

Analysis of the behavior of the polarization of the μ^+ meson of muonium atoms in diamond-structure crystals

Yu. M. Belousov, V. N. Gorelkin, and V. P. Smilga

Moscow Physicotechnical Institute

(Submitted 13 June 1977)

Zh. Eksp. Teor. Fiz. 74, 629-639 (February 1978)

A complete analysis is carried out of the time dependence of the muon polarization in the Mu atom in a crystal lattice with diamond structure. It is shown that for muonium in an octapore the influence of the crystal field leads to a tensor form of the hyperfine-interaction spin Hamiltonian and to a qualitative change in the picture of the muon polarization precession. For muonium in a tetrapore, the Hamiltonian remains isotropic. The developed theory permits a detailed investigation of the structure of the crystal field as well as the determination of the position of the Mu atoms in lattices with diamond structure and in zinc blende. The results explain data on the "anomalous" and "normal" Mu in single-crystal silicon.

PACS numbers: 36.10.Dr

Interest in the μ^+ -meson method of the study of solids is constantly increasing. Most experimental studies are devoted to the muon spin precession in solids. It has been established experimentally that in most semiconductors the μ^+ mesons forms a muonium atoms, and the most promising method of precisely determining the characteristics of the Mu atoms is observation of two-frequency precession.^[1,2] Two-frequency precession in weak and strong magnetic fields is described by the known formulas^[1-4]

$$P_{\text{obs}}(t) = 1/2 \cos(\omega^2 t / \omega_0) \cos \omega t, \quad (1)$$

$$P_{\text{obs}}(t) = \cos(\omega_0 t / 2) \cos(2\zeta \omega + \omega_0^2 / 8\omega) t, \quad (2)$$

where $\zeta = |\mu_{\mu} / \mu_e|$ is the ratio of the magnetic moments of the muon and electron, $\omega = \mu_e B \hbar^{-1}$ is the Larmor frequency of the electron precession in a magnetic field B , and ω_0 is the hyperfine-splitting frequency (the exact formula is given in^[2,4]). Formulas (1) and (2) were written without allowance for polarization relaxation, although the precession itself can be effectively observed only in the case when the relaxation time is large ($\tau \sim 10^{-6}$ sec). In other words, a fundamental analysis of the precession picture in the Mu atom can be carried out by considering an isolated atom in external fields. In the theoretical analysis it was always assumed that the Mu atom is hydrogenlike. Yet it is obvious that the wave function is distorted in the crystal field. The character of its variation is determined by the symmetry group and by the value of the crystal field at the point where the muonium is located.

It will be shown below that even very small s -state muonium wave-function distortions due to violation of spherical symmetry should lead to qualitative changes in the observable precession picture. We consider for the sake of argument muonium in crystal with diamond structure (silicon, germanium, grey tin, etc.). Crystals with diamond structure attract particular interest because muonium has been investigated in detail in silicon and germanium. In particular, it was established that two types of muonium exist in silicon,^[5] with no satisfactory interpretation of the results. We shall return later to a discussion of the experimental results.

1. Thermalized muonium can either remain in an in-

terstice (interstitial impurity) or be trapped in a lattice site (substitutional impurity). There are two types of pores in the considered crystals—tetrapores and octapores.^[6] In a tetrapore there are four nearest atoms at a distance $0.433a$ from the center of the pore, and in an octapore there are six nearest atoms at a distance $0.415a$. The lattice constants of silicon and germanium are $a = 5.43 \text{ \AA}$ and $a = 5.66 \text{ \AA}$ respectively. In a tetrapore the crystal field has tetrahedral symmetry (point group T_d), and in an octapore it has the point symmetry D_{3d} . A theoretical analysis^[6,7] has shown that for hydrogen (muonium) it is easier to occupy an octapore. It must be borne in mind, however, that the calculations are mode-dependent.

We shall consider the hyperfine splitting of the levels of muonium in an octapore. The representation D_1 of the spherical rotation group, describing the triply degenerate level of the Mu atom, is resolved into irreducible representations of the group D_{3d} as follows:

$$D_1 = A_{2u} + E_u, \quad (3)$$

where A_{2u} and E_u are one-dimensional and two-dimensional representations (the notation is from^[8]). It follows from (3) that the triplet splits into a nondegenerate level and one doubly degenerate level, whence it is clear that the hyperfine interaction is no longer described by a scalar. The possibility in principle of splitting the triplet state of muonium was noted in^[9].

We obtain the spin Hamiltonian of the Mu atom by using the crystal-field theory,^[10-13] in analogy with the method used to calculate the spectra of paramagnetic ions in ESR theory. We choose the z axis to be the C_3 axis, and then the expansion of the crystal-field potential in spherical harmonics

$$V(r) = \sum_{k=0}^{\infty} B_k r^k Y_k^q(\theta, \varphi) \quad (4)$$

contain only harmonics with even values of k , since the field has an inversion center, and $q = 6p$, where p is an integer. The wave function of the Mu atom should be transformed in accordance with the representations of the group D_{3d} , so that only wave functions with even l

and with projections m that are multiples of six are mixed with the s function $|100\rangle$ of the ground state of the muonium atom.

