New method of studying the properties of normal metals by a muon technique

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The contribution made to the muon spin precession frequency in normal metals by the muon interaction with the magnetic moments of the nuclei is calculated. The relative frequency shift depends on the crystal orientation and can be of the order 10^{-2} to 10^{-5}. The effect considered uncovers a possibility of directly determining practically all the normal-metal parameters that are obtained by the "traditional" muon technique from the relaxation rate. It permits identification of the type of pore in which the muon was stopped and measurement of the distortion of the crystal cell by the muon, of the rate of muon diffusion, of quadrupole interactions, and others. The effect must be taken into account also in measurements of the Knight shift for muons.

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The accuracy of precision measurements of the muon spin precession frequency in metals has reached 10^{-3}–10^{-4}%, and the possibility of improving it is far from exhausted. Even now, the Knight shift for the muon, 10^{-4}–10^{-5} (Ref. 1), is reliably measured. The observable muon precession frequency in an external field **B**, with allowance for the Knight shift, is usually defined as

$$\omega_\mu=\gamma_\mu(1+K)\,|\langle\mathbf{B}\rangle|. \tag{1}$$

Here $\gamma_\mu = 8.5\times10^4\ \mathrm{sec}^{-1}\cdot\mathrm{G}^{-1}$ is the gyromagnetic ratio of the muon, K is the Knight-shift constant, and $\langle\mathbf{B}\rangle$ is the macroscopic field in the metal. If there is no demagnetizing factor, the average macroscopic field in the sample is $\langle\mathbf{B}\rangle=(1+4\pi\chi)\mathbf{B}$. The modulus of the magnetic susceptibility χ for normal metals is of the order of $(0.1\text{–}5)\times10^{-6}$; $\langle\mathbf{B}\rangle$ can accordingly differ from **B** in the fifth significant figure.

Following the standard terminology, we shall refer hereafter to the precession frequency shift $\Delta\omega$ as the difference between the observed frequency ω_μ and the precession frequency $\omega_0=\gamma_\mu|\langle\mathbf{B}\rangle|$ in the macroscopic field. It will be shown that the Knight-shift constant K can be calculated with the aid of (1) only when the external field is strong ($B\approx3$–5 kG). In medium and weak fields the frequency shift is due mainly to interaction of the magnetic moment of the muon with the stochastic magnetic fields of the sample nuclei.

Thus,

$$\Delta\omega=\delta\omega_{\mathrm{dip}}+\delta\omega_K,$$

where $\delta\omega_{\mathrm{dip}}$ and $\delta\omega_K$ are corrections connected respectively with the dipole fields of the nuclei and with the Knight shift. The appearance of $\delta\omega_{\mathrm{dip}}$ is in full analogy with the change, well known in NMR, of the level-splitting energy in an external field, a change due to the nonsecular part of the interaction operator of the magnetic moments of the sample nuclei.[2]

We consider a nondiffusing muon stopped in some crystal pore. The Hamiltonian of the dipole interaction between the magnetic moment of the muon and the magnetic moments of the nuclei is of the form

$$H_{\mathrm{dip}}=\hbar^2\gamma_\mu\gamma_n\mathbf{S}_\mu\sum_j[3\mathbf{n}_j(\mathbf{n}_j\mathbf{I}_j)-\mathbf{I}_j]r_j^{-3}. \tag{2}$$

As a result, each muon turns out to be in a local magnetic field

$$\mathbf{b}=\sum_j[3\mathbf{n}_j(\mathbf{n}_j\boldsymbol{\mu}_j)-\boldsymbol{\mu}_j]r_j^{-3}. \tag{3}$$

Here $\mathbf{S}_\mu$ and $\mathbf{I}$ are the spin operators of the muon and of the nucleus, γ_n and $\mu=\hbar\gamma_n\mathbf{I}$ are the gyromagnetic ratio and the magnetic moment of the sample nuclei, $\mathbf{r}_j$ is the vector joining the muon with the j-th nucleus, and $\mathbf{n}_j=\mathbf{r}_j/r_j$. The summation is over all the lattice nuclei.

The dipole field **b**, obviously, varies randomly from point to point. As a result, muons stopped at different points precess at different frequencies; this leads to relaxation (damping of the amplitude) of the observable polarization of the muon ensemble.

If the external field is strong ($B\gg b$), the scatter of the precession frequency is determined mainly by the projection b_z of the local field on the direction of the external field. Further, the magnetic moments of the nuclei obviously also precess in the external field, and if the condition $\gamma_nB\gg\gamma_\mu b$ is satisfied (the nuclei precess rapidly) the muon magnetic moment does not notice the rapidly oscillating components of the local field **b**. The depolarization is then determined by the static part $b_{z\,\mathrm{st}}$ of the component b_z. We note also that rapid oscillations of the local field can be caused by interaction of the nuclear quadrupole moments with the muon electric field gradient.

