Theory of the behavior of the spin polarization of negative muons stopped in solids with nonzero nuclear spin

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After a μ^- meson has been captured by an atom in a solid with charge Z of the nucleus, an impurity is formed; it is a μ-nucleon atom or an ion with mesic atom nucleus with charge $Z-1$. A study is made of the effect of the electron shell of the impurity and the conduction electrons on the behavior of the spin polarization of the μ^- meson with allowance for the hyperfine structure of the mesic atom nucleus, a conversion Auger transition being possible between the components of the hyperfine structure. General expressions are obtained for the muon spin polarization, and some specific examples of the possible use of μ^- mesons for investigations in solid-state physics are considered.

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1. The investigation of materials by observation of the behavior of muon polarization (μSR) has become one of the basic experimental methods of solid-state physics. Both in the Soviet Union and abroad, the use of positive muons is now being augmented by an extensive program of investigations by means of negative muons. From the simple observation of the residual polarization in the first studies, a detailed bibliography of which can be found in the review of Ref. 1, the experimental groups are now turning to the measurement of more subtle effects. For example, observations are now made of the muon spin relaxation times T_1 and T_2, the paramagnetic shift and the Knight shifts, and hyperfine anomalies. The measurements are made in a wide range of temperatures; the necessary references to this work can be found in the review paper Ref. 2.

From the theoretical point of view, μSR methods for solids have much in common with the method of nuclear magnetic resonance (NMR) for impurities. However, for target materials with nuclear spin $i \neq 0$ there are important differences. In this paper, we shall not be concerned with questions associated with the phenomena that occur during the stopping, capture, and cascade processes associated with the stopping of negative muons in a target. We shall consider the spin behavior of the muon once it has reached the $1s$ orbit (K shell) of the mesic atom. The components of the muon spin polarization vector $\mathbf{P}(t)$ oscillate because of the interaction with the external fields and the fields due to the hyperfine interactions with the electrons. In addition, $\mathbf{P}(t)$ changes because of conversion Auger transitions between the components of the hyperfine structure of the mesic atom.[3,4] It is shown in Ref. 5 that external fields can change the conversion rate.

We shall show that different relationships between the conversion rates R and the frequency of the $\mathbf{P}(t)$ oscillations significantly change the amplitude and time dependence of the muon spin polarization vector. In addition, R depends strongly in some cases on the state of the outer electron shells and the chemical composition of the target, which, in its turn, changes the effective lifetime of a muon captured by the same element in different chemical compounds. The present paper is devoted to a consistent theory with allowance for these effects, and also to a discussion of their possible uses.

We shall restrict ourselves to taking into account the influence of the electron shell and the conduction electrons on the spin of the μ^- meson, assuming that the back reaction of the muon spin on the behavior of the electrons in the solid is negligible. Such an assumption is quite unjustified for gaseous media, but, as a rule, it is justified in solids at not too low temperatures.

After the μ^- meson has been captured by an atom with charge Z in the solid, an impurity μ-nucleon atom or an ion with charge $Z-1$ is formed. If the impurity does not have a completely closed electron shell, the projection of the total angular momentum J of the resulting paramagnetic center can vary rather rapidly because of the interaction with the environment with characteristic relaxation time τ_e. When the relaxation time τ_e of the angular momentum J of the electron shell is much shorter than the characteristic time of the hyperfine interaction of this shell with the spin I of the impurity nucleus, it is not at all possible, in contrast to a free atom, to speak of the total angular momentum $\mathbf{F}=\mathbf{I}+\mathbf{J}$ of the system. The behavior of the electron shell is then entirely governed by the interaction with the environment. This may be the exchange interaction with the conduction electrons in a metal or a semiconductor or any other interaction considered in ESR theory that leads to a short relaxation time τ_e of the electron angular momentum of the paramagnetic center.

The "nucleus" of the μ-nucleon atom is a mesic atom in whose ground state the total spin I is made up of the spin $s=\frac{1}{2}$ of the muon and the spin i of the target nucleus. If $i \neq 0$, then the nucleus of the mesic atom may be in one of two states of a hyperfine doublet. For $Z<30$, the energy difference in the hyperfine doublet is $\Delta\varepsilon \sim 1-10^3$ eV. From the upper state with spin I_U it is possible for there to be a conversion MI Auger transition to the state with spin I_L, energy being transferred to an Auger electron.

The effect of the electrons on the nucleus of the μ-nucleon atom is manifested in the form of effective

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fields due to the hyperfine interaction of the electrons with the magnetic and quadrupole moments. The conversion transition rate R depends on the wave functions of the electrons of the environment. Different populations of the electron spin states and the selection rules associated with angular momentum conservation in the MI Auger transition lead to a dependence of the transition rate on the polarization of the electron shells from which the Auger electron is stripped. If the paramagnetic shell has an appreciable polarization, the closed inner nlj shells acquire a polarization $\mathscr{P}^{nlj}$ through the configuration interaction. As is shown by the calculations of Refs. 6 and 7, the polarization of the core electrons for the L_I shell of the elements from B to F reaches several percent. A similar situation obtains for the M_I and N_I shells for elements of the third and fourth rows of the periodic table. It should be noted that in ferromagnetic substances the same effects lead to polarization of the inner shells, and the polarization of the conduction electrons is already 5–10%.

We note also that the effects considered here must also be manifested in an MI conversion Auger transition of the excited nucleus of an ordinary atom, but these effects will be manifested much more weakly since for nuclear transitions $\Delta\varepsilon \gtrsim 10$ keV and the Auger electrons are stripped mainly from deep shells with polarization $\mathscr{P}^{nlj} \ll 10^{-3}$. If, however, Auger electrons are detected and separated according to their energies, as, for example, in Ref. 8, then for the electrons of the outer shells one can obviously observe effects associated with the change in the conversion transition rates.