The Hamiltonian of the hyperfine interaction for the Mu atom is best written in the form

$$H_{\text{HFI}} = \left\{ \frac{8\pi}{3} \sigma_e^i \sigma_\mu^j \delta(r) - \sigma_e^i \sigma_\mu^j (3n_p n_q - \delta_{pq}) \right\} \frac{\mu_e \mu_\mu}{r^3}, \quad (5)$$

where σ_e^i and σ_μ^j are the Pauli operators of the Mu electron and of the μ^+ mesons, respectively, and n_p are direction cosines. The first term in the Hamiltonian (5) is a spherically symmetrical contact term that differs from zero only for the s state, while the remaining terms comprise a spherical harmonic of second order. In an axial crystal field (and particularly in a field with symmetry D_{3d}) we obtain, after averaging the Hamiltonian (5) over the orbital wave functions, the spin Hamiltonian of the Mu atom (see, e.g. ^[10,11]):

$$H = 2\hbar\Omega_\perp (\sigma_e^+ \sigma_\mu^- + \sigma_e^- \sigma_\mu^+) + \hbar\Omega_\parallel \sigma_e^z \sigma_\mu^z - \mu_e (\sigma_e B) - \mu_\mu (\sigma_\mu B), \quad (6)$$

where Ω_\perp and Ω_\parallel are the components of the hyperfine-splitting tensor. For convenience we introduce the scalar hyperfine-splitting constant

$$\Omega_\perp = \omega_0/4, \quad \Omega_\parallel = (\omega_0 + \Delta\omega_0)/4. \quad (7)$$

The anisotropy of the spin Hamiltonian (6) is the result of averaging of the second term in the hyperfine-interaction Hamiltonian (5) over the "distorted" wave functions of the ground state.

The experimental data for paramagnetic ions (Mn^+ , Cu^+ , Ag^+) in various types of crystals usually yield $\Delta\omega_0 \sim (10^{-4} - 10^{-2})\omega_0$. It can be assumed that for the Mu atom the effect is of approximately the same order. If the crystal field is weak enough to be able to use perturbation theory, then the correction $\Delta\omega_0$ is linear in the crystal field and is determined by only one parameter B_2^0 and by the polarizability of the Mu atom. The hyperfine-interaction Hamiltonian was averaged for Mn^+ ions in ^[14-16], but the authors considered only a substitutional impurity in a crystal with cubic symmetry, and retained therefore in H_{HFI} the contact term and obtained only the isotropic correction for levels with different n .

2. To determine the hyperfine-interaction levels we choose the C_3 axis to be the quantization axis (the z axis) and choose as the basis the functions

$$\chi_1 = |++\rangle, \quad \chi_2 = |+-\rangle, \quad \chi_3 = |-+\rangle, \quad \chi_4 = |--\rangle. \quad (8)$$

The first sign pertains to the spin of the Mu electron and the second to the spin of the μ^+ meson. The characteristic equation is of the form

$$\det \begin{pmatrix} \Omega_\parallel + \omega_2(1-\zeta) - \lambda & -\zeta\omega_- & \omega_- & 0 \\ -\zeta\omega_+ & -\Omega_\parallel + \omega_2(1+\zeta) - \lambda & 2\Omega_\perp & \omega_- \\ \omega_+ & 2\Omega_\perp & -\Omega_\parallel - \omega_2(1+\zeta) - \lambda & -\zeta\omega_- \\ 0 & \omega_+ & -\zeta\omega_+ & \Omega_\parallel - \omega_2(1-\zeta) - \lambda \end{pmatrix} = 0, \quad (9)$$

where $\hbar\omega_\pm = \mu_e B_x$, $\hbar\omega_\pm = \mu_\mu (B_x \pm iB_y)$.

We present for reference a convenient form of Eq. (9) for the case when $\zeta\omega \ll \Omega = (\Omega_\parallel + \Omega_\perp)/2$:

$$[(\lambda + \Omega_\perp)^2 - 4\Omega^2 - \omega^2][(\lambda - \Omega_\perp)^2 - (\Omega_\parallel - \Omega_\perp)^2 - \omega^2] + 8\Omega(\Omega_\perp - \Omega_\parallel)\omega^2 = 0. \quad (10)$$

Equation (9) can be solved exactly in two cases: 1) the field is parallel to the C_3 axis, i.e., $B_x = B$, $B_y = B_z = 0$, and 2) the field is perpendicular to the C_3 axis, i.e., $B_x = B_y = 0$, $B_z = B$.

In the first case the solutions of (9) are

$$\lambda_{1,4} = \Omega_\parallel \pm \omega(1-\zeta), \quad \lambda_{2,3} = -\Omega_\parallel \pm \Omega_\perp \alpha, \quad (11)$$

where $\alpha = 2[1 + \omega^2(1 + \zeta)^2/4\Omega^2]^{1/2}$. The wave functions are

$$\begin{aligned} \psi_1 &= |++\rangle, \quad \psi_2 = (C_+ |+-\rangle + C_- |-+\rangle)/\sqrt{2}, \\ \psi_3 &= (C_+ |-+\rangle - C_- |+-\rangle)/\sqrt{2}, \quad \psi_4 = |--\rangle. \end{aligned} \quad (12)$$

Here $C_\pm = [1 \pm \omega(1 + \zeta)/\Omega_\perp \alpha]^{1/2}$.

