Muonium stage of positive-muon depolarization in germanium

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Precession in a transverse magnetic field has been used to determine the modulus and the initial phase of the residual polarization of positive muons in single crystals of germanium as a function of the magnetic field, temperature, and dopant concentration. The activation energy for the transition of the muonium atoms to the diamagnetic state has been calculated. Possible channels of interaction of muonium with the crystal lattice and free charge carriers are considered.

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INTRODUCTION

It is well known that muonium atoms are formed with probability close to unity when positive muons come to rest in single-crystal specimens of germanium and silicon.[1,2] Measurements of the hyperfine splitting frequency for muonium in these materials[3,4] have shown that the effect of the crystal lattice of the semiconductor on the wave function of the muonium impurity atoms leads to a reduction in the hyperfine splitting energy of the latter as compared with the analogous quantity measured in vacuum[5] and in a number of media with a large band gap (quartz, corundum,[6] ice,[3] potassium chloride[7]). Nevertheless, this effect is much smaller than predicted by the theory in the case of a medium with macroscopic permittivity $\varepsilon = 12$ (Si) and $\varepsilon = 15.8$ (Ge), and effective electron mass m^* respectively equal to $0.31m_e$ and $0.17m_e$.[8]

As an illustration, Table I shows the hyperfine splitting frequency ω_0 for muonium in the above media divided by the vacuum frequency $\omega_0^{vac} = 2\pi \times 4463$ MHz[5] (these ratios were obtained by two different methods, namely, measurements of the residual polarization of positive muons in an external magnetic field parallel to the initial direction of particle spins as a function of the field strength, and the method of two-frequency precession of muonium in a transverse magnetic field). Estimates of the corresponding quantity for germanium ($\varepsilon \approx 16$, $m^* = 0.17m_e$) yield $\omega_0/\omega_0^{vac} \sim 10^{-6}$. Table I also shows the results obtained for p-type silicon[6] and for germanium,[10] indicating the possible existence of formations incorporating positive muons and characterized by a much smaller ratio ω_0/ω_0^{vac}.

The data listed in Table I must be taken into account in the analysis of experiments in which the measured quantity is the residual polarization of positive muons as a function of the temperature of the medium. It is known from such experiments[1] that the residual polarization of positive muons in germanium, obtained from measurements of the amplitude of precession at the meson frequency in a transverse magnetic field of ~50 Oe, increases with increasing temperature in the range between 150 and 300 °K from the initial level $P \approx 0.13$ up to the maximum possible value. Experiments of this kind yield only that part of the polarization which is due to positive muons in the diamagnetic "environment" (to within the paramagnetism of the neighboring nuclei), i.e., either free particles or positive muons bound to the crystal lattice by a Ge-H type diamagnetic chemical bond.

The proof that part of the residual polarization of positive muons in germanium is due to transitions from the triplet state of muonium ($J = 1$, $m = 1$) to a state with diamagnetic properties, is provided by observations of a shift in the phase of meson precession at the temperature of the target exposed to the positive muons, i.e., ~180 °K.[11] In our view, the three most likely processes producing such transitions that can be used to explain the experimental results are: 1) thermal ionization of the muonium atoms; 2) formation of the diamagnetic Ge-Mu bond, and 3) interactions of muonium with impurity atoms and free charge carriers.

The specific picture of the interactions of positive muons with the crystal lattice of a semiconductor and, consequently, the choice of a particular variant of the theory of the muonium stage of depolarization of positive muons in a medium[12,13] used to calculate the phenomenological parameters of the depolarization process from experimental data, are very dependent on the actual characteristics of muonium as an impurity atom. Possible values of the radius and ionization potential of muonium in germanium and silicon were analyzed

TABLE I. Measured values of the hyperfine splitting frequency of muonium

Material	ω_0/ω_0^{vac}: Measurements in longitudinal fields	ω_0/ω_0^{vac}: Observation of two-frequency precession
Quartz	1.03 ± 0.05[6]	0.987 ± 0.016[3]
Ice	—	1.07 ± 0.07[3]
Corundum	1.04 ± 0.08[6]	—
Potassium chloride	0.97 ± 0.04[7]	—
Silicon, n-type	0.41 ± 0.03[2]	—
Silicon, p-type	—	0.45 ± 0.02[9]; 0.0198 ± 0.0002[9]
Germanium (p-type?)	0.054 ± 0.005*[10]	0.58 ± 0.01[3]

*Measured at 233 °K

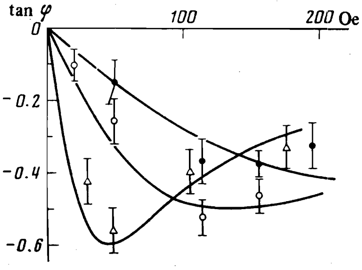

FIG. 1. Measurements of phase shift in the Ge specimen with 10^{14} cm^{-3} As: Δ—177 °K, o—187 °K, •—198 °K. Solid curves calculated from the muonium depolarization theory.

in[8] using experimental values of ω_0/ω_0^{vac} but without taking into account subsequent papers.[9,10] The results of this analysis show that muonium in germanium is a deep donor with probable level energy not less than 1.5 eV. This conclusion can probably be used to exclude the first of the above three processes.

