Polarization precession and relaxation of positive muons in polycrystals and single crystals with diamond or zincblende structure

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A complete analysis is presented of the behavior of muon polarization in anomalous muonium in polycrystals with diamond and zincblende structure. The specific depolarization mechanism in polycrystals, connected with the dependence of the precession frequency in O-Mu on the symmetry-axis orientation relative to the magnetic field and with the corresponding dephasing of the precession frequencies is considered. It is shown that in both weak and strong magnetic fields this depolarization obeys a power law. It is shown that an experiment with polycrystals can determine all the hyperfine-structure parameters of anomalous muonium. The experimentally observed picture is in a certain sense simpler than in single crystals. Muon depolarization due to spin-exchange scattering of the muonium electron by the electrons of the medium is considered. It is shown that the spin-exchange relaxation in T-Mu is determined by a single constant in n- and p-type semiconductors. The relaxation in O-Mu is determined by a single constant in n-type semiconductors and by two constants in p-type. The rate of the "true" depolarization of the muon in O-Mu depends on the orientation of the single crystal in a magnetic field at low as well as at high exchange frequencies.

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INTRODUCTION

In 1972, Crowe's group in Berkeley found that besides the previously known Mu atom another modification of muonium is produced in single-crystal silicon.[1] This new type of Mu atom was named by them "anomalous" muonium. The results were later duplicated and extensively expanded in a number of studies,[2-4] in which detailed studies were made of the hyperfine structure of anomalous muonium in different magnetic fields, and of the dependence of the line intensity of anomalous and normal Mu on the density of the dopants and on the temperature. Anomalous muonium was observed and investigated somewhat later in germanium.[5,6] Indications were found[7] that anomalous muonium exists also in single-crystal quartz. A detailed investigation of the temperature dependence of the polarization of normal and anomalous muonium in n- and p-Si single crystals was investigated in detail in Ref. 4.

To explain the results of Ref. 1 we have developed a theory of the behavior of the polarization of positive muons in single crystals with diamond or zincblende structure.[8-10] It followed from the theory, in particular, that the pattern of the polarization precession should be much more complicated than observed in Ref. 1, and the existence of two types of Mu is determined by the symmetry properties of the crystal rather than by the character and density of the doping impurity. The predictions of the theory were later fully confirmed in Refs. 2–6, and the very same Hamiltonian was proposed in Ref. 2 for the anomalous muonium. In the numerical analysis, the authors have duplicated certain analytic results of Ref. 8.

As shown in Ref. 8, the spin Hamiltonian of normal muonium is isotropic, while that of the anomalous one has axial symmetry about the threefold axis

$$H(\mathbf{n},\mathbf{B})=\hbar\Omega_{\perp}(\sigma_e\sigma_\mu)+\hbar(\Omega_{\parallel}-\Omega_{\perp})(\sigma_e\mathbf{n})(\sigma_\mu\mathbf{n}) -\mu_e\mathbf{B}\sigma_e-\mu_\mu\mathbf{B}\sigma_\mu, \quad (1)$$

where $\Omega_{\parallel}$ and $\Omega_{\perp}$ are the hyperfine-splitting constants, σ_e, σ_μ, μ_e, and μ_μ are the spin operators and the magnetic moments of the muonium electron and of the muon, respectively, n is a unit vector directed along the threefold symmetry axis, and B is the external magnetic field. It follows from experiment[2] that in Si we have $4\Omega_{\perp}=2\pi(92.1\pm0.3)$ MHz and $4\Omega_{\parallel}=2\pi(17.1\pm0.3)$ MHz.

In Ref. 8 the normal muonium was identified with trapping of a muon in a tetrapore of the crystal (T-muonium), while the anomalous muonium was identified with trapping in an octapore[1)] (O-muonium). It was correspondingly predicted that two muonium types can exist in all crystals with diamond or zincblende structure. This interpretation is most logical. The alternate hypothesis, that attributes the anomalous Mu to formation of a paramagnetic chemical complex with the lattice atoms[2,4] (the muonium is shifted away along the threefold axis from the center of the tetrapore towards one of the nearest atoms) is not very convincing and is artificial. In particular, trivial group-theoretical considerations show that the chemical-binding hypothesis excludes trapping of Mu in an octapore.