In μ-nucleon atoms of light elements, the transition energy is $\Delta\varepsilon \gtrsim 10$ eV and Auger electrons can be stripped from the comparatively "crumbly" shells, whose polarization may be of order $\mathscr{P} = 10^{-3} - 5\cdot 10^{-2}$, and, as will be shown, the effects considered in the present paper can be observed and used to investigate the properties of solids at the already existing technological level of μ-meson experiments.

The following circumstance may be very helpful. Having reached the K orbit in a time shorter than 10^{-10} sec, retaining at the same time 3–20% of its initial polarization, the meson may decay with characteristic time $\tau_\mu = 2.2\times 10^{-6}$ sec or be captured by a nucleus at the rate Λ_I, which depends strongly on the total spin $I = i \pm \frac{1}{2}$ of the mesic atom. The difference $\Delta\Lambda^c = \Lambda^c_{i+1/2} - \Lambda^c_{i-1/2}$ between the rates of the K captures is usually estimated in V–A theory and for light elements is two orders of magnitude less than the rate of conversion Auger transitions.[4] As in the case of free μ-nucleon atoms, one can relate $\Delta\Lambda^c$ to the change in the conversion rate for different mutual orientations of the spin polarizations of the μ^- meson and the electrons of the medium. Under experimental conditions, the change in the direction of the polarizations of the electrons of the medium, which is determined by the direction of the external magnetic field, is usually not associated with the introduction of systematic errors in the measurements, and this opens up the possibility of a compensation experiment permitting estimation of the spin constants of the weak interaction for light nuclei. As we shall show, such a compensation experiment can be used to determine the polarization of the individual electron shells.

2. We now obtain the basic relations that describe the system. The Hamiltonian of the nucleus of the μ-nucleon atom or ion must take into account the hyperfine structure of the mesic atom, the interaction of the nucleus with the magnetic field $\mathbf{b}$ produced by the external sources and all the dipoles of the sample, and the interaction of the quadrupole moment of the "nucleus" with the gradients of the electron fields:

$$H_0 = a_{\mu i}\hat{\mathbf{s}}\hat{\mathbf{i}} - 2\mu_-\hat{\mathbf{T}}\mathbf{b} + Q_{\alpha\beta}q_{\alpha\beta}{}^{\rm cr}/6, \tag{1}$$

where

$$\hat{\mathbf{T}} = \hat{\mathbf{s}} + \xi\hat{\mathbf{i}}, \quad \xi = \mu_i/2i\mu_-, \tag{2}$$

$q^{\rm cr}_{\alpha\beta}$ are the gradients of the external crystal fields at the the mesic atom, and $\hat{Q}_{\alpha\beta}$ is the quadrupole moment tensor of the nucleus of the mesic atom. The constant $a_{\mu i}$, which corresponds to the contact interaction of the spin $\mathbf{s}$ of the μ^- meson with the spin $\mathbf{i}$ of the nucleus, must take into account the finite size of the nucleus, and its value can be estimated as

$$a_{\mu i} = 0.045\mu_i Z^4_{\rm eff}/(iZ)\ [\text{eV}].$$

The values of $Z^4_{\rm eff}$ are given, for example, in Refs. 4 and 9. Since the energy of the hyperfine splitting is $\Delta\varepsilon \gtrsim 1-10^3$ eV, and the rates of the conversion transitions are of order 10^5-10^9 sec^{-1}, the total spin $\mathbf{I} = \mathbf{s} + \mathbf{i}$ of the mesic atom can be regarded as a "good" quantum number for both the upper state, $|a_U\rangle = |I_U m_U\rangle$, and the lower state, $|a_L\rangle = |I_L m_L\rangle$. (For $\mu_i > 0$, $I_U = i + \frac{1}{2}$, and for $\mu_i < 0$, $I_U = i - \frac{1}{2}$.)

The interaction of the nucleus with the medium, which results in nonconservation of I_U and leads to the conversion Auger transitions, can be described in the nonrelativistic approximation, which is perfectly adequate for light atoms, by the operator

$$\hat{V} = \sum_e \frac{32}{3}\pi\mu_-\mu_B\delta(\mathbf{r}_e)\hat{\mathbf{T}}\hat{\mathbf{s}}_e + 4\mu_B\mu_-\hat{\mathbf{T}}\sum_e \frac{1}{r_e^3}\hat{\mathbf{t}}_e + \frac{1}{2}\hat{Q}_{\alpha\beta}\sum_e \frac{n_\alpha n_\beta}{r_e^3}, \tag{3}$$

where

$$\hat{\mathbf{t}}_e = \hat{\mathbf{l}}_e - \hat{\mathbf{s}}_e + 3\mathbf{n}(\hat{\mathbf{s}}_e\mathbf{n}), \quad \mathbf{n} = \mathbf{r}_e/r_e.$$

Here, the sum must be taken over all electrons of the atom and the conduction electrons. The nucleus can be regarded as a point, since the electrons of the outer shells responsible for the transitions have a wavelength or orbit radius much greater than the diameter ~10^{-11} cm of the mesic atom. Following the standard Weisskopf–Wigner procedure, we can, as in Ref. 5, show that the spin density matrix of the nucleus satisfies the equation

$$\frac{\partial}{\partial t}\hat{\rho}^I = -\Lambda_I\hat{\rho}^I - \frac{i}{\hbar}[\hat{H}^I, \hat{\rho}^I] + \hat{L}^{I,U}\hat{\rho}^U, \tag{4}$$

where the first term

$$\Lambda_I = \Lambda_I{}^c + 1/\tau_\mu + R' \tag{5}$$

on the right-hand side takes into account K capture, the decay of the μ^- meson, and the possibility of radiative conversion transitions, whose probability R' for light elements is negligibly small.[4] The matrix elements of the Hamiltonian $\hat{H}^I$ include the diagonal (in I) part of the perturbation operator (3) averaged over the states of the electrons, which are described by the functions $|e\rangle = |\mathbf{k}, l_e, j_e, m_j\rangle$ ($\mathbf{k}$ is the electron wave vector, and $\mathbf{j}_e = \mathbf{l}_e + \mathbf{s}_e$).