Since the nuclear magnetization in normal metals is almost always vanishingly small, the average observable muon precession frequency is determined by Eq. (1) if the local-field components transverse to the external field are neglected. It can be easily seen, however, that these components must be taken into account in precision measurements. Indeed the precession frequency of an individual muon is equal to

$$\omega=\gamma_\mu|(1+K)\langle\mathbf{B}\rangle+\mathbf{b}_{\mathrm{st}}|. \tag{4}$$

The observable frequency ω_μ is the result of averaging over the muon ensemble

$$\omega_\mu=\langle\omega\rangle\approx(1+K)\,\omega_0+{}^1/{}_2\gamma_\mu^2\omega_0^{-1}(\langle b_{x\,\mathrm{st}}^2\rangle+\langle b_{y\,\mathrm{st}}^2\rangle).$$

It can be seen that allowance for the transverse components

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shifts the precession frequency by the amount

$$\delta\omega_{\rm dip}/\omega_0={}^1/{}_2\gamma_\mu{}^2\omega_0{}^{-2}(\langle b_{x\,{\rm st}}^2\rangle+\langle b_{y\,{\rm st}}^2\rangle). \quad (5)$$

We shall illustrate the mechanism that produces the frequency shift using an example in which the dipole field has a normal distribution. For the polarization components perpendicular to the external field we have

$$P_+(t)=\int e^{i\omega t}W(\mathbf{b}_{\rm st})\,d\mathbf{b}_{\rm st}P_+(0),$$

where $P_+ = P_x + iP_y$, and ω is defined by Eq. (4). Expanding ω in powers of the ratio $b_{\rm st}/B$ up to the quadratic terms inclusive and integrating, we find

$$P_+(t)\approx\exp[-{}^1/{}_2\gamma_\mu{}^2\langle b_{z\,{\rm st}}^2\rangle t^2+i\omega_0 t](1-2i\delta\omega_{\rm dip}t)^{-1/2}P_+(0). \quad (6)$$

Since we have neglected terms of order $(b_{\rm st}/B)^3$ in the argument of the exponential, Eq. (6) is valid under the condition $\omega_0 t(b_{\rm st}/B)^3\ll 1$. If the more stringent inequality $\delta\omega_{\rm dip}t\ll 1$ is satisfied, the radical in (6) can be approximately replaced by $\exp(i\delta\omega_{\rm dip}t)$. The concept of the dipole shift of the precession frequency is thus really applicable for times $t\ll\delta\omega_{\rm dip}^{-1}$. In normal metals the modulus of the dipole field does not exceed 5–7 G. We then obtain from (5) the estimate

$$t\ll 2\cdot 10^{-6}B/b_{\rm st}\approx\tau_\mu B/b_{\rm st},$$

where $\tau_\mu\approx 2.2\,\mu$sec is the muon lifetime. In experiment, the observation time is limited to several times τ_μ (realistically, 5–10 μsec). Therefore Eq. (5) can at any rate be used at $B\gtrsim 50$ G. In such fields the precession frequency shift can reach 1%, which is substantially higher than the sensitivity threshold of the experiment.

As seen from (6), the depolarization rate of nondiffusing muons is determined by the parameter

$$\sigma^2={}^1/{}_2\gamma_\mu{}^2\langle b_{z\,{\rm st}}^2\rangle.$$

Comparison with Eq. (5) shows that the frequency shift contains practically the same information on the properties of matter as σ^2. At the same time, the frequency is determined with an accuracy higher by several orders than that of the measured relaxation rate. The dipole frequency shift is therefore of considerable interest by itself.

We proceed now to calculate $\delta\omega_{\rm dip}$. We consider first nuclei with spin $I=\frac{1}{2}$, when there are no quadrupole interactions between the nuclei and the electric field gradients. From (3) we have for the static part of the dipole field

$$b_{x\,{\rm st}}=\sum_j 3n_{jx}n_{jz}\mu_{jz}r_j{}^{-3},$$

$$b_{y\,{\rm st}}=\sum_j 3n_{jy}n_{jz}\mu_{jz}r_j{}^{-3},$$

$$b_{z\,{\rm st}}=\sum_j (3n_{jz}{}^2-1)\mu_{jz}r_j{}^{-3}.$$

We substitute these expressions in (5) and average, with account taken of the relations

$$\mu_{jz}=\hbar\gamma_n I_{jz},\qquad \langle I_{jz}I_{j'z}\rangle={}^1/{}_3I(I+1)\delta_{jj'}$$