O- and T-muonium were recently observed in single-crystal diamond,[11] thus confirming the conclusions of Ref. 8. Muonium in diamond is of particular interest. In fact, the equation for the density matrix of muonium in diamond contains practically no relaxation terms that could lead to "true" depolarization of the muon spin. First, diamond is an ideal insulator, so that spin-exchange scattering of the muonium electron by the conduction electrons is excluded. Second, the spins of almost all the nuclei are zero, and there is no dipole-dipole relaxation.[2)]

The hyperfine structure constants $\Omega_{\parallel}$ and $\Omega_{\perp}$ differ by approximately five times in both Si and Ge,[2,5] and can naturally not be attributed to the presence of a quadrupole moment in an isolated Mu atom.[12]

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Crystals with diamond structure have four threefold axes. The observed precession picture is therefore determined by four modifications of O-muonium (in accordance with the relative orientation of n and B) and is quite complicated. At certain single-crystal orientations relative to the field the picture becomes much simpler. The most convenient is the orientation along $\langle 100\rangle$, when the behavior of the polarization in all octapores is equivalent.[9,10]

MUONIUM IN POLYCRYSTALS

2. The theory of the behavior of the muon polarization in single crystals with diamond or zincblende structure must be generalized to include polycrystals, primarily in the interest of the experimenters. The study of many important materials in single-crystal form is difficult. We shall show that experiments with polycrystals can yield practically all the hyperfine-structure information obtainable from experiments with single crystals (the only exception is a precision measurement of the g-factor of the muonium electron). Moreover, the experimentally observed picture of the precession in polycrystals is in general simpler to decode.

For the T-Mu atom with spherically symmetrical Hamiltonian, single crystals with polycrystals are identical. In O-muonium the behavior of the polarization, and in particular of the precession frequency, depends essentially on the mutual orientation of the external magnetic field and the symmetry axis of the pore. Therefore in polycrystals one observes effectively, even in the absence of true depolarization due to relaxation processes, rapid depolarization of the muon spin because of the "dephasing" of the precession frequencies upon averaging over all possible orientations. Anomalous muonium may therefore not be observed at all in polycrystals, and the corresponding part of the polarization will be simply "lost." Let us examine this mechanism.

3. In Refs. 9 and 10 we obtained for the total muon polarization in the O-Mu atom in a single crystal the equation

$$P_i(t)=\tfrac{1}{4}\sum_{\alpha}S_{ik}(\mathbf{n}_\alpha,\mathbf{B},t)P_k(0),$$
$$S_{ik}(\mathbf{n}_\alpha,\mathbf{B},t)=\tfrac{1}{4}\,\mathrm{Sp}\{\sigma_i^\mu\otimes\chi\exp[-i\hbar^{-1}H(\mathbf{n}_\alpha,\mathbf{B})t]\sigma_k^\mu\otimes\chi \times\exp[i\hbar^{-1}H(\mathbf{n}_\alpha,\mathbf{B})t]\}. \tag{2}$$

The summation is over all four octapores that are nonequivalently positioned relative to the magnetic field; $P_k(0)$ is the initial polarization of the muon, σ_i^μ are the spin matrices of the muon, χ is a unit 2×2 matrix, and the symbol $\otimes$ denotes the direct product.

For a nontextured polycrystal it is necessary to average (2) over the equally probable orientations of the crystallites. The averaging for each octapore is then identical and

$$P_i(t)=\langle S_{ik}(\mathbf{n},\mathbf{B},t)\rangle P_k(0)=S_{ik}(\mathbf{B},t)P_k(0). \tag{3}$$

It follows from (2) that the tensor $S_{ik}(\mathbf{n},\mathbf{B},t)$ depends only on the angle θ between n and B. In Ref. 9 we obtained simple formulas for the tensor $S_{ik}(\mathbf{n},\mathbf{B},t)$ in two limiting cases: weak ($\omega\ll(\Omega_\parallel+\Omega_\perp)/2=\Omega$) and strong ($\omega\gg\Omega$) fields. A coordinate system was chosen with $z\|\mathbf{B}$ and with the x axis lying in the plane passing through n and B; then $S_{ik}(\mathbf{n},\mathbf{B},t)\equiv S_{ik}(\theta,B,t)$. We shall use for polycrystals likewise a coordinate system with $z\|\mathbf{B}$.

We write down the components of the averaged tensor

$$S_{ik}(\mathbf{B},t)=\langle R_{il}(\varphi)S_{lm}(\theta,B,t)R_{mk}^{-1}(\varphi)\rangle_{\theta,\varphi}, \tag{4}$$

where $R_{il}(\varphi)$ is the matrix of rotation through an angle φ about the z axis. After averaging we obtain

$$S_{xx}(\mathbf{B},t)=S_{yy}(\mathbf{B},t)=S_\perp(t)=\tfrac{1}{2}[\langle S_{xx}(\theta,B,t)\rangle_\theta+\langle S_{yy}(\theta,B,t)\rangle_\theta],$$
$$S_{zz}(\mathbf{B},t)=S_\parallel(t)=\langle S_{zz}(\theta,B,t)\rangle_\theta,\quad S_{xz}(\mathbf{B},t)=S_{yz}(\mathbf{B},t)=0,$$
$$S_{yx}(\mathbf{B},t)=-S_{xy}(\mathbf{B},t)=\langle S_{yx}(\theta,B,t)\rangle_\theta, \tag{5}$$
$$\langle S_{ik}(\theta,B,t)\rangle_\theta=\tfrac{1}{2}\int_0^\pi S_{ik}(\theta,B,t)\sin\theta\,d\theta.$$