The last term on the right-hand side of (4) is an operator of Liouville type and describes the conversion transitions:

$$\hat{L}_{a_L a_L', a_U a_U'} = \sum_{e, e', \tilde{e}} \langle a_L \tilde{e} | \hat{V} | a_U e\rangle \langle e | \hat{\rho}_e | e'\rangle \langle a_U e' | \hat{V} | a_L' \tilde{e}\rangle. \tag{6}$$

Then

$$\langle a_L | \hat{L}^{LU} \hat{\rho}^U | a_L'\rangle = \hat{L}_{a_L a_L' a_U a_U'} \langle a_U | \hat{\rho}^U | a_U'\rangle, \tag{7}$$

$$\langle a | \hat{L}^{UU} \hat{\rho}^U | a_U'\rangle = -\frac{1}{2} \hat{L}_{a_L a_L a_U'' a_U} \langle a_U'' | \hat{\rho}_U | a_U'\rangle - \frac{1}{2} \langle a_U | \hat{\rho}^U | a_U''\rangle L_{a_L a_L a_U' a_U''}. \tag{8}$$

In Eqs. (6)–(8), $\tilde{e}$ and e are the quantum numbers describing the electron states $|e\rangle$ and $|\tilde{e}\rangle$ before and after the Auger transition, respectively, $|\tilde{e}\rangle = |\mathbf{k}, l_e, j_e, m_j\rangle$. In (7) and (8), summation over the repeated indices a_L and a_U'' is understood.

It is assumed that the interaction between the shell electrons is weak. Therefore, each shell makes an independent contribution to the conversion process. The density matrix of the electron states factorizes in this case into the density matrices of the nlj shells. The single-electron density matrix can be specified by the angular momenta of the shell. For our purposes, it is sufficient to know $n^{nlj} = N^{nlj}/N$, the population, $\mathscr{P}_\alpha^{nlj} = \mathrm{Sp}\{\hat{\rho}^{nlj} \hat{j}_\alpha\}/j$, the polarization, and

$$\pi_{\alpha\beta}^{nlj} = \mathrm{Sp}\{\hat{\rho}^{nlj} [\hat{j}_\alpha \hat{j}_\beta + \hat{j}_\beta \hat{j}_\alpha - {}^2/_3 j(j+1)\delta_{\alpha\beta}]/2\}. \tag{9}$$

The magnetic hyperfine interaction makes the main contribution to the rate of the conversion transitions. Therefore, we ignore the contribution from the quadrupole interaction [the last term in Eq. (3)]. In this case, the operator (6) can be written as

$$\hat{L}_{a_L a_L' a_U a_U'} = \Gamma_{\alpha\beta} \langle a_L | \hat{T}_\alpha | a_U\rangle \langle a_U' | \hat{T}_\beta | a_L'\rangle, \tag{10}$$

$$\Gamma_{\alpha\beta} = \sum_n{}' N^{n,0,1/2} \Gamma_{\alpha\beta}^{n,0,1/2} + \sum_{n, l\neq 0, j}{}' N^{nlj} \Gamma_{\alpha\beta}^{nlj}. \tag{11}$$

Here and in all that follows, the primed summation means that the summation is over the shells for which the electron binding energy is less than $\Delta\varepsilon$:

$$\Gamma_{\alpha\beta}^{nlj} = \Gamma^{nlj} \delta_{\alpha\beta} - (i) B^{nlj} j e_{\alpha\beta\gamma} \mathscr{P}_\gamma^{nlj} + C^{nlj} \pi_{\alpha\beta}^{nlj}. \tag{12}$$

For $l \neq 0$, the coefficients in Eq. (12) are

$$\Gamma^{n,l,l\pm 1/2} = \frac{(2l+1\mp 1)[1+2(2l+1\mp 1)(2l+1\pm 2)]}{24(2l+1)} \gamma_n(l,l) + \frac{3}{8}\frac{(2l+1\pm 3)}{(2l+1\pm 2)} \gamma_n(l, l\pm 2), \tag{13}$$

$$B^{n,l,l\pm 1/2} = \left[\frac{(2l+1\mp 1)^2}{2(2l+1)^2} \pm \frac{l}{4(2l+1)^2}\right] \gamma_n(l,l) \mp \frac{9}{8}\frac{(2l+1\pm 3)}{(2l+1\pm 2)^2} \gamma_n(l, l\pm 2), \tag{14}$$

$$C^{n,l,l\pm 1/2} = \frac{4(2l+1\mp 1)^2 - 1}{4(2l+1)^2} \gamma_n(l,l) - \frac{9}{4}\frac{\gamma_n(l, l\pm 2)}{(2l+1\pm 2)^2}. \tag{15}$$

In Eq. (12) and what follows, i is the imaginary unit, and

$$\gamma_n(l, l') = 2\pi\hbar(4\mu_B\mu_-)^2 \sum_\mathbf{k} \delta(\varepsilon_{\mathbf{k},e'} - \varepsilon_{n,e} + \Delta\varepsilon) |\langle \mathbf{k} l' | \frac{1}{r_e^3} | nl\rangle|^2. \tag{16}$$

For transitions from the state with $l = 0$, $\pi_{\alpha\beta}^{n,0,1/2} \equiv 0$, and the coefficients in Eq. (12) have the form

$$\Gamma^{n,0,1/2} = \gamma_n(0,2)/2 + \gamma_n(0,0)/4,$$

$$B^{n,0,1/2} = [\gamma_n(0,2) + \gamma_n(0,0)]/2, \tag{17}$$

$$\gamma_n(0,0) = 2\pi\hbar(32\pi\mu_-\mu_B/3)^2 |\psi_n(0)|^2 \times \sum_\mathbf{k} \delta(\varepsilon_{\mathbf{k},0} - \varepsilon_{n,0} + \Delta\varepsilon) |\psi_\mathbf{k}(0)|^2. \tag{18}$$

For transitions from the s shell to an s wave, the table of $\gamma_n(0,0)/4$ values for a number of elements is given in Refs. 4 and 5.