The behavior of the levels is shown schematically in Fig. 1. In contrast to the isotropic case, besides the crossing of the levels 1 and 2 in strong fields $\omega \approx \Omega_\perp/\zeta$, the levels 1 and 2 cross at $\omega \approx 2|\Omega_\perp - \Omega_\parallel|$ if $\Omega_\perp > \Omega_\parallel$ and the levels 2 and 4 cross in the opposite case.

In the second case (field perpendicular to C_3 axis) the solutions of (9) are

$$\lambda_{1,4} = \Omega_\perp \pm |\Delta\Omega|\alpha, \quad \lambda_{2,3} = -\Omega_\perp \pm \Omega_\perp \alpha, \quad (13)$$

where

$$\begin{aligned} \Delta\Omega &= 2(\Omega_\perp - \Omega_\parallel), \quad \alpha_1 = 1/2[1 + 4\omega^2(1-\zeta)^2/\Delta\Omega^2]^{1/2}, \\ \alpha_2 &= 2[1 + \omega^2(1+\zeta)^2/4\Omega^2]^{1/2}. \end{aligned}$$

The dependence of the energy levels on the field is shown schematically in Fig. 2. In this case there is only a crossing of levels 1 and 2 in strong fields.

In the case of a perpendicular field it is convenient to direct the quantization axis along the field (along the x axis); the functions then take the form

$$\begin{aligned} \psi_1 &= (D_+ |++\rangle + D_- |--\rangle)/\sqrt{2}, \quad \psi_2 = (C_+ |+-\rangle + C_- |-+\rangle)/\sqrt{2}, \\ \psi_3 &= (C_+ |-+\rangle - C_- |+-\rangle)/\sqrt{2}, \quad \psi_4 = (D_- |++\rangle - D_+ |--\rangle)/\sqrt{2}, \end{aligned} \quad (14)$$

where

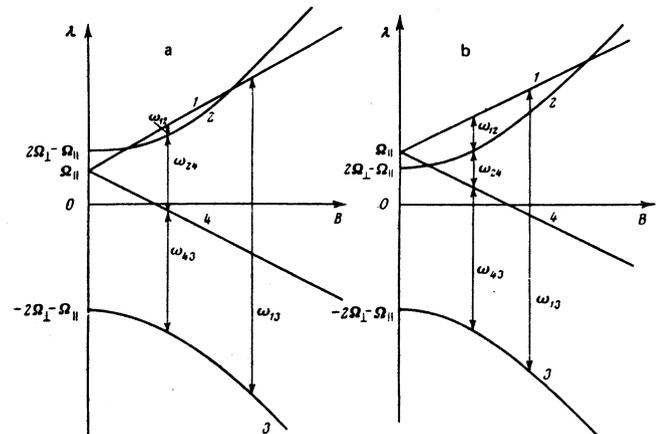


FIG. 1. Energy spin levels in a magnetic field parallel to the symmetry axis: a) $\Omega_\perp > \Omega_\parallel$, b) $\Omega_\perp < \Omega_\parallel$.

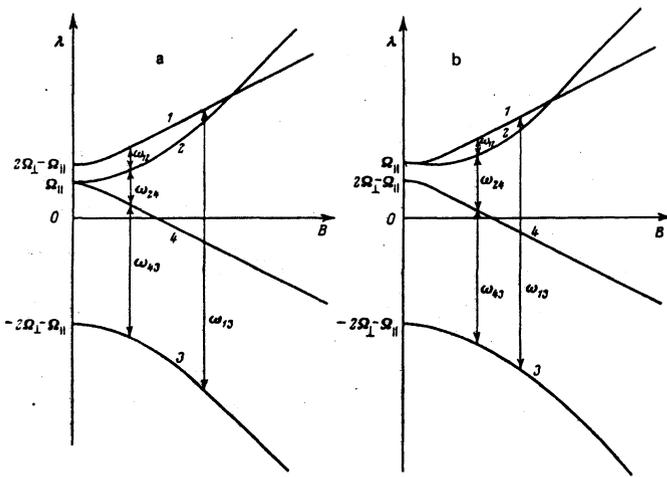


FIG. 2. Energy spin levels in a magnetic field perpendicular to the symmetry axis: a) $\Omega_{\perp} > \Omega_{\parallel}$, b) $\Omega_{\perp} < \Omega_{\parallel}$.

$$C_{\pm}' = [1 \pm \omega(1 + \zeta) / \Omega \alpha_{\pm}]^{1/2}, \quad D_{\pm} = [1 \pm \omega(1 - \zeta) / |\Delta \Omega| \alpha_{\pm}]^{1/2}.$$

3. We proceed to determine the dependence of the μ^+ -meson polarization on the time: $P(t) = S \rho(\sigma_{\mu} \rho)$. The spin density matrix ρ in the considered "pure" case is a known bilinear combination of spin functions. In contrast to the "isotropic" muonium, the problem is now determined by three vectors: the magnetic field, the symmetry axis, and the initial-polarization vector. Accordingly the behavior of the muonium in longitudinal and transverse fields reduce to five variants:

variants I, $B \parallel C_3$:

a) $P(0) \parallel C_3$, b) $P(0) \perp C_3$;

variants II, $B \perp C_3$:

a) $P(0) \parallel B$, b) $P(0) \perp C_3$ и $P(0) \perp B$, c) $P(0) \parallel C_3$.

