Concerning the existence of muonium in metals

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The Wangsness-Bloch equations for muonium are solved in the presence of a magnetic field with allowance for the two-time relaxation of the μ^+-meson spin. It is shown that an analysis of the average polarization of the μ^+ meson in superconductors should reveal whether a muonium atom is produced in a metal. It is noted that in a rather large number of cases the two-time relaxation must be taken into account in the analysis of the average polarization.

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It is not known at present whether a muonium atom is produced in metals. It is obvious that this question is practically identical with the question of the charge state of the proton in metals. It is this circumstance which determines the importance of the problem. Indeed, the question of the behavior of monatomic hydrogen (of the proton!) in metals is extremely important for a large group of most varied problems in applied and theoretical metal physics. However, despite the very large number of investigations, this problem has not yet been solved. Theoretical calculations are willy-nilly based on rather crude models, and the experiments can be ambiguously interpreted. As a result, directly contradictory statements can be encountered both in the experimental and in the theoretical literature (see, e.g., [1-3]).

We propose here a method that makes it possible to detect the existence of a Mu atom by analyzing the polarizations of the μ^+ meson in metals. We note immediately that the corresponding experiments must be carried out in superconductors, and we therefore confine ourselves to an analysis of the μ^+-meson polarization in metals in the absence of an external magnetic field.

Following[4-6], we use the Wangsness-Bloch formalism. Generally speaking the use of the Wangsness-Bloch equations to describe the spin density matrix in a metal calls for a special justification. However, in the absence of external magnetic fields only one requirement must be satisfied, namely the condition $T \gg \hbar\omega_0$, where T is the temperature and ω_0 is the characteristic frequency of the hyperfine splitting in the muonium atom. For muonium in vacuum we have $\omega_0 = 2.8 \times 10^{10}$ sec^{-1} and $\hbar\omega_0 = 0.184°$K. In metals, if muonium does exist, a noticeable decrease of ω_0 and hence of the critical temperature should be expected. It is known that the experiments at high temperature do not reveal whether muonium exists in metals.[4-6] Indeed, since in metals the metals the muonium electron-spin relaxation frequency is large in comparison with the muonium hyperfine-splitting frequency (simple estimates show that $\nu \sim 10^{12}-10^{14}$ at room temperatures), the spin-spin coupling is broken and, even of the muonium atom exists, the muon behaves spinwise almost like a free particle.

We assume now that muonium does exist in metals and consider the behavior of the muonium polarization. In contrast to[4-7], we consider Wangsness-Bloch equations in which account is taken of the muon spin relaxation as a result of direct interaction of its magnetic moment with random magnetic fields in the medium (we shall henceforth call this mechanism "direct relaxation"). We write down the Wangsness-Bloch equations for the muonium spin density matrix, assuming the following: the medium is isotropic; the relaxation rate depends linearly on the muonium density matrix; the interaction of the μ^+ meson and of the electron with the medium is incoherent. It is easily seen that under these assumptions the most general form of the equations is

$$\frac{\partial\hat{\rho}}{\partial t}=-\frac{i}{\hbar}[\hat{H},\hat{\rho}]+\frac{\nu_e}{2}(\hat{\sigma}_e\hat{\rho}\hat{\sigma}_e-3\hat{\rho})+\frac{\nu_\mu}{2}(\hat{\sigma}_\mu\hat{\rho}\hat{\sigma}_\mu-3\hat{\rho}). \tag{1}$$

Here $\hat{H} = \frac{1}{4}\hbar\omega_0\hat{\boldsymbol{\sigma}}_e\hat{\boldsymbol{\sigma}}_\mu$ is the spin Hamiltonian of the system; $\hat{\boldsymbol{\sigma}}_e$ and $\hat{\boldsymbol{\sigma}}_\mu$ are the Pauli operators for the electron and muon, and ν_e and ν_μ are the relaxation frequencies of the electron and muon spins. We note that in our case, when $\nu_e \gg \nu_\mu$, allowance for the coherent interaction of the electron and the muon with the medium leads effectively to corrections that are not very significant.

In[4-6], where the depolarization of the μ^+ meson was investigated, the relaxation due to direct interaction of the muon magnetic moment with the random fields of the medium was neglected, since ν_μ is much smaller than ν_e. Indeed, it can be readily shown[8] that the ratio ν_e/ν_μ is at least of the order of the ratio of the squares of the corresponding magnetic moments:

$$\nu_e/\nu_\mu \sim (m_e/m_\mu)^2 \approx 4\cdot 10^4.$$

However, when $\nu_e \gg \omega_0$ and the spin coupling of the μ^+ meson with the rapidly relaxing electron spin is broken, it is necessary to take into account also the direct interaction of the muon magnetic moment with the medium.