The tensor $\Gamma_{\alpha\beta}$ can always be represented by a sum of symmetric and antisymmetric components:

$$\Gamma_{\alpha\beta} = \Gamma(\delta_{\alpha\beta} - i e_{\alpha\beta\gamma} \mathscr{P}_\gamma' + \pi_{\alpha\beta}'),$$

$$\Gamma = \sum_{nlj}{}' \Gamma^{nlj} N^{nlj}, \quad \mathscr{P}_\gamma' = \Gamma^{-1} \sum_{nlj}{}' j B^{nlj} \mathscr{P}_\gamma^{nlj} N^{nlj}, \tag{19}$$

$$\pi_{\alpha\beta}' = \Gamma^{-1} \sum_{nlj}{}' C^{nlj} \pi_{\alpha\beta}^{nlj} N^{nlj}.$$

We recall that the largest contribution to the sum is usually made by the lowest ns shell,[4] and it is therefore helpful to express $\Gamma^{n,0,1/2}$ and $\mathscr{P}^{n,0,1/2}$ in terms of the densities of the electrons with different spin projections $|\psi_{n\uparrow}(0)|^2$ and $|\psi_{n\downarrow}(0)|^2$ at the nucleus:

$$\Gamma^{n0 1/2} = \frac{8}{9}\frac{\pi\alpha^4 z'}{m_\mu^2} \rho_n,$$

$$\rho_n = |\psi_{n\uparrow}(0)|^2 + |\psi_{n\downarrow}(0)|^2, \tag{20}$$

$$\mathscr{P}_z^{n0 1/2} = (|\psi_{n\uparrow}(0)|^2 - |\psi_{n\downarrow}(0)|^2)\rho_n^{-1}.$$

Substituting (19) in (10) and noting that

$$\langle a_U | \hat{\Gamma} | a_U'\rangle = \hat{L}_{a_L a_L, a_U' a_U} = \Gamma_{\alpha\beta} \langle a_U | \hat{T}_\beta | a_L\rangle \langle a_L | \hat{T}_\alpha | a_U'\rangle, \tag{21}$$

we write down for the density matrix of the upper state the equation

$$\frac{\partial}{\partial t} \hat{\rho}^U = -\frac{i}{\hbar} [\hat{H}_{\mathrm{eff}}^U, \hat{\rho}^U] - \frac{1}{2} \{\hat{\Gamma} + \Lambda_U, \hat{\rho}^U\}. \tag{22}$$

The diagonal matrix element of the perturbation operator $\hat{V}$ is included in the effective Hamiltonian $\hat{H}_{\mathrm{eff}}^U$, which now takes into account the contact and the orbital Knight shift as well as the polarizability of the inner shells:

$$\hat{H}_{\mathrm{eff}}^I = -2\mu_-\lambda_I \hat{\mathbf{I}} \mathbf{b}_{\mathrm{eff}} - \frac{e^2}{2}\frac{Q_I}{I(2I-1)} \hat{\Pi}_{\alpha\beta} q_{\alpha\beta}^{\mathrm{eff}}, \tag{23}$$

$$(b_\alpha)_{\mathrm{eff}} = b_\alpha - \frac{8}{3}\pi\mu_B \sum_{n,0,1/2} \mathscr{P}_\alpha^{n,0,1/2} \rho_n - 2\mu_B \sum_{n,l,l\pm 1/2} \mathscr{P}_\alpha^{n,l,l\pm 1/2} (l+1/2) \frac{2l+1\mp 1}{2l+1} \left\langle nl \left| \frac{1}{r^3} \right| nl \right\rangle, \tag{24}$$

$$\lambda_I=(2i+1)^{-1}\left[\pm1+\xi(2i+1\mp1)\right], \tag{25}$$

$$q_{\alpha\beta}^{\text{eff}}=q_{\alpha\beta}^{\text{cr}}+\frac{3}{2}\sum_{nlj}\frac{\pi_{\alpha\beta}^{nlj}}{j(j+1)}\left\langle nl\left|\frac{1}{r^3}\right|nl\right\rangle, \tag{26}$$

$q_{\alpha\beta}^{\text{eff}}$ is the tensor determined by the gradient of the electric field at the nucleus produced by the crystal lattice and the quadrupole polarizability $\pi_{\alpha\beta}^{nlj}$ of the electron shells, Q_I is the quadrupole moment of the nucleus of the mesic atom,[10,11] and

$$\hat{\Pi}_{\alpha\beta}=\frac{1}{2}\left[\hat{I}_\alpha\hat{I}_\beta+\hat{I}_\beta\hat{I}_\alpha-\frac{2}{3}I(I+1)\delta_{\alpha\beta}\right] \tag{27}$$

are the Cartesian components of the quadrupole polarization tensor of the state with given I.

The operator (21), which describes the conversion, can be written as

$$\hat{\Gamma}=R\left[\hat{E}\mp\frac{2\mathscr{P}_\alpha'\hat{I}_\alpha}{2i+1}-\frac{2\pi_{\alpha\beta}'\hat{\Pi}_{\alpha\beta}}{(2i+1)(2i+1\mp1)}\right], \tag{28}$$

$$R=\Gamma(1-\xi)^2\frac{2i+1\mp1}{2(2i+1)}. \tag{29}$$

Here and in what follows, the upper and lower signs are taken for nuclei with magnetic moment $\mu_i>0$ and $\mu_i<0$, respectively, and R is the conversion rate without allowance for polarization. A table of R values for a number of elements is given in [4].