(we assume for simplicity that the spins of all the nuclei are identical). As a result we arrive at the equation

$$\frac{\delta\omega_{\rm dip}}{\omega_0}=\frac{3}{2}(\hbar\gamma_\mu\gamma_n)^2I(I+1)\omega_0{}^{-2}\sum_j n_{jz}{}^2(1-n_{jz}{}^2)r_j{}^{-6}. \quad (7)$$

For metals with crystal lattice of cubic symmetry, it is easy to determine the explicit form of the angular dependence of the frequency shift:

$$\delta\omega_{\rm dip}/\omega_0={}^1/{}_4(\hbar\gamma_\mu\gamma_n)^2I(I+1)\omega_0{}^{-2}[a_1+a_2(\lambda_1{}^4+\lambda_2{}^4+\lambda_3{}^4)], \quad (8)$$

where λ_1, λ_2, and λ_3 are the direction cosines of the external-field vector in a coordinate frame whose axes coincide with the crystal axes;

$$a_1=\sum_j(9\cos^4\alpha_j-1)r_j{}^{-6},\quad a_2=3\sum_j(1-5\cos^4\alpha_j)r_j{}^{-6}, \quad (9)$$

where α_j is the angle between the vector $\mathbf{n}_j$ and the (1, 0, 0) direction. For polycrystalline samples

$$\frac{\delta\omega_{\rm dip}}{\omega_0}=\frac{1}{5}(\hbar\gamma_\mu\gamma_n)^2I(I+1)\omega_0{}^{-2}\sum_j r_j{}^{-6}. \quad (10)$$

Naturally, the formulas obtained are valid also for nuclei with $I>\frac{1}{2}$ in strong external fields, when the energy of the interaction of the nuclei with the external field is substantially higher than the energy of the quadrupole interaction of the nuclei with the muon.

There are rather few metals with $I=\frac{1}{2}$ and with more or less noticeable values of μ (see the table). Thus, greatest interest attaches to elements with $I>\frac{1}{2}$. Since the dipole frequency shift is inversely proportional to the modulus of the external field, to observe the effect it is more convenient to use weak fields of the order of 50–500 G. If the external magnetic field is strong, the principal role is played by quadrupole interactions of the nuclei with the electric-field-gradient tensor of the muon. In this case, as seen from (5) and (6), $\delta\omega_{\rm dip}=2\sigma^2/\omega_0$.

We confine ourselves hereafter to metals with cubic crystal-lattice symmetry. It is known that in this case the Hamiltonian of the quadrupole interaction can be well approximated by

$$H_{Q_j}=A_j(r_j)(\mathbf{I}_j\mathbf{n}_j)^2+B_j(r_j). \quad (11)$$

We introduce for each nucleus its own coordinate frame $x'_jy'_jz'_j$, with axes z'_j directed along the vectors $\mathbf{n}_j$, and the axes x'_j and y'_j along some unit vectors $\mathbf{e}_{1j}$ and $\mathbf{e}_{2j}$. In this coordinate frame the dipole field (2) can be represented in the form

$$\mathbf{b}=\hbar\gamma_n\sum_j(2\mathbf{n}_jI_{jz'_j}-\mathbf{e}_{1j}I_{jx'_j}-\mathbf{e}_{2j}I_{jy'_j})r_j{}^{-3}. \quad (12)$$

The eigenfunctions of the quadrupole operator (11) are states with definite spin projections on the z'_j axis:

$$H_{Q_j}|m_j\rangle=[A_j(r_j)m_j^2+B_j(r_j)]|m_j\rangle.$$

Since states with $m_j=|m_j|$ and $m_j=-|m_j|$ are mutually degenerate, not only the diagonal matrix elements of an arbitrary operator $\langle m_j|\hat{A}|m_j\rangle$ but also the elements $\langle m_j|\hat{A}|-m_j\rangle$ are independent of time. The operators $I_{jx'_j}$

TABLE I. Isotopes of metals and semimetals with $I = \frac{1}{2}$.