To obtain a detailed understanding of the reasons for the sharp increase in the residual polarization of positive muons in germanium, and the possible effect on this process of the presence of impurity atoms, we have investigated two specimens of germanium doped with arsenic to a concentration of 10^{14} and 1.5×10^{15} atoms per cm^3 in the temperature range between 80 °K (the temperature at which muonium precession was observed in[1]) and 350 °K (when residual polarization of the positive muons approaches unity). These experiments were largely confined to measurements of the amplitude and initial phase of the residual polarization vector of the positive muons (meson precession frequency) as functions of the transverse magnetic field and specimen temperature.

EXPERIMENTAL DATA

The measurements were carried out in the meson channel of the synchrocyclotron of the Laboratory for Nuclear Problems of the Joint Institute for Nuclear Research. Longitudinally polarized positive muons came to rest in the specimen under investigation, which was located in a thermostat, and the decay positrons were recorded with a telescope consisting of two scintillation counters at about 0° to the positive-muon beam. The apparatus, whose operation was described in[14], was used to determine the μ-e decay-time distribution in a transverse magnetic field. The field strength was varied between 0 and 200 Oe.

Computer analysis of the experimental data was used to determine the amplitude and initial phase of the positive-muon spin precession. To determine the absolute polarization P, and to introduce small corrections to the observed phase shift φ connected with the possible geometric asymmetry of the positron telescope, each cycle of measurements corresponding to given temperature and field strength was preceded and followed by experiments with a standard target of bromoform ($CHBr_3$), a material in which the polarization of the positive muons was close to unity and independent of the magnetic field whilst the phase-shift effect was absent.

The absence of systematic errors in phase shift measurements was also checked by reversing the direction of the external magnetic field in successive experiments.

Preliminary experiments were carried out to determine the temperature range (which turned out to be the same for both specimens of germanium) in which measurements of the residual polarization and phase shift as functions of the transverse field could be used to determine the parameters of the theory with sufficient statistical accuracy.[11] In subsequent experiments, detailed measurements (four or five values of field strength for each temperature) were confined to the temperature range between 160 and 200 °K. As an example, Fig. 1 shows the results obtained in a series of experiments with a Ge specimen (10^{14} cm^{-3} As). At lower temperatures, the lifetime of free muonium atoms in germanium is found to be longer and, if the phase shift effect is to be observed, this requires the use of magnetic fields of such low strength that the meson-precession period exceeds the lifetime of the positive muon. This substantially reduces the statistical precesion with which φ is determined.

The possibility of direct observation of precession with the muonium frequency in a transverse field of about 10 Oe was investigated at still lower temperatures (about 100 °K). This precession was detected in the Ge specimen containing 10^{14} cm^{-3} As and was found to have a relaxation time of 57 ± 18 nsec and an initial amplitude corresponding to a polarization of $P=0.42\pm0.05$. Since, under the conditions of this experiment, positive muons in the muonium atoms with $m=0$ are completely depolarized, and a proportion of the particles precess with the meson frequency because of the presence of the prompt channel (its contribution to the residual polarization of the positive muons is $\beta=0.10$), the admixture of depolarization channels which we have ignored ($1-2P-\beta=0.06\pm0.010$) can be regarded as unimportant. In similar experiments performed with the Ge specimen containing 1.5×10^{15} cm^{-3} As, the muonium precession was not detected. The minimum relaxation time for this precession, when it could be reliably identified in the above experiments, did not exceed 20 nsec.

The residual polarization of positive muons in a magnetic field of about 50 Oe was determined at temperatures in excess of 200 °K. Figure 2 shows the results. The phase shift of the muonium precession under these conditions did not exceed the statistical error of the experiments. The reduction in the residual polarization of the positive muons during the observation time, which occurred only for the Ge specimen with 10^{14} cm^{-3} As, was noted at temperatures in excess of 300 °K. The relaxation time of the meson precession was found to decrease monotonically with increasing temperature from 25 ± 12 μsec at 300 °K to 6 ± 1 μsec at 350 °K.