In what follows, Eq. (22) will be solved for the case when the system has a symmetry axis of higher than the third order. The external magnetic field **b** is directed along this symmetry axis (the Z axis). Then all the operators in Eq. (22) are diagonal. Denoting $\Gamma_m=\langle m|\hat{\Gamma}|m\rangle$, we obtain

$$\hat{\rho}^U_{m,m'}(t)=\hat{\rho}^U_{m,m'}(0)\exp\left[-(i)\omega^U_{mm'}t\right], \tag{30}$$

$$\operatorname{Im}\omega^U_{m,m'}=-\Lambda_U-(\Gamma_m+\Gamma_{m'})/2. \tag{31}$$

The real part of the frequency corresponding to the eigenvalues of the Hamiltonian (23) is

$$\operatorname{Re}\omega^I_{m,m'}=-2\mu_-\lambda_I b_{\text{eff}}(m-m')+\frac{3}{4}\frac{e^2Q_Iq_{zz}}{I(2I-1)}(m'^2-m^2). \tag{32}$$

We now consider the evolution of the density matrix of the lower state. In this case, Eq. (4) acquires the inhomogeneous term

$$\langle a_L|\hat{L}^{L,U}\hat{\rho}^U|a_L'\rangle=\Gamma_{\alpha\beta}\langle a_L|\hat{T}_\alpha|a_U\rangle\langle a_U|\hat{\rho}^U|a_U'\rangle\langle a_U'|\hat{T}_\beta|a_L'\rangle \tag{33}$$

[see Eqs. (7) and (10)], which describes the arrival of particles in the lower state as a result of the conversion. The expression (33) is a known function of the time, since $\hat{\rho}_U(t)$ is determined by the relation (30). It can be directly verified that the solution of (4) for the lower state with the inhomogeneous term (33) is

$$\langle m|\hat{\rho}^L|m'\rangle=\hat{\rho}^L_{mm'}(0)\exp\left[-(i)\omega^L_{mm'}t\right]+\Gamma_{\alpha\beta}\langle I^Lm|\hat{T}_\alpha|I^Um''\rangle$$

$$\times\langle m''|\hat{\rho}^U(0)|m'''\rangle\langle m'''I^U|\hat{T}_\beta|I^Lm'\rangle f_{mm'}^{m''m'''}, \tag{34}$$

where

$$f_{mm'}^{m''m'''}=(i)\left(\omega^L_{mm'}-\omega^U_{m''m'''}\right)^{-1}\left\{\exp\left[-(i)\omega^L_{mm'}t\right]-\exp\left[-(i)\omega^U_{m''m'''}t\right]\right\}$$

$$\langle i\mp{}^1\!/_2m|\hat{T}_+|i\pm{}^1\!/_2,m'\rangle=\mp\delta_{m,m'+1}\frac{1-\xi}{2i+1}\left[(i+{}^1\!/_2\mp m)(i+{}^1\!/_2\mp m\pm1)\right]^{1/2}, \tag{35}$$

$$\langle i\mp{}^1\!/_2,m|\hat{T}_z|i\pm{}^1\!/_2m'\rangle=-\delta_{mm'}\frac{1-\xi}{2i+1}\left[(i+{}^1\!/_2)^2-m^2\right]^{1/2}.$$

In principle, the expressions (30) and (34) completely describe the behavior of the spin of the μ^- meson with allowance for the conversion Auger transitions.

3. We now discuss the physical consequences and some experimental applications associated with the possibility of changing the rate of the conversion Auger transitions. In the μSR method, the experimentally observed quantity is usually the time distribution of the number of μ^--decay electrons emitted in a definite direction **k**:

$$\frac{dN_e}{dt}=\frac{AN}{\tau_\mu}\left[n(t)+a\mathbf{k}\mathbf{P}(t)\right]. \tag{36}$$

The constants A and a are associated with the characteristics of the detecting counters, and N is the number of stoppings in the target. Thus, one measures $n(t)=n^U(t)+n^L(t)$ and $\mathbf{P}(t)=\mathbf{P}^U(t)+\mathbf{P}^L(t)$, where

$$n^I(t)=\operatorname{Sp}\hat{\rho}^I(t),$$

$$\mathbf{P}^I=2g_I\operatorname{Sp}\{\hat{\mathbf{I}}\hat{\rho}^I\},\quad g_{i\pm1/2}=\pm1/(2i+1). \tag{37}$$

As follows from (30) and (34), the population $n(t)$ is equal to

$$n(t)=\exp(-\Lambda^Lt)-\Delta\Lambda\sum_m\frac{e^{-\Lambda_Lt}-e^{-(\Lambda^U+\Gamma_m)t}}{\Delta\Lambda+\Gamma_m}\rho_{mm}{}^U(0). \tag{38}$$

It is made up of exponentials with different arguments, so that a muon in matter does not have a lifetime in the strict sense. For the upper state, the arguments of the exponentials may differ by amounts of order $R\mathscr{P}'$ or $R\pi'$ [see Eq. (28)], which for the light elements Be, B, ^{13}C, ^{14}N, F, ^{21}Ne, ^{25}Mg, and ^{57}Fe can be a few percent of Λ^U.

