ \hat{S}_y \cos\alpha_k \cos\beta_k + \hat{S}_z \sin\beta_k) \hat{F}_{y_k} - 2(\hat{S}_x \sin\beta_k \sin\alpha_k$$
$$- \hat{S}_y \sin\beta_k \cos\alpha_k + \hat{S}_z \cos\beta_k) \hat{I}_{z_k}]. \tag{16}$$

where the operators $\hat{S}_x, \hat{S}_y, \hat{S}_z$ act on the muon spin wave function in the laboratory frame (x,y,z), the operators $\hat{F}_{x_k}, \hat{F}_{y_k}, \hat{F}_{z_k}$ act on the nuclear spin wave functions in the radial coordinate frames (x_k, y_k, z_k) α_k and β_k are the angles through which the axes of the radial frame of coordinates have to be rotated to make them coincident with the laboratory axes; α_k is the angle defining the rotation around the x axis, i.e., around the direction of the primary muon polarization, and β_k defines the rotation around the x_k axis whose direction is taken to be perpendicular to the plane containing the vectors $\mathbf{z}$ and $\mathbf{n}_k$. It is readily seen that this choice of the direction of the x_k axis enables us to superimpose the radial and laboratory coordinate frames by the above two rotations alone. It may be shown that the Hamiltonian given by (16) describes the muon spin dynamics not only for the electric quadrupole interaction of maximum strength, as defined by (4), but also for $I=\frac{1}{2}$, for which there is no quadrupole interaction.

The relaxation function evaluated with the aid of (14) can be written in the form of the series

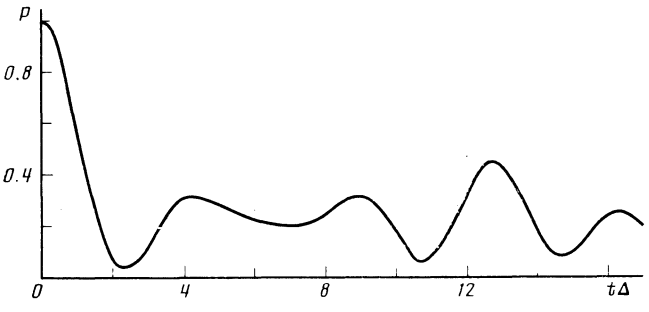

FIG. 1. Muon poarization $P(t)$ in the absence of an external magnetic field. The time t is shown in units of Δ^{-1}, defined by (19). The polarization $P(t)$ was calculated from (17) for a muon localized in the octahedral interstitial space of an **fcc** crystal interacting with six nearest neighbors with spin $I=\frac{1}{2}$.

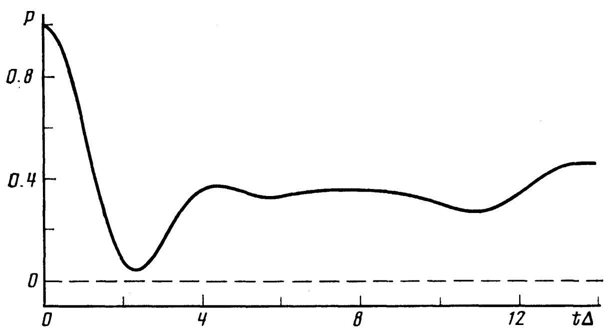

FIG. 2. The function $P(t)$ for a muon in the octahedral interstitial space of an **fcc** crystal interacting with the six nearest neighbors with spins $I = 3/2$.

$$P(t)=\sum_{n=0}^{\infty} a_n t^n i^n, \tag{17}$$

where

$$a_n=\frac{1}{n!}\operatorname{Sp} \hat{s}_z \hat{R}_n, \qquad \hat{R}_1=[\hat{s}_z,\hat{\mathscr{H}}], \qquad a_0=1.$$

The commutators $\hat{R}_n$ can be evaluated from the recursion formula $n>1$ for any $\hat{R}_n = [\hat{R}_{n-1}, \hat{\mathscr{H}}]$.

Figures 1–5 show the relaxation function $P(t)$ at the octahedral and tetrahedral interstices of an **fcc** crystal, calculated from (17) for different values of the nuclear spin I, taking into account one or two coordination spheres containing the nuclei nearest to the muon. In these figures, the horizontal axis gives the time t in units of Δ^{-1} where Δ is the second moment in the expression

$$P(t)=P(0)\exp(-\Delta^2t^2/2),$$

which describes the function $P(t)$ for $t\to 0$.For the case $\omega_k^Q \gg \omega_k^D$ in which we are interested here, we have[8]

$$\Delta^2=\frac{4}{3}I(I+1)\sum_{k=1}^{N}\left(\frac{\hbar\gamma_\mu\gamma_I}{r_k^3}\right)^2\sin^2\theta_k \tag{18}$$

for integral spin I and

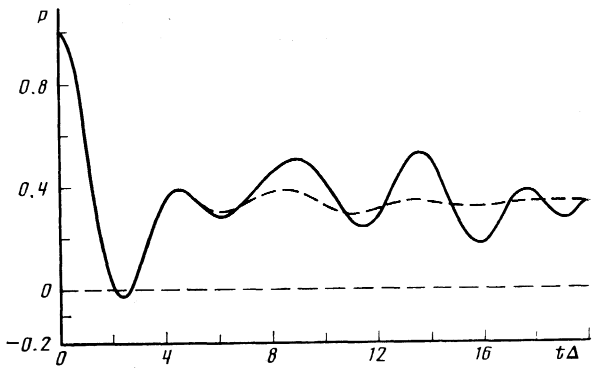

FIG. 4. The function $P(t)$ for a muon in the octahedral interstital space of an **fcc** crystal. The solid curve corresponds to the interaction between the muon and six nuclei in the nearest coordination sphere; dashed curve—interaction between the muon and 6 + 8 nuclei in the two nearest coordination spheres. The nuclear spin is $I = 1$. The quantity Δ is given by (18) in this case.