4. Assume that there is no external field. The behavior of the polarization in polycrystals and single crystals is the same and is determined by the formula

$$P(t)=\tfrac{1}{3}P(0)\,[\tfrac{1}{2}(1+\cos 4\Omega_\perp t)+\cos 2(\Omega_\parallel-\Omega_\perp)t+\cos 2(\Omega_\parallel+\Omega_\perp)t]. \tag{6}$$

The damping is not produced by dephasing but is determined only by the true relaxation. In contrast to isotropic muonium, only one-sixth of the polarization is preserved. Therefore, if the trapping of the muon in the different pore types is equally probable, the ratio of the conserved components of the polarization in the octa- and tetrapores is $P^O_{\text{cons}}/P^T_{\text{cons}}=2/3$, although there are twice as many octapores as tetrapores.

In a weak field, a complicated multifrequency precession is observed in single crystals.[9,10] In polycrystals all the terms that oscillate with frequencies that depend on the magnetic field attenuate in power-law fashion at a rate proportional to the frequency ω [$\hbar\omega=|\mu_e|B$, see Eqs. (45)–(47) of Ref. 10.] Even in a field $B\sim 10$ G the amplitude of these terms in $S_\perp(t)$ and the off-diagonal components of the tensor decrease by one order of magnitude within 10^{-6} sec. Actually, therefore, starting with $t>10^{-7}$ sec only polarization oscillation with one frequency

$$S_\parallel(t)=S_\perp(t)=(1+\cos 4\Omega_\perp t)/6, \tag{7}$$

will be observed.

5. In intermediate fields ($\omega\sim\Omega$) all the frequencies of the transitions between the O-Mu hyperfine-structure levels are of the order of Ω; the transition probabilities are high, so that the muons are rapidly depolarized. This case is of little interest. We proceed to the most interesting case, with abundant possibilities, of strong magnetic fields (corresponding to $B>100$ G for Si and Ge).

In single crystals, the tensor components $S_{ik}(\mathbf{n},\mathbf{B},t)$ have terms dependent as well as independent of the time, $S_{ik}(\mathbf{n},\mathbf{B},t)=S^c_{ik}+S^v_{ik}(t)$. As shown in Ref. 9, S^c_{ik} has a unity trace. Using the results of Ref. 9 and of Eq. (5), we obtain

$$S_\perp{}^c=(1-S_\parallel{}^c)/2=S_0(a_-,b_-)+S_0(a_+,b_+). \tag{8}$$

Here

$$S_0(a_\pm, b_\pm)=\frac{(\Omega_\parallel-\Omega_\perp)^2}{2b_\pm}\left[\frac{a_\pm^2}{b_\pm}+\frac{2}{3}-\left(\frac{a_\pm^2}{b_\pm}\right)^{1/2}\left(1+\frac{a_\pm^2}{b_\pm}\right)\operatorname{arctg}\left(\frac{b_\pm}{a_\pm^2}\right)^{1/2}\right], \tag{9}$$

where $a_\pm=\Omega_\perp\pm\zeta\omega$, $b=(\Omega_\parallel-\Omega_\perp)(\Omega_\parallel+\Omega_\perp\pm2\zeta\omega)$, and $\zeta=|\mu_\mu/\mu_e|$. It is seen that in a strong magnetic field $\zeta\omega\gg\Omega_\parallel+\Omega_\perp$ the time-independent part of the transverse polarization components tends to zero, and $S_\parallel^c\to1$.

6. We consider now the time-dependent polarization components, assuming that there is not true damping. The damped component $S_\parallel^v(t)$ of the longitudinal component can be represented in the form

$$S_\parallel^v(t)=1/2[S_\parallel(a_-, b_-, t)+S_\parallel(a_+, b_+, t)], \tag{10}$$

$$S_\parallel(a_\pm, b_\pm, t)=(\Omega_\parallel-\Omega_\perp)^2\int_0^1 x^2(1-x^2)\frac{\cos[2(a_\pm^2+b_\pm x^2)^{1/2}t]}{a_\pm^2+b_\pm x^2}dx. \tag{11}$$

In strong fields, the precessions are large ($\omega_{ik}\sim10^7$ sec^{-1}) and the functions $S(a_\pm, b_\pm, t)$ reach their asymptotics within times $t\sim10^{-7}$ sec. We obtain an asymptotic estimate of the integral (11) by the stationary-phase method (the critical-point method),[13] according to which the main contribution to an integral of a rapidly oscillating function is made by the vicinity of the points where the phase has extrema. Equation (10) at $t>10^{-7}$ sec then takes the form