Element		Abundance, %	Magnetic moment	Type of crystal lattice
Cd *	111	12.75	−0.59	hcp
	113	12.26	−0.62	
Sn *	115	0.33	−0.92	α Sn – diamond structure
	117	7.51	−1	β Sn – complex
	119	8.45	−1.05	
Pt *	195	35.2	0.61	fcc
Tl *	203	29.5	1.62	α Tl hcp
	205	70.5	1.63	β Tl fcc
Pb	207	22.6	0.59	fcc

*The remaining stable isotopes have $I = 0$.

and $I_{jy'_j}$ have matrix elements only between the states for which $|m_j - m'_j| = 1$. Thus, for integer spin these operators contain no static elements at all. For a half-integer spin, however, the following elements are independent of time:

$$\langle \pm {}^1/_2 | I_{jx'_j} | \mp {}^1/_2 \rangle = {}^1/_2 (I + {}^1/_2), \qquad \langle \pm {}^1/_2 | I_{jy'_j} | \mp {}^1/_2 \rangle = \pm {}^1/_2 (I + {}^1/_2)\, i. \tag{13}$$

For the frequency shift we obtain in accord with (5)

$$\frac{\delta\omega_{\text{dip}}}{\omega_0} = \frac{2}{3}(\hbar\gamma_\mu\gamma_n)^2 I(I+1)\,\omega_0^{-2} \times \sum_j \left[(1-n_{jz}^2) + \beta_I \frac{3}{16}\frac{I+{}^1/_2}{I(I+1)}(1+n_{jz}^2) \right] r_j^{-6}, \tag{14}$$

where $\beta_I = 0$ for integer spins and $\beta_I = 1$ for half-integer spins. For cubic crystals and polycrystals, this expression takes the form

$$\frac{\delta\omega_{\text{dip}}}{\omega_0} = \frac{2}{3}(\hbar\gamma_\mu\gamma_n)^2 I(I+1)\,\omega_0^{-2} \left[\frac{2}{3} + \beta_I \frac{I+{}^1/_2}{4I(I+1)} \right] \sum_j r_j^{-6}. \tag{15}$$

The frequency shift in weak magnetic fields is thus isotropic.

The equations obtained pertain to nondiffusing muons. It was shown in Ref. 3 that the precession frequency of rapidly diffusing muons ($\lambda \gg \gamma_\mu b, \gamma_n b$, where λ is the frequency of the muon hops over the interstices) differs from ω_0 by an amount

$$\delta\omega_{\text{dip}}/\omega_0 = {}^1/_3 \gamma_\mu^2 \langle b^2 \rangle / (\lambda^2 + \omega_0^2). \tag{16}$$

Using the calculation method proposed in these references, it is easy to obtain a generalization of Eq. (16) to include the case when the relation between λ and the characteristic precession frequencies of the nuclear spins is arbitrary:

$$\delta\omega_{\text{dip}}/\omega_0 = {}^1/_2 \gamma_\mu^2 [\langle b_{x\,\text{st}} \rangle + \langle b_{y\,\text{st}} \rangle] / (\lambda^2 + \omega_0^2), \tag{17}$$

where $b_{x\,\text{st}}$ and $b_{y\,\text{st}}$ must be taken to mean that part of the dipole field which does not manage to change during the time $\tau = \lambda^{-1}$ spent by the muon in one interstice. Although Eq. (17) was obtained under the assumption $\lambda \gg \gamma_\mu b$, if we put $\lambda = 0$ in it it goes over into Eq. (5) which is valid for muons at rest. It can thus be assumed that Eq. (17) can be used for interpolation at arbitrary diffusion velocities.

The authors know of only one study[4] in which separate investigations were made of the temperature dependences of the relaxation rate and of the precession frequency. Two vanadium samples with different impurity densities were investigated in a field of 70 G. It was found that at $T = 10$ K the depolarization rate is $\Lambda \approx 0.4\,\mu\text{sec}^{-1}$ for one sample and $\Lambda \approx 0.35\,\mu\text{sec}^{-1}$ for the other, while at $T = 300$ K the respective values were $\Lambda \approx 0.1\,\mu\text{sec}^{-1}$ and $\Lambda \approx 0.04\,\mu\text{sec}^{-1}$. It follows from the results of the experiment that the muons are at rest at 10 K and diffuse rapidly at 300 K. The precession frequency in the two samples is approximately the same, 6.14 rad/μsec at 10 K and 6.08 rad/μsec at 300 K. It can be seen that these results are in splendid agreement with our relation $\delta\omega_{\text{dip}} = 2\sigma^2/\omega$. Final conclusions, however, call for systematic investigations.

Information on the points of muon localization, on the distortion of the crystal lattice by the muons, on the diffusion velocity, etc. are obtained at present from data on the muon depolarization rates. Our equations show that the same information can be obtained also in precision measurements of the muon precession frequency. It is possible that this alternate method will turn out to be even more convenient for experimenters since, as noted earlier, the precession frequency can be determined with much higher accuracy than the damping rate.

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