It is therefore obvious that there are now more opportunities of investigating the precession pictures than in the case of "isotropic" muonium.

Variant Ia (longitudinal field): at the initial instant of time $P(0) = P_z(0)$. We obtain for $P(t)$

$$P_x(t) = \frac{1}{2} P(0) \frac{4\Omega_{\perp}^2}{4\Omega_{\perp}^2 + \omega^2(1 + \zeta)^2} \left[1 + \frac{\omega^2(1 + \zeta)^2}{2\Omega_{\perp}^2} + \cos 2\Omega_{\perp} t \right], \quad (15)$$

$$P_z(t) = P_y(t) = 0.$$

In the observation, the rapidly oscillating cosine ($\Omega_{\perp} \alpha \sim \omega_0$) averages out, and we obtain ultimately the known^[1] formula in which ω_0 is replaced by $4\Omega_{\perp}$:

$$P_{x, \text{obs}}(t) = P(0) \frac{2\Omega_{\perp}^2 + \omega^2(1 + \zeta)^2}{4\Omega_{\perp}^2 + \omega^2(1 + \zeta)^2}. \quad (16)$$

In this case the behavior of the polarization does not differ from the behavior of "isotropic" muonium in a longitudinal field. Experiments in longitudinal fields can determine only one hyperfine splitting constant Ω_{\perp} .

Variant Ib (transverse field): the field is directed along the symmetry axis z , and the initial polarization along the x axis. The polarization $P(t)$ precesses in the xy plane. The formula for $P(t)$ is outwardly similar to

the results obtained in [2]:

$$P_x(t) = \frac{1}{4} P(0) \left[\cos \omega_{12} t + \cos \omega_{23} t + \cos \omega_{45} t + \cos \omega_{13} t \right. \\ \left. + \frac{\omega(1 + \zeta)}{\Omega_{\perp} \alpha} (\cos \omega_{12} t + \cos \omega_{23} t - \cos \omega_{45} t - \cos \omega_{13} t) \right], \quad (17)$$

$$P_z(t) = 0.$$

The frequencies correspond to transitions between the energy levels shown in Fig. 1. The splitting of the triplet state leads to new qualitative effects for $P(t)$ in weak fields. It is seen that at $\omega \approx |\Delta \Omega|$ ($|\Delta \Omega| \ll \Omega$) precession will be observed with one frequency $\omega_{\pi \rho} = 2|\Delta \Omega|$. In weak fields $\omega \ll \Omega$ the observed polarization takes the form

$$P_{x, \text{obs}}(t) = \frac{1}{2} P(0) \cos \left(\Delta \Omega + \frac{\omega^2}{4\Omega_{\perp}} \right) t \cos \omega t. \quad (18)$$

It follows from (18) that if $\Delta \Omega > 0$ then we should again observe at the point $\omega = 2(\Omega_{\perp} |\Delta \Omega|)^{1/2}$ only one precession frequency. At the present-day resolution of the apparatus, experiments makes it possible to determine the sign of the splitting $\Delta \Omega$ if $(\Omega_{\perp} |\Delta \Omega|)^{1/2} \lesssim 10^8 \text{ sec}^{-1}$. It is seen that so long as the splitting turns out to be in the interval $10^8 > |\Delta \Omega| > 10^6$, oscillations of the polarization will be observed even in the absence of an external field, with a frequency $|\Delta \Omega|$ and corresponding to transitions between the split states of the triplet. Thus, compared with "isotropic" muonium, a fundamentally new measurement possibility arises. It should be recalled, however, that for a real observation of the oscillations the following obvious condition must be satisfied: the polarization relaxation time must exceed the period of the oscillations. As shown by experiment, in well prepared insulator crystals (quartz) the relaxation time reaches several microseconds. In the better semiconductor samples investigated to date the relaxation time reaches values 3×10^{-7} to 5×10^{-7} sec at $T = 77$ K.

In strong fields we have for the observable polarization

$$P_{x, \text{obs}}(t) = P(0) \cos 2\Omega_{\perp} t \cos 2 \left(\zeta \omega + \frac{\Omega_{\perp}^2}{\omega} \right) t, \quad (19)$$

is full analogy with formula (2) for two-frequency pre-

cession in the "isotropic" case, but now the high and low frequencies are different by different hyperfine-interaction constants (longitudinal and transverse, respectively).