Various representations can be used to solve Eqs. (1). Definite advantages are offered by a representation in which the muonium spin-density matrix is in the form of an expansion of orthonormalized spin operators[5-7]:

$$\hat{\rho}=\sum_{\varkappa,k=0}^{3}\rho_{\varkappa k}u_\varkappa\otimes u_k. \tag{2}$$

Here $u_0 = \chi/\sqrt{2}$, $\mathbf{u} = \hat{\boldsymbol{\sigma}}/\sqrt{2}$, χ is a unit two-by-two matrix, the Greek indices label the muon, and the Latin ones the electron. It is easily seen that in this representation the polarization of the μ^+ meson in the muonium is $P(t) = \rho_{10}(t)$, where the subscript 1 corresponds to the axis along which the muon is initially polarized. In the absence of an external magnetic field, Eqs. (1) reduce in this representation to a simple linear system:

$$\begin{aligned}
\frac{d\rho_{10}}{dt}&=\frac{\omega_0}{2}(\rho_{32}-\rho_{23})-\nu_\mu\rho_{10}, & \frac{d\rho_{23}}{dt}&=\frac{\omega_0}{2}(\rho_{10}-\rho_{01})-(\nu_e+\nu_\mu)\rho_{23},\\
\frac{d\rho_{01}}{dt}&=\frac{\omega_0}{2}(\rho_{23}-\rho_{32})-\nu_e\rho_{01}, & \frac{d\rho_{32}}{dt}&=\frac{\omega_0}{2}(\rho_{01}-\rho_{10})-(\nu_e+\nu_\mu)\rho_{32},
\end{aligned} \tag{3}$$

with initial conditions $\rho_{10}(0) = 1$ and $\rho_{\varkappa k}(0) = 0$. Since Eqs. (3) for the depolarization of the muon spin contain two relaxation parameters, we shall henceforth refer also to the process of "two-time relaxation" of the μ^+-meson spin in the muonium.

Before we proceed to an analysis of the solution of the system (3), we shall see what changes in the formulas for the experimentally observed quantities result from taking into account the direct relaxation of the μ^+-meson spin. In the analysis of the observed polarization it must be borne in mind that the muonium can enter in a chemical reaction and form a diamagnetic compound. This important circumstance was first emphasized by Nosov and Yakovleva.[5] A phenomenological theory that makes it possible to use the observed polarization to analyze chemical reactions of muonium was developed in[9], but no account was taken there of the muon polarization relaxation due to direct interaction of its magnetic moment with the medium.

It is well known (see, e.g.,[10-12]) that the law governing the asymmetry of the distribution of the μ^+-meson-decay positrons, under the condition that positrons of all energies are registered, leads to the relation

$$\frac{P(t)}{3}=\frac{dN_-(t)-dN_+(t)}{dN_-(t)+dN_+(t)}. \tag{4}$$

Here $dN_-(t)$ and $dN_+(t)$ are respectively the numbers of the counts in the interval dt in counters occupying a small solid angle and disposed symmetrically relative to the initial muon-polarization vector ahead and behind the target, respectively. In addition to this quantity, interest attaches in the experiments also to the time average of the polarization

$$\langle P\rangle=\frac{3(N_--N_+)}{N_0}=\frac{1}{\tau_\mu}\int_0^\infty P(t)e^{-t/\tau_\mu}dt, \tag{5}$$

where N_0 is the total number of the decays registered in the experiment, N_0 and N_+ are the total numbers of decays backward and forward. If the relaxation after the entry of the muonium in the chemical reaction is neglected, then the formula for $\langle P\rangle$ is trivial:

$$\langle P\rangle=\frac{1}{\tau}\int_0^\infty \rho_{10}(t)e^{-t/\tau}dt. \tag{6}$$

Here $1/\tau - 1/\tau_\mu + 1/\tau_1$, where $\tau_\mu = 2.2\times10^{-6}$ sec is the average lifetime of the muon and τ_1 is the average lifetime of the free muonium atom prior to its entry into a chemical reaction in which a diamagnetic compound is produced.