$$S_\parallel^v(t)\approx\frac{1}{8}\left(\pi\left|\frac{\Omega_\parallel-\Omega_\perp}{\Omega_\perp-\zeta\omega}\right|\right)^{1/2}\frac{\cos[2|\Omega_\perp-\zeta\omega|t\pm3\pi/4]}{(\Lambda_-t)^{3/2}}$$
$$+\frac{1}{8}\left(\pi\left|\frac{\Omega_\parallel-\Omega_\perp}{\Omega_\perp+\zeta\omega}\right|\right)^{1/2}\frac{\cos[2(\Omega_\perp+\zeta\omega)t\pm3\pi/4]}{(\Lambda_+t)^{3/2}}, \tag{12}$$

where the upper sign corresponds to $b_\pm>0$ and the lower to $b_\pm<0$; the damping rates are

$$\Lambda_\pm=|\Omega_\parallel+\Omega_\perp\pm2\zeta\omega|. \tag{13}$$

7. The stationary-phase method cannot be used for $S_\parallel(a_-, b_-, t)$ at two values of the external field, namely $\zeta\omega=(\Omega_\parallel+\Omega_\perp)/2$ (then $\Lambda_-=0$) and $\zeta\omega=\Omega_\perp$ (singularity in the amplitude).

The case of particular interest is $\zeta\omega=(\Omega_\parallel+\Omega_\perp)/2$. In such a field $a_-=(\Omega_\perp-\Omega_\parallel)/2$, $b_-=0$ and $S_\parallel(\Omega_\parallel-\Omega_\perp)/2, 0, t)=(8/15)\cos(\Omega_\perp-\Omega_\parallel)t$. The frequency ω_{21} is then independent of the angle between B and C_3 for all "types" of O-Mu, and only the true depolarization will be observed.[9,10]

The function $S_\parallel(a_+, b_+, t)$ attenuates as before practically to zero within $t\sim10^{-7}$ sec. Therefore at $t>10^{-7}$ sec there will be observed an oscillation of the longitudinal polarization with one frequency:

$$P_z(t)=[S_\parallel^c+(4/15)\cos(\Omega_\perp-\Omega_\parallel)t]P_z(0), \tag{14}$$

where the time-independent contribution to the polarization $S_\parallel^c$ is determined from Eq. (8).

As shown by experiments, the necessary external fields are of the order of 1, 1.5, and 2 kG for Si, Ge, and diamond, respectively. A simple estimate shows that at a field deviation $\delta B\sim10$ G, Eq. (14) operates well up to an observation time $t\sim10^{-5}$ sec. An interesting effect is thus observed near the point $\zeta\omega=(\Omega_\parallel+\Omega_\perp)/2$: the longitudinal component of the polarization acquires an oscillating component that attenuates very slowly with time.

In a field $\zeta\omega=\Omega_\perp$ the integral (11) can be calculated exactly and decreases asymptotically like t^{-2}.

8. We restrict the analysis of the damped component $S_\perp^v(t)$ of the transverse polarization component likewise to an asymptotic behavior determined by the equation

$$S_\perp^v(t)\approx\frac{1}{4}\left(\pi\left|\frac{\Omega_\perp-\zeta\omega}{\Omega_\perp-\Omega_\parallel}\right|\right)^{1/2}\frac{\cos[2|\Omega_\perp-\zeta\omega|t\pm\pi/4]}{(\Lambda_-t)^{1/2}}$$
$$+\frac{1}{4}\left(\pi\frac{\Omega_\perp+\zeta\omega}{|\Omega_\perp-\Omega_\parallel|}\right)^{1/2}\frac{\cos[2(\Omega_\perp+\zeta\omega)t\pm\pi/4]}{(\Lambda_+t)^{1/2}}. \tag{15}$$

The component $S_{yx}(t)$ is defined in similar fashion [see Eq. (81) of Ref. 10], has the same order of damping $O(t^{-1/2})$, and the same depolarization rates $\Lambda_\pm$(13). It is seen that the precessing part of the transverse component of the polarization attenuates more slowly than the oscillating part of the longitudinal polarization.

An undamped precession with frequency close to $|\Omega_\perp-\Omega_\parallel|$ should also be observed in the vicinity of the point $\zeta\omega=(\Omega_\parallel+\Omega_\perp)/2$:

$$S_\perp^v(t)\approx(11/30)\cos(\Omega_\perp-\Omega_\parallel)t, \quad S_{yx}(t)\approx1/6\sin(\Omega_\perp-\Omega_\parallel)t. \tag{16}$$

Equations (16) are subject to the same conditions on the observation time and on the field mismatch δB as Eq. (14).

MUON DEPOLARIZATION IN SINGLE CRYSTAL WITH DIAMOND OR ZINCBLENDE STRUCTURE

9. We consider now the true muon depolarization in single crystals, due to spin-exchange scattering of the muonium electron by the electrons of the medium. We assume that the muonium atom does not diffuse over the lattice. The experimental data now available lead to no concrete conclusions whatever concerning the diffusion. It can only be stated that O-muonium does not diffuse once its characteristic frequencies are observed. Diffusion would cause the precession picture to vanish as a result of transition from the octapore to another one with a different orientation relative to the magnetic field. On the whole, the problem calls for detailed theoretical and experimental research.