Variant IIa (longitudinal field): the initial polarization and the field are perpendicular to the symmetry axis and are directed along the x axis; then

$$P_x(t) = \frac{1}{2} P(0) \left\{ \frac{1}{4\alpha_1^2} \left[\frac{4\omega^2(1-\zeta)^2}{\Delta\Omega^2} + \cos \Delta\Omega\alpha_1 t \right] + \frac{4}{\alpha_2^2} \left[\frac{\omega^2(1+\zeta)^2}{4\Omega^2} + \cos 2\Omega\alpha_2 t \right] \right\}, \quad (20)$$

$$P_z(t) = P_y(t) = 0.$$

In weak fields, in contrast to "isotropic" muonium, oscillations of the polarization should be observed. The polarization observable in fields $\omega \gg |\Delta\Omega|$ does not depend on the time and is determined by Eq. (16) if the frequency Ω_1 is replaced by Ω . Experiments in longitudinal fields (variants Ia and IIa) make it possible to determine both hyperfine splitting constants, and hence also the sign of $\Delta\Omega$.

Variant IIb (transverse field): the initial polarization is $P(0) = P_y(0)$ and the field is perpendicular to the symmetry axis. The polarization is determined by the formulas

$$P_y(t) = \frac{1}{4} P(0) \left[\cos \omega_{12}t + \cos \omega_{13}t + \cos \omega_{21}t + \cos \omega_{31}t + \frac{1}{\alpha_1\alpha_2} \left(1 + \frac{\omega^2(1-\zeta^2)}{\Omega|\Delta\Omega|} \right) (\cos \omega_{12}t - \cos \omega_{13}t - \cos \omega_{21}t + \cos \omega_{31}t) \right], \quad (21)$$

$$P_x(t) = \frac{1}{4} P(0) \left[\frac{\omega(1-\zeta)}{|\Delta\Omega|\alpha_1} (\sin \omega_{12}t + \sin \omega_{13}t + \sin \omega_{21}t + \sin \omega_{31}t) + \frac{\omega(1+\zeta)}{\Omega\alpha_2} (\sin \omega_{12}t - \sin \omega_{13}t - \sin \omega_{21}t + \sin \omega_{31}t) \right], \quad (22)$$

$$P_z(t) = 0.$$

The frequencies correspond to transitions between the energy levels determined by formulas (13) (see Fig. 2). As before, the main deviations from the "isotropic" case should be observed in weak fields.

For very weak fields $\omega \ll |\Delta\Omega|$, two-frequency precession should be observed

$$P_{y \text{ obs}}(t) = \frac{1}{2} P(0) \cos \frac{1}{2} \Delta\Omega t \cos \left(\frac{1}{2} \Delta\Omega + \frac{\omega^2}{\Delta\Omega} \right) t. \quad (23)$$

It is seen that the behavior of the polarization differs substantially both from that of "isotropic" muonium [formula (1)] and from the case when the external field is parallel to the symmetry axis [formula (18)], since one frequency is now independent of the field.

If $\omega \geq |\Delta\Omega|$, $\omega \ll \Omega$, the polarization is determined by the formula

$$P_{y \text{ obs}}(t) = \frac{1}{2} P(0) \left[\cos \left(\Delta\Omega - \frac{\omega^2}{4\Omega} \right) t \cos \Delta\Omega\alpha_1 t - \frac{1}{2\alpha_1} \sin \left(\Delta\Omega - \frac{\omega^2}{4\Omega} \right) t \sin |\Delta\Omega|\alpha_1 t \right]. \quad (24)$$

For fields $\omega \gg |\Delta\Omega|$ the formula for two-frequency precession is of the form

$$P_{y \text{ obs}}(t) = \frac{1}{2} P(0) \cos \left(\Delta\Omega - \frac{\omega^2}{4\Omega} \right) t \cos \left(\omega + \frac{\Delta\Omega^2}{\omega} \right) t. \quad (25)$$

It is seen that at the point $\omega = 2(\Omega|\Delta\Omega|)^{1/2}$ there will be observed one precession frequency if $\Delta\Omega > 0$ (one-frequency precession should be observed at $\Delta\Omega < 0$ in the case B || C_3). Thus, experiments in transverse fields (variants Ib and IIb) make it possible to determine from the one-frequency precession not only the sign of $\Delta\Omega$ but also both hyperfine splitting constants.

For strong fields we have

$$P_{y \text{ obs}}(t) = P(0) \cos 2\Omega_1 t \cos 2 \left(\zeta\omega + \frac{\Omega^2}{\omega} \right) t \quad (26)$$

and $P(t)$ is determined by the two hyperfine-interaction constants.

Variant IIc (transverse field): $P(0)$ is directed along the symmetry axis and the field along the x axis. The time dependence of the polarization is given by

$$P_x(t) = \frac{1}{4} P(0) \left[\cos \omega_{12}t + \cos \omega_{31}t + \cos \omega_{13}t + \cos \omega_{21}t - \frac{1}{\alpha_1\alpha_2} \left(1 - \frac{\omega^2(1-\zeta^2)}{\Omega|\Delta\Omega|} \right) (\cos \omega_{12}t + \cos \omega_{31}t - \cos \omega_{13}t - \cos \omega_{21}t) \right], \quad (27)$$

$$P_z(t) = 0.$$

The frequencies are determined by the energy levels from (13), and $P_y(t)$ is determined by (22). In weak fields $\omega \ll |\Delta\Omega|$ one should observe precession with one frequency that is quadratic in the field:

$$P_{z \text{ obs}}(t) = \frac{1}{2} P(0) \cos \left[\frac{1}{2} \left(|\Delta\Omega| - \Delta\Omega - \frac{2\omega^2}{\Delta\Omega} \right) t \right]. \quad (28)$$