If account is taken of the μ^+-meson spin relaxation in the chemical compound, the corresponding formulas become somewhat more complicated. It can be readily verified that they take the form

$$P(t)=\rho_{10}(t)e^{-t/\tau_1}+\frac{1}{\tau_1}\int_0^t e^{-t'/\tau_1}\rho_{10}(t')k(t-t')dt'. \tag{7}$$

The kernel $k(t-t')$ determines here the law governing the decrease of the polarization in the chemical compound. If there is no depolarization in the chemical compound and $k(t-t')\equiv 1$, then by substituting (7) in (5) and then changing the order of integration we obtain directly formula (6). As seen from the system (3), in our case we have $k(t)=\exp(-\nu_\mu t)$, and we readily get

$$\langle P\rangle=\left[\frac{1}{\tau_\mu}+\frac{1}{\tau_1}\frac{1}{1+\nu_\mu\tau_\mu}\right]\int_0^\infty \rho_{10}(t)e^{-t/\tau}dt. \tag{8}$$

To determine finally when and to what extent the allowance for the two-dimensional relaxation can influence the results, it is necessary to solve the system (3), inasmuch as besides modification of the numerical factor preceding the integral in (8) in comparison with (6), a change takes place also in the character of the $\rho_{10}(t)$ dependence. The system (3) reduces to a third-order equation relative to $\rho_{10}(t)$ and we obtain in the standard manner

$$\rho_{10}(t)=Ae^{-\varkappa_1 t}+(Be^{i\omega t}-B^*e^{-i\omega t})e^{-\varkappa_2 t}. \tag{9}$$

Here

$$A=\frac{(\varkappa_2-\nu_\mu)^2+\omega^2-2}{\omega^2+(\varkappa_1-\varkappa_2)^2}, \tag{10}$$

$$B=\frac{(\nu_\mu-\varkappa_1)(\varkappa_2-\nu_\mu+i\omega)+2}{2i\omega(\varkappa_2-\varkappa_1-i\omega)}, \tag{11}$$

and κ_1, κ_2, and ω are connected with the roots of the corresponding characteristic equation by the relations

$$\varkappa_1=-\lambda_1,\quad -\varkappa_2\pm i\omega=\lambda_{2,3}. \tag{12}$$

The characteristic equation for the system (3) takes the form

$$(\lambda+\nu_\mu)(\lambda+\nu_e)(\lambda+\nu_e+\nu_\mu)+{}^1/_8\omega_0^2(2\lambda+\nu_e+\nu_\mu)=0. \tag{13}$$

The roots of this equation are determined by the known Cardan formulas. In the case when $\nu_e/\nu_0 \ll 1$ or $\nu_e/\nu_0 \gg 1$ we have

$$\begin{gathered}\varkappa_1\approx\nu_\mu+\frac{\nu_e/2}{1+4\nu_e^2/\omega_0^2},\\ \varkappa_2=\nu_e+\nu_\mu-\frac{\varkappa_1}{2}\approx\nu_e\left(1-\frac{1/4}{1+4\nu_e^2/\omega_0^2}\right),\\ \omega^2=\frac{\nu_e+\nu_\mu}{\varkappa_1}\left(\nu_e\nu_\mu+\frac{\omega_0^2}{4}\right)-\varkappa_2^2.\end{gathered} \tag{14}$$

At $\nu_e/\nu_0 \sim 1$ the error in the approximate formula (14) for κ_1 does not exceed 30%.

With the aid of (7)–(14) we can easily obtain the expression for the polarization P(t) in the absence of external fields also when account is taken of the direct relaxation of the μ^+-meson spin. However, the formula for P(t) is quite cumbersome and will not be written out here, all the more since the presently employed experimental technique is incapable of separating the high-frequency components of the polarization (the characteristic resolution time of the apparatus is $\Delta t \sim 10^{-9}$ sec). After averaging over an interval on the order of Δt, the observed time dependence of P(t) is given by

$$\begin{gathered}P(t)=\left[1-\frac{\tau_1^2\omega_0^2/2}{(1+\tau_1\varkappa_2-\tau_1\nu_1)^2+\tau_1^2\omega^2}\right]\frac{e^{-\nu_1 t}}{1+\tau_1\varkappa_1-\tau_1\nu_\mu}\\ +A\frac{\tau_1(\varkappa_1-\nu_\mu)}{1+\tau_1\varkappa_1-\tau_1\nu_\mu}\exp\left\{-\left(\frac{1}{\tau_1}+\varkappa_1\right)t\right\}.\end{gathered} \tag{15}$$

Accordingly we obtain for the average polarization $\langle P\rangle$ the simple formula

$$\frac{\langle P\rangle(1+2\nu_\mu\tau_\mu)}{1-\langle P\rangle(1+2\nu_\mu\tau_\mu)}=\frac{2(1+2\nu_\mu\tau_\mu)(1+2\nu_\mu\tau+2\nu_e\tau)}{\tau^2\omega_0^2}+\frac{1+2\nu_\mu\tau}{1+2\nu_e\tau}. \tag{16}$$

For the sake of simplicity we have assumed throughout that the μ^+-meson spin relaxation frequency ν_μ remains unchanged when the μ^+ meson enters into a chemical compound. The contrary assumption leads to an inessential change of formulas (15) and (16).