If it is difficult to separate from the decay curve the exponentials with nearly equal arguments, an effect can be detected by measuring the total probability of μ^- decay:

$$W_e\approx\tau_\mu^{-1}\Lambda_L^{-1}\left\{1-\frac{\Delta\Lambda}{\Lambda_U+R}\left[n^U(0)+\frac{R}{\Lambda_U+R}\left(\mathscr{P}_\alpha'\bar{P}_\alpha{}^U(0)\right.\right.\right.$$

$$+\frac{2\pi_{\alpha\beta}'\langle\Pi_{\alpha\beta}(0)\rangle}{(2i+1)(2i+1\mp1)}+\frac{1}{3}n^U(0)\mathscr{P}_z'^2\frac{R}{\Lambda_U+R}\frac{2i+1\pm2}{2i+1}$$

$$\left.\left.\left.+\frac{1}{30}n^U(0)\pi_{\alpha\beta}^2\frac{R}{\Lambda_U+R}\frac{(2i+1\pm2)\left[(2i+1)(2i+1\pm2)-3\right]}{(2i+1)(2i+1\mp1)^2}\right)\right]\right\} \tag{39}$$

or the probability of K capture: $W_K=1-W_e$. In (39), we have retained the terms up to those quadratic in $\mathscr{P}'$ and π'. Note that the existence of a symmetry axis is not required for the derivation of Eq. (39).

At low temperatures $T\sim1-10$°K and in external fields $b\sim10$ kG, a paramagnetic center for semiconducting boron compounds may have $\mathscr{P}'\sim0.01$, and the change in W_e will be of order 10^{-2}. In compounds of the type LiF one can expect values $\pi'\sim0.1$ for $^\mu$O, and the contribution from the last two terms in (39) will be important.

There will be a particularly strong variation of W_e

when the value of R itself changes; as can be seen from (29), (13), (16), and (18), R is sensitive to the form of the electron wave function. For the light elements listed at the start of this section, this means the wave functions of the outer electrons. For this reason, the conversion rate for these elements is sensitive to changes associated, for example, with the entry of a μ-nucleon atom into a chemical compound. There is a particularly strong change in R on ionization or when the μ-nucleon atom enters in to a strongly ionic chemical bond, since the binding energy of the remaining electrons of the ion is appreciably higher than in the atom. For example, in B_2O_3 there should be no conversion at all, since the energy released in an Auger transitions is appreciably less than the second ionization potential of the μ-nucleonic $^{11}Be^+$. It is convenient to compare the probabilities of the processes in a target of pure elements and the hydrides of the elements, since even in the case of capture by H the meson will be rapidly recaptured by the heavy element. In this case, the change in the probability of K capture in, for example, pure ^{11}B and its hydride, $\Delta W_K = W_K(^{11}B) - W_K(^{11}BH_3)$, will be of order 20%, while $\Delta W_e \approx 0.5\%$. For Be and BeH_2, $\Delta W_K \approx 21\%$ and $\Delta W_e < 10^{-3}$ and for ^{25}Mg and $^{25}MgH_2$ we have $\Delta W_K \sim 10^{-3}$ and $\Delta W_e \sim 1\%$. In graphite and methane with ^{13}C, $\Delta W_e \sim 1\%$.

We now discuss effects associated with measurement of the polarization. We begin the analysis by considering $P_z(t)$. As follows directly from the expressions (30) and (34) for the density matrices, the polarizations of the upper and lower states are

$$P_z^U(t) = \exp(-(\Lambda_U + R)t)\left[P_z^U(0) + 4\frac{n^U(0)Rt}{(2i+1)^2}\left(\mathscr{P}_z'\langle I_z^2(0)\right.\right.$$
$$\left.\left.\pm\frac{3}{2}\frac{\pi'\langle I_z\Pi_{zz}\rangle}{(2i+1\mp1)}\right)\right], \qquad (40)$$

$$P_z^L(t) = P_z^L(0)e^{-\Lambda_L t} \mp 2R\frac{2i+1\mp2}{(2i+1)^2}\sum_{m=-I_U}^{I_U} f_{mm}^{mm}(t)\rho_{mm}(0)\left[m + \mathscr{P}_z'\frac{2i+1\mp4m^2}{2(2i+1\mp1)}\right.$$
$$\left.\pm\pi' m\frac{1}{2i+1\mp2} - 3\pi'\frac{\langle m|\Pi_{zz}|m\rangle}{(2i+1\mp1)(2i+1\mp2)}\right], \qquad (41)$$

where

$$\pi' = \pi_{zz}' = -{}^1/_2\pi_{xx}' = -{}^1/_2\pi_{yy}'.$$

For the arbitrary values of the spin i of the nucleus, the expression for the polarization is cumbersome. Therefore, as an illustration, we give the formula for P_z in the simplest case $i = \frac{1}{2}$, which reflects qualitatively the time dependence of the polarization for arbitrary nuclei:

$$P_z(t) = \exp(-(\Lambda_U + R)t)\{P_z(0) + n^U(0)Rt[{}^2/_3\mathscr{P}_z'$$
$$+\mathscr{P}_z'\langle\Pi_{zz}^U(0)\rangle + {}^3/_2\pi'\langle I_z\Pi_{zz}^U(0)\rangle]\}. \qquad (42)$$

In the expression (42) and all that follow, we take into account only the terms linear in $\mathscr{P}_z'$ and π'.

In the case of nuclei with arbitrary values i of the spin, we can derive an expression for the polarization taking into account only the terms linear in $\mathscr{P}'_z$, $P_z^U(0)$, and $P_z^L(0)$. If we restrict ourselves to time $t \ll (\Lambda_U + R)^{-1}$, for which $f_{mm}^{mm} \approx t$, then the total polarization has the form

$$P_z(t) = P_z(0) - \Lambda_L t P_z^L(0) - \Lambda_U t P_z^U(0) + \frac{2}{3}Rt\mathscr{P}_z' n^U(0) - 2RtP_z^U(0)\frac{2i+1\mp1}{2i+1}. \qquad (43)$$