For fields $\omega \geq |\Delta\Omega|$ the observed polarization takes the form

$$P_{z \text{ obs}}(t) = \frac{1}{2} P(0) \left[\cos \left(\Delta\Omega - \frac{\omega^2}{4\Omega} \right) t \cos \Delta\Omega\alpha_1 t + \frac{1}{2\alpha_1} \sin \left(\Delta\Omega - \frac{\omega^2}{4\Omega} \right) t \sin |\Delta\Omega|\alpha_1 t \right]. \quad (29)$$

The polarization is determined by formula (25) at $\omega \gg |\Delta\Omega|$, and by formula (26) for strong fields $\omega \gg \Omega$.

From the analysis of the time dependence of the polarization it is seen that the precession frequencies are shifted by an amount equal to the splitting $\Delta\Omega$, and if the splitting is large ($|\Delta\Omega| \geq 10^8 \text{ sec}^{-1}$) polarization at the muonium frequency may not be observed in weak fields. For a complete analysis it is therefore necessary to carry out the experiments in both weak and strong fields.

We have not considered cases of arbitrary mutual orientations of the vectors $P(0)$, C_3 , and B . It is obvious that in principle it is easy to analyze all possible variants. The calculations are simple and merely require some definite amount of work. We emphasize in conclusion that the entire philosophy of the theory is based only on the use of the group properties of the crystal field at the place where the muonium is situated.

4. We shall show that the results offer a most natural explanation of the data on the behavior of muons in silicon and germanium. Observation^[5] of muonium in single-crystal silicon at $T=77$ K has revealed the existence of two types of muonium, "normal" with hyperfine interaction frequency $\omega_0^n=0.45\omega_{ov}$ ($\omega_{ov}=2.8\cdot 10^{10}$ sec⁻¹) and "anomalous" with very low hyperfine-interaction frequency $\omega_0^a=0.02\omega_{ov}$. The experiment was performed in pure *p*-type crystal and the muonium-signal relaxation time was 4×10^{-7} sec, so that two-frequency precession could be distinctly observed for both types of muonium. This reference contains data for the Fourier transform of $P(t)$, where the signals of the two-frequency precession of the normal and anomalous muonium are clearly separated. The dopant concentrations in the samples were 10^{12} , 5×10^{12} , and 5×10^{13} cm⁻³. Anomalous muonium was observed in all samples, and the maximum signal amplitude was observed in the sample with impurity density 5×10^{12} cm⁻³. In this sample, the signal from the anomalous muonium exceeded substantially the signal from the normal one. The dependence of the precession frequency of the anomalous muonium on the external field (in the interval up to 1 kG) was in general in agreement with formula (2), which describes two-frequency precession in strong fields. This has shown directly that the frequency of the hyperfine splitting of the anomalous muonium is much lower than the vacuum frequency. No change in the precession frequencies with changing orientation of the crystal relative to the external magnetic field was noted for normal muonium, but a change was distinctly observed for the anomalous muonium.

The reduction of the data with the aid of formula (2) yielded the following results: The hyperfine-splitting frequency of the anomalous muonium with the field oriented along the [111] axis is $\omega_0^a=0.0198\omega_{ov}$ and $\omega_0^a=0.0205\omega_{ov}$ for orientation along [100]. The experiment could be interpreted, however, only by assuming that the *g* factor of the electron in the muonium does not depend on the crystal orientation in the field and is equal to 13 ± 3 . Obviously, when the ground state is not degenerate with respect to orbital motion (orbital signal), this assumption is absolutely unsatisfactory. In fact, data on double electron spin resonance for donor centers in semiconductors show that usually the changes of the *g* factor appear only in the fourth or fifth significant figure.^[11,17] For example, for lithium in silicon the principal values of the tensor *g* and $g_{11}=1.9978$ and $g_{\perp}=1.9992$. We note here that the wave function of the lithium electron is strongly smeared ($E_i=0.033$ eV) so that the effect of the crystal field on the spin *g* factor manifests itself fully. The authors of^[5] have noted that there is actually no reasonable description of the anomalous muonium whatever, but in the subsequent discussions in the literature the situation with the *g* factor was simply ignored. Thus, the "usual" precession theory was unable to offer any satisfactory explanation of the data on anomalous muonium.

Since we are not in possession of the primary experimental material we are unable to perform an exact quantitative analysis, but qualitatively the data of both

[5] and [18-20] find a rather simple and natural explanation if the anomalous muonium is identified with trapping of the muonium in an octapore, and normal muonium with trapping in a tetrapore. An alternative assumption is that normal muonium correspond to trapping in a free vacancy site. The latter, however seems less natural, primarily because of the relatively small number of vacancies.