Comparing (16) with the analogous formula for the case when $\nu_\mu = 0$ (see[9]) we see that the main but generally speaking significant difference lies in the change of the form of the left-hand side of the equation. In particular, as shown by experiment,[10,12,13] situations with $\nu_\mu = 10^5\ \text{sec}^{-1}$ are realistic in a great variety of media. In this case $2\nu_\mu\tau_\mu \lesssim 0.44$ and must be taken into account. On the other hand if the free muonium has a long lifetime ($\tau_1 \geq \tau_\mu$) and the condition $2\nu_\mu\tau_\mu \lesssim 1$ is satisfied, a correction term must be taken into account also in the right-hand side of the equation. It can therefore be concluded that situations in which the direct relaxation of the muon spin must be considered when calculating the average polarization are quite realistic.

We proceed now to analyze the experimental possibilities in ordinary metals and superconductors. In ordinary metals, as already indicated, the electron spin relaxation frequency is $\nu_e \gg \omega_0$ and therefore the relaxation picture is determined entirely by the direct interaction of the muon spin with the medium. The large value of ν_e in metals is due to exchange scattering by free electrons of the metal. Since the electrons participating in this process have a momentum close to the Fermi momentum, it is obvious that ν_e changes with changing temperature in proportion to T. Estimates show that ν_e can become comparable in order of magnitude with ω_0 only at very low temperatures, on the order of 10^{-3}—10^{-2} °K, although the uncertainty in the value of ω_0 in metals (if, of course, muonium exists) does not permit any concrete conclusions to be drawn.

Thus, "detection" of muonium in ordinary metals is quite problematic. In superconductors, however, the situation is much more favorable. As is well known, the nuclear-spin relaxation rate at temperatures much lower than T_c decreases roughly speaking like $\nu \sim e^{-\Delta/T}$, where Δ is the width of the gap.[13,14] Qualitatively this is explained by the fact that the number of free electrons in a superconductor decreases exponentially with temperature. Although the quantitative calculation of the electron-spin relaxation rate in the electronic subsystem in a superconductor is difficult, since the BCS perturbation theory used to calculate the nuclear-spin relaxation rate is not applicable, the qualitative regularity should, naturally, remain in force. Thus, by using temperatures much lower than T_c one can decrease the electron-spin relaxation rate by several orders of magnitude. (We note that an empirical relation $\Delta \approx 2T_c$ at $T < T_c$ holds for many superconductors).

Thus, if the muon does produce muonium in the superconductor, the picture of the relaxation should be determined by (16) and the residual polarization, equal approximately to unity in a normal metal, will decrease sharply to values close to 1/2 (see[5,6,9]) when the region where $\nu_e \ll \omega_0$ is reached as the temperature is lowered. Depending on the value of τ_1, it can then turn out that $\langle P \rangle$ goes through a minimum in the intermediate region.[6] However, if the muon and electron do not form a bound state in the superconductor, then the polarization remains close to unity and responds little to a temperature change. In fact, the rates of relaxation of the nuclear spin in the electron system at low temperature are small and will not lead to a noticeable change of the polarization during the possible time of muon observation ($t \sim 10^{-5}$ sec). It must be emphasized that the described picture will be observed only if the magnetic moments of the superconductor nuclei are equal to zero and there is no dipole-dipole relaxation of the electron and muon spins. It is therefore necessary to choose for the experiment suitable isotopes of the superconductors. There are many known superconductors with isotopes that have zero nuclear spin, for example Pb (T_c = 7.2), Hg (T_c = 4.2), Zn (T_c = 0.88), Sn (T_c = 3.7), and others. (A complete table of superconducting elements is given, e.g., in the monograph[15]). Therefore the choice of suitable objects for the experiments entails no difficulty.

We note in conclusion that for a complete analysis it is of great interest to organize parallel experiments on superconductor isotopes with nonzero nuclear spins. The rate of dipole relaxation of the muon spin, as shown by estimates and experiment, should be of the order of 10^{-4}—10^{-6} sec. Accordingly, one should expect values 10^{-8}—10^{-10} sec for the electron-spin dipole-relaxation rate. If muonium does not exist, then when measuring P(t) and $\langle P \rangle$ at temperatures much lower than T_c we can count on measuring the rate of the dipole-dipole relaxation of the nuclear spin (or, in other words, T_2) in bulky superconductor samples. Insofar as we know, there is no other analogous possibility. If muonium does exist, then, by performing the measurements in isotopes with different magnetic moments and thus varying ν_e it is possible, first, to determine parameters ω_0 and τ_1 for muonium from the measurements of P(t) and $\langle P \rangle$, and second, to obtain the dependence of ν_e on the value of the magnetic moment of the nuclei.

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100