For times $t \gg (\Lambda_U + R)^{-1}$ and under the same restrictions as in (43), the polarization can be expressed as

$$P_z = P_z^L(0)e^{-\Lambda_L t} - \frac{Re^{-\Lambda_L t}}{R+\Lambda_U-\Lambda_L}\frac{2i+1\mp2}{2i+1}$$
$$\times\left[P_z^U(0) - \frac{1}{3}n^U(0)\mathscr{P}_z'\left(1 - \frac{R}{R+\Lambda_U-\Lambda_L}\frac{2i+1\pm2}{2i+1}\right)\right]. \qquad (44)$$

It is meaningful to use the expression (43) when the conversion rate is of the order of the μ^--meson lifetime. But if the conversion transition takes place during times shorter than the characteristic observation time in the experiment, which is determined either by the lifetime or the resolution of the instrument ($10^{-8}-10^{-9}$ sec), it is necessary to use the expression (44). In the first case, one can observe a nontrivial time dependence of the polarization. Even when $P_z^U(0) = P_z^L(0) = 0$ there is at short times a linear growth of the polarization with t. An estimate shows that for ferromagnetic substances like MnB and ^{57}Fe or the ferromagnet $CrCl_3$ the maximal value of the polarization is certainly greater than 10^{-3}, and after the maximum at $t \sim R^{-1}$ an exponential decay of the polarization at rate Λ_L commences. If at the same time the lifetime of the μ^- meson is short, it may be convenient to measure the time average of the polarization,

$$\langle P_z\rangle = \frac{1}{\tau}\int_0^\infty P_z(t)\,dt, \qquad (45)$$

for the measurement of which it is not necessary to bother about the time dependence, since it is sufficient to determine the number of decays in two directions for known number of μ^--meson stoppings in the target.

If the relaxation of the spin of the nucleus of the μ-nucleon atom leads to an appreciable change in the polarization during the μ^--meson lifetime, this effect can be taken into account by replacing Λ^I in Eqs. (42)–(44) by the sum $\Lambda^I + \tau_1^{-1}$, in which the relaxation time τ_1 must be calculated in the same way as the time T_1 in NMR theory.[6,7,12]

A general expression for the perpendicular, or oscillating, component of the polarization can, as in the case of (40) and (41) for $P_z(t)$, be readily obtained directly from the expressions (30) and (34) for the density matrix, the complex form of expression $P_+ = P_x + iP_y$ being convenient. However, there is no point in writing out the cumbersome expression, since allowance for the electron polarization is important only in the shift of the frequency (32), while in the amplitude of the oscillations it leads to small corrections. With allowance for these remarks, the expression for $P_+(t)$ can be written as

$$P_+(t) = P_+^L(0)\varphi^L(t) + P_+^U(0)\varphi^U(t) + P_+^U(0)\frac{2i+1\mp2}{2i+1}Rf(t), \qquad (46)$$

$$\varphi^I(t) = -\sum_{m=-I}^{I}\frac{3}{2}\frac{I(I+1)-m(m-1)}{I(I+1)(2I+1)}\exp[-(i)\omega_{m-1,m}^I t],$$

$$f(t) = \sum_{m=-(i+1/2)}^{i+1/2}\frac{6(i\pm m-{}^1/_2)(i\mp m\pm1+{}^1/_2)}{(4i^2-1)4i(i+1)(2i+3)}\{2(i+{}^1/_2\mp m)(i+{}^1/_2+m\mp1)f_{m-1,m}^{m-1,m}$$
$$+(i+{}^1/_2\mp m\mp1)(i+{}^1/_2\mp m)f_{m,m+1}^{m-1,m} + (i+{}^1/_2\pm m\mp2)(i+{}^1/_2\pm m\mp1)f_{m-2,m}^{m-1,m}\}. \qquad (47)$$

As is shown by estimates,[12-18] the probability of conversion (in unit time) may be of the order of the frequency of the oscillations resulting from the interaction of the quadrupole moment of the nucleus of the mesic atom with the gradients of the electric fields, and therefore both these effects must be taken into account at once for $i > 1/2$. A detailed analysis of quadrupole effects would go beyond the scope of the present paper; we merely note that for the most frequently encountered nuclear spin $i = 3/2$ the polarization $P_+(t)$ can be represented as

$$P_+(t) = \exp(-(\Lambda_s + R + (i)\omega^U)t)[A\cos(\Omega t + \varphi_1) + B\cos(\Omega t/3 + \varphi_2)] + \exp[-(\Lambda_L + (i)\omega^L)t]C\cos(\Omega t + \varphi_3), \tag{48}$$

in which, in the general case, the complex constants A, B, C and the phases $\varphi_1, \varphi_2, \varphi_3$ can be expressed in terms of $P_+^U(0)$ and $P_+^L(0)$ and all the remaining parameters of the problem $\Lambda_I, R, b_{\text{eff}}, \Omega = 3/8e^2Q_1q_{zz}$.

On the other hand, the part played by conversion is very important for understanding the "purely quadrupole" effects. For example, for b = 0 it significnatly changes both the amplitude and the phase of oscillations at the same frequencies. For example, for Na in the semiconducting compound Na_3Sb or for Cl in the compounds CCl_4 and $SiCl_4$, we can expect $R \gg \Lambda_U \sim \Lambda_L$, and for $t \gg R^{-1}$ when $\Omega \ll R$

$$P_+(t) \simeq [P_+^L(0) - P_+^U(0)]e^{-\Lambda_L t}\cos\Omega t, \tag{49}$$

and when $\Omega \gg R$

$$P_+(t) \approx [P_+^L(0) - 2P_+^U(0)/5]e^{-\Lambda_L t}\cos\Omega t. \tag{50}$$

According to the estimate of Refs. 19–23, the relation $P_+^L(0) \ll P_+^U(0)$ usually holds, and therefore the contribution to $P_+(t)$ from the initial polarization of the upper state will be predominant, and the amplitude of the oscillations will be determined by the ratio $x = \Omega/(R + \Lambda_U - \Lambda_L)$. Sometimes, Ω may depend appreciably on the temperature, and at phase-transition points very strongly, so that the amplitude of the oscillations will also change appreciably.