As already shown, the spin Hamiltonian for muonium in an octapore is given by formula (6), and is isotropic in a tetrapore (or a site). We shall dub muonium in octapores and tetrapores O-muonium and T-muonium, respectively. We shall assume furthermore that the density of the wave function at zero is much smaller for O-muonium than for T-muonium, and the O-muonium is correspondingly more "swollen." There are no reliable calculations of the wave functions of O- and T-muonium. We note only that mode-dependent estimates^[21] point to a tendency of the wave function at zero to have a lower density for O-muonium than for T-muonium. That the muon can in principle settle in different pores is apparently confirmed by the data of^[22], where precession in iron at low temperatures was investigated.

The possibility of being trapped in both octapores and tetrapores, and the absence of a transition from one type of pore to another within a time on the order of $10^{-6}-10^{-4}$ sec, are even much more probable for the Mu atom than for a muon in a metal. Naturally, one of the muonium states is metastable. However, first, the symmetry of the electron wave function in an octapore is substantially different from that in a tetrapore, and this leads, as usual, to the appearance of hindrances and to a strong decrease of the corresponding matrix elements. Second, as is shown from experiment, the ionization potentials of the muonium electron in the octa- and tetrapores should differ by a value on the order of an electron volt. We recall that $\omega_0^T\approx 0.45\omega_{ov}$, but $\omega_0^O\approx 0.02\omega_{ov}$. The heat of the reaction when the muonium goes from one state to the other can be easily obtained by going through the standard Born cycle:

$$\Delta E=E_T-E_O=V_T-V_O+U_T-U_O. \quad (30)$$

Here E_T , V_T , U_T and E_O , V_O , and U_O are the binding energies of the muonium and the μ^+ meson and the electron ionization potential in the tetrapore and the octapore, respectively. Obviously that at $U_T-U_O\sim 1$ eV the difference ΔE should also be of the order of 1 eV, provided that out of pure coincidence the ionization-potential difference is not offset by the binding energy of the muons. The transition can thus take the form of a complicated multiphonon process with relatively low probability. Finally, muonium has in each given position a much more probable channel, namely diffusion over "its own" pores. All these arguments show that if the muonium has settled in the course of thermalization in a pore of a definite type, then it should not change the type of pore during its lifetime.

It is seen thus that the precession picture depends on the mutual orientation of the crystal and of the magnetic field for O-muonium and is independent of the field for

T-muonium; this explains qualitatively the effect observed in [5].

Formulas (11) and (13) lead to simple relations that are useful in the reduction of the experimental data. At $B \parallel C_3$ ([111] axis)

$$\begin{aligned} \omega_{12} &= 2\Omega_{\parallel} - \Omega_{\perp} \alpha + \omega(1 - \zeta), & \omega_{13} &= 2\Omega_{\parallel} - \Omega_{\perp} \alpha - \omega(1 - \zeta), \\ \omega_{13} &= 4\Omega_{\parallel} - \omega_{12}, & \omega_{12} - \omega_{13} &= \omega_{13} - \omega_{12} = 2\omega(1 - \zeta). \end{aligned} \quad (31)$$

At $B \perp C_3$

$$\begin{aligned} \omega_{12} &= 2\Omega_{\perp} + |\Delta\Omega| \alpha_1 - \Omega \alpha_2, & \omega_{13} &= 2\Omega_{\perp} - |\Delta\Omega| \alpha_1 - \Omega \alpha_2, \\ \omega_{13} &= 4\Omega_{\perp} - \omega_{12}, & \omega_{12} - \omega_{13} &= \omega_{13} - \omega_{12} = 2|\Delta\Omega| \alpha_1. \end{aligned} \quad (32)$$

We used (31) and (32) to obtain estimates made possible by a diagram given in [5]. At $g=2$ we obtained for O-muonium $4\Omega \approx 0.02\omega_{0p}$, $4\Omega_{\perp} \approx 0.01\omega_{0p}$. As already noted, such a substantial anisotropy of the hyperfine-splitting tensor is typical of paramagnetic centers in crystals. Unfortunately, weak fields were not investigated in [5]. At $B=1$ to 20 G, all four precession frequencies will be observed, but ω_{42} vanishes at $B \sim 5-10$ G. Finally, in a zero field one should observe two-frequency precession.

The fact that, as a rule, no precession signal is seen for O-muonium can be readily explained qualitatively. In fact, the relation $\nu_e^O/\omega_0^O \gg \nu_e^T/\omega_0^T$, where ν_e^O and ν_e^T are the muonium electron spin relaxation frequencies, is always valid. First, $\omega_0^O \ll \omega_0^T$, and second, $\nu_e^O > \nu_e^T$, since the O-muonium radius is larger than the T-muonium radius and therefore the cross section for exchange scattering of the conduction electrons is much larger for O-muonium. Thus, O-muonium can be observed only in very pure crystals at low temperatures. Accordingly, when the temperature is raised and the region $\nu_e^O > \omega_0^O$ is reached, the muonium signal from O-muonium will vanish and precession at the muon frequency will appear. On going back to low temperatures, the O-muonium signal reappears. We note, finally, that the data of [18-20], where the polarization of the muons in germanium and silicon in longitudinal fields and the relaxation rate were investigated are in qualitatively good agreement with the assumption that O- and T-muonium can exist simultaneously.