The form of the relaxation equation for the spin density matrix of the muonium depends on the symmetry of the exchange-scattering operator. In the general case this operator is determined by six independent functions (see, e.g., Refs. 14 and 15); specifically, it is determined by the symmetry of the wave functions of the electrons of the medium relative to the scattering center.

The ground state of the muonium electron is an orbital singlet, therefore the wave function of the muonium electron is factorized in the spin and coordinate variables. Usually the conduction band in semiconductors with diamond or zincblende structure is not degenerate, and the spin and coordinate variables are separated in the conduction electron wave function. It is known (see, e.g., Refs. 14 and 15) that in this case the exchange-scattering spin operator is isotropic. The relaxation term depends then on one constant ν, namely "the frequency of the spin-exchange scattering."

The valence band is usually degenerate, and for the electrons of the valence band the spin and coordinate variables of the wave function do not separate. Thus, the spin operator of the exchange scattering of the muon electron by the valence-band electrons is not necessarily isotropic. T-Mu is located at the center of a tetrapore with tetrahedron symmetry (point group T_d). For this group, only the isotropic exchange-scattering spin operator is isotropic. Accordingly the T-muon relaxation is determined by a single constant independently of the type of semiconductor conductivity.

Located at the center of the octapore is O-Mu. The symmetry group is D_{3d} for a crystal with diamond structure and C_{3v} for a crystal with zincblende structure. Invariant for these groups is the exchange-scattering spin operator, which has axial symmetry relative to a threefold axis. We direct the z axis along a threefold axis, and then the axially symmetric exchange-scattering operator takes in the general case the form

$$\hat{A}=A_1(I+\sigma_1^z\otimes\sigma_2^z)+A_2(\sigma_1^z\otimes\chi+\chi\otimes\sigma_2^z)+A_3(\sigma_1^-\otimes\sigma_2^++\sigma_1^+\otimes\sigma_2^-), \qquad (17)$$

where $\sigma_1^{z,\pm}$ and $\sigma_2^{z,\pm}$ are the spin operators of the muonium electrons and of the medium electrons, respectively, I is a unit 4×4 matrix, and A_i are the exchange integrals. From the invariance of the operator (17) to reflection in a plane passing through a threefold axis (the z axis) we obtain $A_2=0$.

The relaxation equation can be obtained by using the scattering-matrix formalism. For isotropic scattering, such an equation was obtained in Ref. 16. In our case an axially symmetric exchange-scattering operator corresponds to an axially symmetric scattering matrix. The relaxation equation also has axial symmetry relative to a threefold axis of the crystal.

As is customary (see, e.g, Refs. 17 and 18), we introduce the effective spin Hamiltonian corresponding to the exchange-scattering spin operator (17). We can then use the equations obtained in Ref. 19 from the known NMR and ESR equations.[20,21] At high temperatures we can neglect the influence of the polarization of the electrons of the medium on the relaxation term.[19] Then the spin density matrix is determined from the Wangsness-Bloch equation, which has axial symmetry:

$$\dot{\rho}+i\hbar^{-1}[H,\rho]=\nu_\perp(\sigma_e^i\otimes\chi\rho\sigma_e^i\otimes\chi-3\rho)+\Delta\nu(n_in_k\sigma_e^i\otimes\chi\rho\sigma_e^k\otimes\chi-\rho), \qquad (18)$$

where $\Delta\nu=\nu_\parallel-\nu_\perp$, $\nu_\parallel$ and $\nu_\perp$ are the "frequencies" of the spin-exchange scattering, n_i is a unit vector directed along the threefold symmetry axis $\langle111\rangle$, and H is the Hamiltonian of the hyperfine interaction (1). The polarization of the electrons of the medium leads to a renormalization of the magnetic moment of the muonium electron (see, e.g., Refs. 16 and 22). However, since usually the polarization of the electrons of the medium in semiconductors is low, the indicated corrections can be neglected.

We arrive thus at an important qualitative result: the relaxation term in the Wangsness-Bloch equation is isotropic for T-Mu in n- and p-type semiconductors, and for O-Mu it is isotropic in n-semiconductors and has axial symmetry in p-semiconductors.

The relaxation equation for T-Mu with isotropic spin Hamiltonian is isotropic ($\nu_\perp=\nu_\parallel=\nu$) and the solution is well known.[23,24] We consider now the depolarization of a muon in O-Mu with the anisotropic spin-Hamiltonian (1). We confine ourselves only to the region of strong fields ($\omega\gg\Omega$), when the precession picture is simplest. In weak fields, the behavior of the polarization is complicated enough even without allowance for relaxation. We obtain next the solution of the anisotropic relaxation Eq. (18) for low ($\nu\ll\Omega$) and high ($\nu\gg\Omega$) exchange frequencies.