We now consider a different case when the dominant role is played by precession in a magnetic field, and the quadrupole effects can be ignored. This means that the precession frequency is much higher than the quadrupole frequencies of the oscillations, and the energy levels are equidistant. The functions $\varphi^I(t)$ and $f(t)$, which determine the precession in accordance with Eq. (47), take the form

$$\begin{gathered} \varphi^U(t) = \exp[-\Lambda_U + R + (i)\omega^U)t], \\ \varphi^L(t) = \exp[-(\Lambda_L + (i)\omega^L)t], \\ f(t) = (\varphi^U(t) - \varphi^L(t))[\Lambda_U + R - \Lambda_L + (i)(\omega^U - \omega^L)]^{-1}. \end{gathered} \tag{51}$$

If the field causing the precession is set equal to zero, then (46) with φ^I and f from (51) goes over into the expression obtained in Ref. 19. For $Z > 11$ the conversion rate satisfies $R > \Lambda^I$, and it is therefore convenient to write down (46) for $t \gg R^{-1}$:

$$P_+(t) = \left[P_+^L(0) - P_+^U(0)R\frac{2i+1\mp 2}{2i+1}[R + (i)(\omega^U - \omega^L)]\right] \times \exp[-(\Lambda_L + (i)\omega^L)t]. \tag{52}$$

As can be seen from (52), the precession amplitude depends on the frequency. This effect must be manifested particularly strongly in the metals Na, K, and Al, in which there are no quadrupole effects because of the cubic symmetry. For example, in Al the conversion rate is $R = 4.1 \times 10^7$ and $i = 5/2$, and therefore the influence of a magnetic field on the amplitude will be important for 1 kG, when the precession frequency is $\omega^L > 10^7$ sec^{-1}. The same effect will occur in compounds of these elements of the NaCl type with cubic lattice. The impurity that is formed will simulate an atom of an inert gas at a lattice site. Note that in the case of capture by a halogen atom the situation is entirely different because of the formation of a paramagnetic center and distortion due to the Jahn–Teller effect. A similar situation must obtain in ionic compounds of elements of the groups II–VI. Formation of a paramagnetic center with an As atom in ZnSe was observed in Ref. 24 by the double ESR method. The rapid depolarization on the capture of a μ^- meson by O in MgO and ZnO, also noted in Ref. 25, obviously indicates the formation of a paramagnetic center. If the τ_e^{-1} of the acceptor paramagnetic center is not large compared with ω_{st}, one must observe a paramagnetic shift of the precession frequency of order $206\hbar\omega_{\text{st}}/T$, where $\hbar\omega_{\text{st}} \sim 0.1$°K. Thus, even at high $T \sim 10^3$°K the frequency shift will be of order 2%. By raising the temperature, one can decrease the relaxation rate of the μ^--meson spin, and precession will be observed in both MgO and ZnO at $T \sim 1000$°K.

In ZnO, it is interesting to investigate precession when an ion is captured by a Zn atom. If the μ-nucleon ion $\mu_{\text{Cu}+}$ is in the same state as the ordinary impurity ion Cu^+, then the hyperfine coupling will be noticeable at low temperatures $T \sim 10$°K. The results of an experiment can be compared with the analogous measurements of the ESR method.[26]

For d metals, such measurements give information about the Knight shift at the impurity, which reaches 1%. For example, the Knight shift at V in Cr is 0.57% according to the NMR data of Ref. 29, and at Ni in Cu the shift is 1.28%.[27] In metals, in particular in Cu, measurement of the frequency shift and the damping rate of the polarization as functions of the temperature gives information about the exchange integral and, thus, about the Kondo temperature. In addition, in Cu one can expect different effects associated with the unfilled d shell of the μ-nucleon $^{\mu}$Ni at both low and high temperatures.

In magnetically ordered systems or for paramagnetic centers at low $T \lesssim b\mu_U$, the polarization of the electron shells is such that the effective field at the nucleus reaches 10^2–10^3 kG and the observation of precession becomes impossible. However, one can observe precession in the case of capture by a diamagnetic ion in a magnetically ordered compound provided the impurity that is formed is also diamagnetic. The diamagnetic shell is then polarized and one must observe a shift of the precession frequency similar to the paramagnetic shift observed in MnO.[28]

Evidence for appreciable polarizability in antiferromagnets with diamagnetic shell can be seen in the complete depolarization of the muon in Fe_2O_3,[25] whereas in the chemically analogous paramagnetic Mn_2O_3, Co_2O_3, and Ni_2O_3 there is an appreciable residual polarization. A similar situation obtains in ferromagnets. For example, at the B nucleus in MnB there is in accordance with the NMR data of Ref. 6 a field $b_{\rm eff}=23.7$ kG, and at Mn in Fe a field $b_{\rm eff}=227$ kG, so that in them there is no real possibility of observing precession. In these substances, one can directly measure the polarizability of the outer electron shells by observing the component of the muon spin polarization parallel to an external magnetic field. The time dependence and mean value of the longitudinal polarization are given by Eqs. (40)–(45) which contain, not the total field at the nucleus determined by the NMR method, but the polarizability of the shells from which an Auger transition is possible. As yet, no experiment has been made to measure the polarization of a μ^- meson induced by conversion transitions in a magnetized ferromagnet, though such experiments could give information about the exchange forces in magnetically ordered systems that is currently inaccessible by other methods.

There is no doubt that the examples listed here do not cover all the possibilities opened up by the use of negative muons in investigations in different fields of physics and chemistry. We have here discussed the experimental possibilities whose realization requires virtually no modifications in the standard experimental method. The undoubted possibilities are much greater, and they will come to the fore as the new method of investigation is developed.

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