We emphasize in conclusion that the entire theory can be applied with practically no changes to crystal with zincblende structure (InSb, GaAs, and others). The only difference is that in the octapore the crystal field has the symmetry C_{3v} . Particular interest attaches to crystals with axial symmetry (quartz, corundum, crystals with wurtzite structure, etc.), in which the hyperfine splitting has a tensor character, when the muonium can be an interstitial or a substitutional impurity.

The authors are grateful to I. A. Malkin for a useful discussion of the results, and the referee of JETP for useful remarks.

- ¹I. G. Ivanter and V. P. Smilga, Zh. Eksp. Teor. Fiz. **54**, 559 (1968) [Sov. Phys. JETP **27** 301 (1968)].
- ²I. I. Gurevich, I. G. Ivanter, E. A. Meleshko, B. A. Nikol'skiĭ, V. S. Roganov, V. I. Selivanov, V. P. Smilga, B. V. Sokolov, and V. D. Shestakov, Zh. Eksp. Teor. Fiz. **60**, 471 (1971) [Sov. Phys. JETP **33**, 253 (1971)].
- ³I. I. Gurevich, B. A. Nikol'skiĭ, and V. I. Selivanov, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 640 (1972) [JETP Lett. **15**, 453 (1972)].
- ⁴I. I. Gurevich and B. A. Nikol'skiĭ, Preprint IAE-2437, Moscow, 1972.
- ⁵J. H. Brewer, K. M. Crow, F. N. Gygax, R. F. Johnson, B. D. Patterson, and D. F. Fleming, Phys. Rev. Lett. **31**, 143 (1973).
- ⁶K. Weiser, Phys. Rev. **126**, 1427 (1962).
- ⁷K. Weiser, J. Phys. Chem. Solids **17**, 149 (1960).
- ⁸L. D. Landau and E. M. Lifshitz, Kvantovaya mekhanika-Nerelyativistskaya teoriya (Quantum Mechanics-Nonrelativistic Theory), Moscow, Nauka, 1974 [Pergamon].
- ⁹V. G. Baryshevskii, Yadernaya optika polarizovannykh sred (Nuclear Optics of Polarized Media), Belorussian State University, Minsk, 1976.
- ¹⁰A. Abragam and B. Bleaney, Electron Paramagnetic Resonance of Transition Ions, Oxford Univ. Press, 1970.
- ¹¹S. A. Al'tshuler and B. M. Kozyrev, Élektronnyĭ paramagnitnyĭ rezonans. Soedineniya elementov promezhutochnykh grupp (Electron Paramagnetic Resonance. Compounds of Intermediate-Group Elements), Nauka, 1972.
- ¹²R. J. Elliot and K. W. H. Stevens, Proc. R. Soc. London Ser. A **215**, 437 (1952).
- ¹³R. J. Elliot and K. W. H. Stevens, Proc. R. Soc. London Ser. A **219**, 387 (1953).
- ¹⁴P. G. Baranov and R. A. Zhitnikov, Fiz. Tverd. Tela (Leningrad) **10**, 3279 (1968) [Sov. Phys. Solid State **10**, 2592 (1969)].
- ¹⁵P. G. Baranov and R. A. Zhitnikov, Phys. Status Solidi **30**, 859 (1968).
- ¹⁶P. G. Baranov, R. A. Zhitnikov, and N. I. Mel'nikov, Fiz. Tverd. Tela (Leningrad) **13**, 2841 (1971) [Sov. Phys. Solid State **13**, 2392 (1972)].
- ¹⁷G. Feher, Phys. Rev. **114**, 1219 (1959).
- ¹⁸I. I. Gurevich, B. A. Nikol'skiĭ, V. I. Selivanov, B. V. Sokolov, Zh. Eksp. Teor. Fiz. **68**, 806 (1975) [Sov. Phys. JETP **41**, 401 (1975)].
- ¹⁹D. G. Andrianov, L. A. Goncharov, E. V. Minaichev, G. G. Myasishcheva, Yu. V. Obukhov, V. S. Roganov, G. I. Savel'ev, V. G. Firsov, and V. I. Fistul', Fiz. Tekh. Poluprovodn. **10**, 1167 (1976) [Sov. Phys. Semicond. **10**, 692 (1976)].
- ²⁰V. I. Kudinov, E. V. Minaichev, G. G. Myasishcheva, Yu. V. Obukhov, V. S. Roganov, G. I. Savel'ev, V. M. Samoĭlov, and V. G. Firsov, Zh. Eksp. Teor. Fiz. **70**, 2041 (1976) [Sov. Phys. JETP **43**, 1065 (1976)].
- ²¹J. S.-Y. Wang and C. Kittel, Phys. Rev. B **7**, 713 (1973).
- ²²M. Camani, F. N. Gygax, W. Rüegg, A. Schenk, H. Graf, H. Schilling, W. Kündig, B. D. Patterson, P. Roggwiler, H. Fischer, and P. W. Percival, Hyperfine Interaction **2**, 83 (1976).

Translated by J. G. Adashko