10. We consider first the case $\nu\ll\Omega$, when precession with muonium frequency is observed. The Wagsness-Bloch Eq. (18) with the Hamiltonian (1) is equivalent to a system of 15 coupled equations for the density-matrix components. In the considered region (damping rate Λ much less than all the observed precession frequencies $|\omega_{ik}|$), however, in a basis that diagonalizes the hyperfine-interaction Hamiltonian (1), this system breaks up into a system of three equations for the diagonal components of the density matrix, into a system of four equations for the components oscillating with observable frequencies, and a system of equation for the components having unobservably high frequencies. The oscillating and nonoscillating components of the tensor S_{ik} are determined respectively by the off-diagonal and diagonal components of the density matrix. The damping of the oscillating and nonoscillating components of the tensor S_{ik} is thus determined from equation systems that are not interrelated.

In order not to encumber the text with the straightforward but unwieldy calculations, we present only the final results. The damping of the nonoscillating components of the tensor $S_{ik}(\theta,B,t)$ is determined by two rates:

$$\lambda_\pm=\nu_\perp\{2+(\gamma_1+\gamma_2)/2\pm[\alpha^2+(\gamma_1-\gamma_2)^2/4]^{1/2}\}. \qquad (19)$$

Here

$$\gamma_{1,2}=(\bar\Omega_\perp+\Delta\bar\Omega_2)^2(1+\eta\sin^2\theta)\,(1+\cos\vartheta_{3,4})+\bar\Omega_\perp^2(2+\eta\sin^2\theta)+\eta\sin^2\theta,$$

$$\alpha=2(\bar\Omega_\perp+\Delta\bar\Omega_2)^2(1+\eta\sin^2\theta)\cos2\vartheta_3\cos2\vartheta_4-(1-\bar\Omega_\perp^2)\delta(2+\eta\sin^2\theta)$$
$$+\eta[\Delta\bar\Omega_2(2\bar\Omega_\perp+\Delta\bar\Omega_2)\,(1-2\delta\cos^2\theta)-\bar\Omega_\perp\sin2(\vartheta_3-\vartheta_4)\,\sin2\theta],$$
$$\delta=\cos2(\vartheta_3-\vartheta_4), \qquad (20)$$

where $\Omega_\perp=\Omega_\perp/\omega$, $\bar\Omega_\parallel=\Omega_\parallel/\omega$, $\bar\Omega=\bar\Omega_\parallel\cos^2\theta+\bar\Omega_\perp\sin^2\theta$, $\Delta\bar\Omega_1=(\bar\Omega_\parallel-\bar\Omega_\perp)\sin\theta\cos\theta$, $\Delta\bar\Omega_2=(\bar\Omega_\parallel-\bar\Omega_\perp)\sin^2\theta$, and $\eta=\Delta\nu/\nu$. The parameters ϑ_3 and ϑ_4 are determined by a transformation that permits approximate diagonalization of the Hamiltonian (1) in strong fields[9]:

$$\operatorname{tg}2\vartheta_{3,4}=\Delta\bar\Omega_1(\bar\Omega\mp\zeta)^{-1}. \qquad (21)$$

The solution (19) was obtained accurate to terms $\bar\Omega^2$. This accuracy is necessary in the case when $\theta\approx0$ or $\pi/2$. Indeed, in that case we have $\delta\approx1$ and $\alpha\approx-2$, and $\lambda_-=0$ accurate to $\bar\Omega^2$ terms. At $\theta=0$ and $\pi/2$ the expression for the depolarization rates becomes much simpler.

Let $\theta=0$, then $\gamma_1=\gamma_2=2\Omega_1^2(2+\eta)$, $\alpha=-2[1-\Omega_1^2(2+\eta)]$, and we obtain for the damping rates

$$\lambda_+=4\nu_\perp, \quad \lambda_-=\tau_1^{-1}=2\bar\Omega_\perp^2(\nu_\parallel+\nu_\perp). \qquad (22)$$

It turns out here that damping of the nonoscillating components will be observed only at the low rate τ_1^{-1}.

Let $\theta=\pi/2$, then $\gamma_1=\gamma_2=2(\Omega_\parallel^2+\Omega_\perp^2)+(1+\Omega_\parallel^2)\eta$ and $\alpha=-\eta(1-\Omega_\parallel^2)-2(1-\Omega_\parallel^2-\Omega_\perp^2)$. In this case the nonoscillating components attenuate likewise only at the slow rate

$$\tau_1^{-1}=\lambda_-=2\bar{\Omega}_\parallel^2(\nu_\parallel+\nu_\perp)+4\bar{\Omega}_\perp^2\nu_\perp. \tag{23}$$