$$\Delta^2=\sum_{k=1}^{N}\left(\frac{\hbar\gamma_\mu\gamma_I}{r_k^3}\right)^2\left[\frac{1}{8}(2I+1)(2-\sin^2\theta_k)\right.$$
$$\left.+\frac{4}{3}I(I+1)\sin^2\theta_k\right], \tag{19}$$

for half-integral spin I, where θ_k is the angle between the direction of the primary muon polarization $\mathbf{P}(0)$ and the direction of the vector $\mathbf{n}_k$. The sum is evaluated over all the nuclei interacting with the muon. We note that the expressions given by (19) for Δ^2 and by (17) for the function $P(t)$ are also valid for $I=\frac{1}{2}$, for which there is no quadrupole interaction between the muon and the nuclei, i.e., $\omega_k^Q=0$. The accuracy with which the function $P(t)$ is calculated is determined by the number of terms taken in the series (17), and decreases with increasing time t. To calculate $P(t)$ with $\delta P/P \lesssim 0{,}1\%$ for $t=15\Delta^{-1}$ (see Figs. 1–5) we need fifty to a hundred terms in (17), depending on the number of nuclei interacting with the muon.

It is clear from Figs. 1–5 that the function $P(t)$ has a deep minimum for $t\Delta \sim 2$, as was shown in Ref. 1. The function $P(t)$ oscillates[3–8] when $t\Delta > 4$, and these oscillations become better defined for small values of I and for a relatively

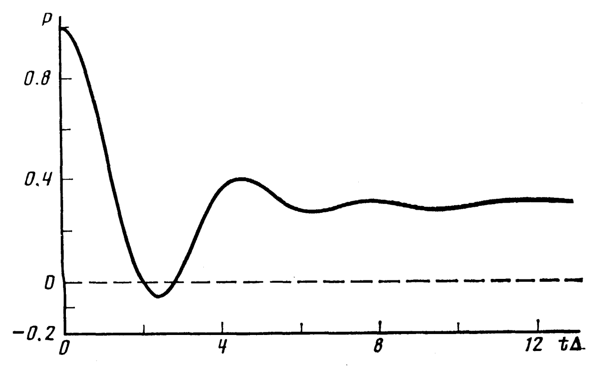

FIG. 3. The function $P(t)$ for a muon in the tetrahedral interstitial space of an **fcc** crystal interacting with four nearest neighbor nuclei with spins $I = 9/2$.

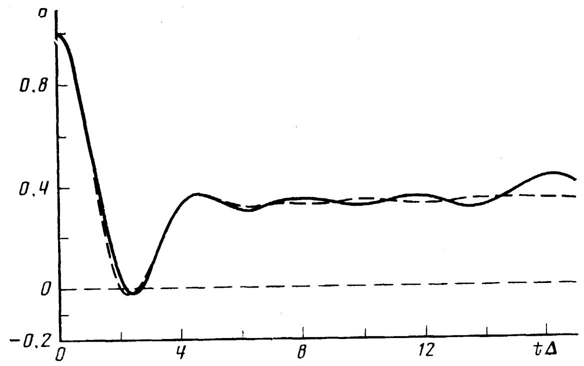

FIG. 5. The function $P(t)$ for a muon in the octahedral interstitial space of an **fcc** crystal. The muon spin $I = 2$. The remaining notation is the same as Fig. 4.

small number N of nuclei interacting with the muon. The oscillations on the function $P(t)$ are a consequence of the finite number of frequencies used to describe the dipole interaction between the muon spin and the ambient nuclei. The low value of I, and the small number of nuclei interacting with the muon, lead to a smaller number of frequencies and deeeper oscillations on $P(t)$.

Figures 4 and 5 illustrate how the second coordination sphere, containing nuclei nearest to the muon, affects the oscillations of $P(t)$. It is clear from these figures that the amplitude of the oscillations is reduced when the interaction between the muon and the nuclei in the second coordination sphere is taken into account. This reduction is particularly noticeable in Fig. 4 in which there are large oscillations due to the interaction between the muon and the nuclei in the first coordination sphere.

The graphs of $P(t)$ shown in Figs. 1 and 4 are in agreement with calculations performed by a different method in Ref. 8 for nuclear spins $I = \frac{1}{2}$ and $I = 1$, and for the relatively short times $t < 8\Delta^{-1}$.

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[1]R. S. Hayano, Y. J. Uemura and J. Imazato *et al.*, Phys. Rev. **B20**, 850 (1979).
[2]R. Kubo, and T. Toyabe, *Magnetic Resonance and Relaxation*, North-Holland, Amsterdam, 1967, p. 810.
[3]M. Celio and P. F. Meier, Phys. Rev. **B27**, 1908 (1983).
[4]K. G. Petzinger and S. H. Wei, Hyperfine Inter. **17–19**, 441 (1984).
[5]E. Holzschuh and P. F. Meier, Phys. Rev. **B29**, 1129 (1984).
[6]M. Celio, Hyperfine Inter. **31**, 41 (1986).
[7]R. Kadono, J. Imazato, K. Nishiyama *et al.*, Phys. Lett. **107A**, 279 (1985).
[8]M. Celio and P. F. Meier, Hyperfine Inter. **17–19**, 435 (1984).

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