We consider now the damping rates at the angles θ encountered in the three single-crystal orientations that are simplest for experimental observation: B$\|\langle 100\rangle$, B$\|\langle 111\rangle$, and B$\|\langle 110\rangle$. In the first case one observes one O-Mu for which $\cos\theta=1/\sqrt{3}$. In the second case two O-Mu are observed, with $\theta=0$ for one and $\cos\theta=1/3$ for the other. In the third case, too, two O-Mu are observed, with $\theta=\pi/1$ for one and $\cos\theta=(2/3)^{1/2}$ for the other. Thus, if $\theta\neq 0,\pi/2$, we have $\delta<1$ and $1-\delta\sim 1$ for all O-muonium orientations, so that there is no need to take the quadratic terms $\bar{\Omega}^2$ into account, and we obtain $\gamma_1=\gamma_2=\eta\sin^2\theta$ and $\alpha=-\delta(2+\eta\sin^2\theta)$. Within the limits of the indicated accuracy, the nonoscillating terms of the components of the tensor S_{ik} will again attenuate at only one rate

$$\tau_1^{-1}=\nu_\perp(1-\delta)(2+\eta\sin^2\theta). \tag{24}$$

We see that the damping rate is no longer small but is of the order of the exchange frequency ν.

Solution of the system of equations for the off-diagonal components of the density matrix, which oscillate with observable frequencies (ω_{21} and ω_{43}), leads to a simple expression for the damping rate of the oscillating components of the tensor S_{ik}:

$$\tau_2^{-1}=2(\nu_\parallel\cos^2\theta+\nu_\perp\sin^2\theta). \tag{25}$$

The components of the tensor S_{ik}, which determine the muon polarization in O-Mu in strong fields in the indicated single-crystal directions that are simplest from the experimental point of view, take thus, with allowance for the relaxation at low exchange frequency, the form

$$\begin{aligned}
S_{zz}&=2\left[\left(\frac{\Omega-\zeta\omega}{\omega_{21}}\right)^2+\left(\frac{\Omega+\zeta\omega}{\omega_{43}}\right)^2\right]e^{-t/\tau_1}\\
&+2\left[\left(\frac{\Delta\Omega_1}{\omega_{21}}\right)^2\cos\omega_{21}t+\left(\frac{\Delta\Omega_1}{\omega_{43}}\right)^2\cos\omega_{43}t\right]e^{-t/\tau_2},\\
S_{xx}&=2\left[\left(\frac{\Delta\Omega_1}{\omega_{21}}\right)^2+\left(\frac{\Delta\Omega_1}{\omega_{43}}\right)^2\right]e^{-t/\tau_1}\\
&+2\left[\left(\frac{\Omega-\zeta\omega}{\omega_{21}}\right)^2\cos\omega_{21}t+\left(\frac{\Omega+\zeta\omega}{\omega_{43}}\right)^2\cos\omega_{43}t\right]e^{-t/\tau_2},\\
S_{yy}&=1/2(\cos\omega_{21}t+\cos\omega_{43}t)e^{-t/\tau_2},\\
S_{yx}&=\left(\frac{\Omega-\zeta\omega}{\omega_{21}}\sin\omega_{21}t+\frac{\Omega+\zeta\omega}{\omega_{43}}\sin\omega_{43}t\right)e^{-t/\tau_2},\\
\omega_{21}&=2[(\Delta\Omega_1)^2+(\Omega-\zeta\omega)^2]^{1/2},\quad \omega_{43}=2[(\Delta\Omega_1)^2+(\Omega+\zeta\omega)^2]^{1/2}.
\end{aligned} \tag{26}$$

The damping rate τ_1^{-1} is determined by Eqs. (22)–(24), and τ_2^{-1} by Eq. (25).

The total polarization of the muon is O-Mu in the three simplest single-crystal orientations is determined by the same combination (26) of the components of the tensor S_{ik} as in the absence of relaxation [see Eqs. (36)–(41) of Ref. 9]. We see that the simplest way to determine the exchange-scattering constants $\nu_\parallel$ and $\nu_\perp$ is to measure the damping rate τ_2^{-1} of the precessing components of the muon polarization in a strong external magnetic field at various single-crystal orientations.

We point out an interesting possibility of determining the g-factor of the muonium electron in a strong magnetic field at low exchange frequencies. The muon depolarization rate in O-Mu in an octapore whose symmetry axis is parallel or perpendicular to the magnetic field is proportional to ω^{-2} [Eqs. (22) and (23)]. However, in view of the low rate of depolarization one cannot expect good accuracy.

11. At high exchange frequencies, no precession with muonium frequency will be observed. Just as for low exchange frequencies, the Wangsness-Bloch Eq. (18) leads to a system of 15 coupled equations for all the muonium density-matrix components. This system can be solved by perturbation theory with the small parameter Ω/ν or $\Delta\nu/\omega$. In the zeroth approximation, the hyperfine structure is inessential and the solution is obtained directly. Leaving out the cumbersome calculations, we present the final results.

The damping rate of the longitudinal (parallel to the external magnetic field) polarization is

$$\tau_1^{-1}=4\nu_\perp\frac{[4+\eta(1+\cos^2\theta)][2\bar{\Omega}_\perp(\bar{\Omega}_\perp+\Delta\bar{\Omega}_2)+(\Delta\bar{\Omega}_2)^2]}{\bar{\nu}_\perp^2[4+\eta(1+\cos^2\theta)]^2+4}+\frac{2(\Delta\Omega_1)^2}{\nu_\perp(2+\eta\sin^2\theta)}. \tag{27}$$

Here $\bar{\nu}_\perp=\nu_\perp/\omega$. If $\eta=0$ (n-type semiconductor), the expression simplifies greatly ($\bar{\nu}_\parallel=\bar{\nu}_\perp=\bar{\nu}$):

$$\tau_1^{-1}=4\nu\frac{[2\bar{\Omega}_\perp(\bar{\Omega}_\perp+\Delta\bar{\Omega}_2)+(\Delta\bar{\Omega}_2)^2]}{4\bar{\nu}^2+1}+\frac{(\Delta\Omega_1)^2}{\nu}. \tag{28}$$

The longitudinal polarization attenuates slowly with time, without oscillations:

$$P_z(\theta,t)=P_z(0)e^{-t/\tau_1}. \tag{29}$$

We emphasize that even if the electron relaxation is isotropic, the rate of depolarization of the longitudinal polarization component of the muon in O-Mu depends on the orientation of the single crystal in the magnetic field.

The transverse (perpendicular to the magnetic field) polarization precesses at the muon frequency with a "renormalized" magnetic moment and attenuates:

$$P_+(\theta,t)=P_x+iP_y=P_+(0)\exp(i2\tilde{\zeta}\omega-\tau_2^{-1})t, \tag{30}$$

$$\tilde{\zeta}=\zeta+4[\bar{\Omega}_\perp(\bar{\Omega}_\perp+\Delta\bar{\Omega}_2)+\Delta\bar{\Omega}_1^2]\{\bar{\nu}_\perp^2[4+\eta(1+\cos^2\theta)]^2+4\}^{-1}, \tag{31}$$

$$\tau_2^{-1}=2\nu_\perp\frac{[4+\eta(1+\cos^2\theta)][2\bar{\Omega}_\perp(\bar{\Omega}_\perp+\Delta\bar{\Omega}_2)+(\Delta\bar{\Omega}_2)^2]}{\bar{\nu}_\perp^2[4+\eta(1+\cos^2\theta)]^2+4}+\frac{2\Omega^2+(\Delta\Omega_1)^2}{\nu_\perp(2+\eta\sin^2\theta)}. \tag{32}$$

We note that just as for the longitudinal polarization, even at $\eta=0$ the damping rate of the transverse component of the muon polarization in O-Mu depends strongly on the orientation of the single crystal in a magnetic field:

$$\tau_2^{-1}=2\nu[2(\bar{\Omega}_\perp+\Delta\bar{\Omega}_2)\bar{\Omega}_\perp+(\Delta\bar{\Omega}_2)^2]/(4\bar{\nu}^2+1)+[2\Omega^2+(\Delta\Omega_1)^2]/2\nu. \tag{33}$$

By investigating the dependence of the damping rate of the polarization component that precesses with the muon frequency on the orientation of the single crystal in the magnetic field, we can determine whether the muon goes over from an octapore to a tetrapore or back during the lifetime. Indeed, if the muon goes over into a tetra-

pore, then the damping rate of the transverse polarization is isotropic. If O-Mu turns out to be stable, then the damping rate of the transverse polarization (32) depends on the orientation of the single crystal in the magnetic field. We note that the polarization of the medium leads in principle to a shift of the precession frequency.[25] However, the dependence of the depolarization rate on the single-crystal orientation in a magnetic field is undoubtedly more substantial and of greater interest.

The total polarization of the O-muonium muon is equal to the sum of the muon polarizations in all the octapores that are not equivalent relative to the magnetic field. The result is therefore somewhat simpler in form than for low exchange frequencies ν. We note in this connection that the simplest picture for experimental observation is obtained in the noted three cases,[9] when not more than two different polarization rates should be observed, in analogy with the observation of several types of anomalous muonium. In addition $\tilde{\xi}$ depends on the orientation of the single crystal in the magnetic field, and this must also be taken into account in the analysis of the experimental results.

We emphasize that the study of muon polarization is particularly important in the investigation of semiconductors with narrow forbidden bands (InSb, HgTe, and others), and also of semimetals (e.g., grey tin). Indeed, owing to the high frequency ν of the spin-exchange scattering, the muonium frequencies may become unobservable. In this case a study of the damping rate may turn out to be the main method of determining the parameters of the hyperfine structure.

[1)]In another terminology, these interstices are called respectively tetragonal and hexagonal.

[2)]The admixture of the stable isotope C^{13} with $\mu = 0.74\mu_{nuc}$ is only 1.11%.

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