ANALYSIS OF RESULTS

Analysis of the experimental data, most of which consisted of calculating the parameters of the theory of the muonium stage of positive-muon depolarization in germanium as functions of specimen temperature, was

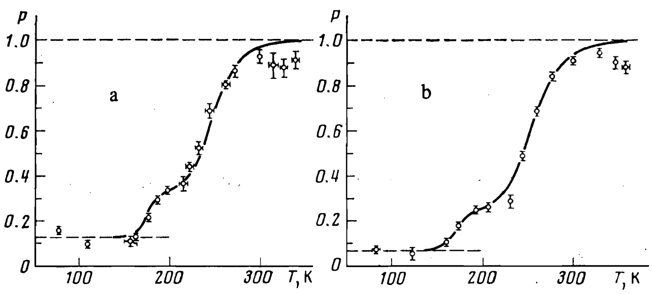

FIG. 2. Residual polarization of positive muons as a function of temperature: a—Ge specimen with 10^{14} cm^{-3} As, b—Ge specimen with 1.5×10^{15} cm^{-3} As. Magnetic field ~50 Oe. Solid curve calculated from the muonium depolarization theory: a—$\beta=0.13$, $\nu\tau=0.9$, $\Delta E=0.184$ eV; b—$\beta=0.07$, $\nu\tau=1.5$, $\Delta E=0.166$ eV.

based on the variant of the theory given by Ivanter and Smilga.[13] This involves a consideration of the four processes which, in the last analysis, determine the residual polarization of positive muons:

1. Depolarization of muonium atoms, formed in the state with $m=0$, due to the hyperfine interaction. The parameter of the theory that characterizes the rate of depolarization is the frequency ω_0 of hyperfine splitting of muonium in the medium. We assume that $\omega_0 = 2\pi(2580\pm50)$ MHz, i.e., the value that was obtained experimentally by Gurevich *et al.*[3] This value was taken to be independent of temperature and the same for all the specimens that were investigated. The fact that formations including positive muons can exist with a substantially reduced hyperfine splitting frequency[9,10] undoubtedly requires further detailed consideration, and this will be possible only when more experimental data become available.

2. Exchange of electrons between the muonium atoms and the medium, which leads to the relaxation of the electron spin in the muonium with frequency ν, a process which enhances the depolarizing influence of the medium on the positive muons for $\nu<\omega_0$ and tends to suppress it for $\nu\gg\omega_0$.

3. Formation of the diamagnetic chemical bond or thermal ionization of muonium (these two processes are indistinguishable in the experiment unless the dependence of the experimental results on temperature is investigated), which leads to the completion of the muonium stage of the depolarization process in an average time τ. It is assumed that post-muonium depolarization is absent to within processes occurring in a time much greater than τ (slow depolarization of positive muons[15]).

4. Interaction of positive muons or muonium atoms with the ambient medium in the epithermal region, or with nonequilibrium products formed at the end of the positive-muon track. If the final result of such interactions is the appearance of diamagnetic formations containing the positive muons, the residual polarization β of the positive muons due to these factors should be independent of the external field but may vary with the temperature of the specimens because this temperature influences the rate at which radiation damage in the crystal lattice is reduced.

Ivanter and Smilga have obtained relatively complicated expressions[13] for the measured P and φ as functions of the parameters of the theory and the external magnetic field H. These expressions can be substantially simplified when the muonium depolarization stage is long in comparison with the characteristic time for the depolarization of positive muons in muonium atoms with $m=0$, i.e., when $\omega_0\tau\gg1$. Under these conditions, the phase shift of the meson precession is also a maximum (for a given transverse field). The result of these simplifications is:

$$P=\frac{1}{2}\left[\frac{(1+\beta+2\beta\nu\tau)^2+4\beta^2(\omega'\tau)^2}{(1+\nu\tau)^2+(\omega'\tau)^2}\right]^{1/2},$$
$$\operatorname{tg}\varphi=-\omega'\tau\left\{(1+\nu\tau)+\frac{2\beta}{1-\beta}\left[(1+\nu\tau)^2+(\omega'\tau)^2\right]\right\}^{-1}, \quad (1)$$

provided $\omega_0\tau\gg1$, $\omega'/\omega_0<1$. In this expression, $\omega'=g'H$ is the muonium precession frequency and $g'=8.76\times10^6$ sec$^{-1}\cdot$G^{-1} is the gyromagnetic ratio for the muonium atom. In accordance with (1), the parameters τ, ν (in the form of the product $\nu\tau$), and β were determined by applying a least-squares analysis to the measured P and φ as functions of the transverse magnetic field. The results of this analysis are given in Table II.

It follows from the final results that, firstly, the lifetime τ of muonium atoms in germanium is rapidly reduced with increasing temperature (as compared with the $T^{-1/2}$ law), which suggests that there is an energy barrier for the Mu → μ^+ transitions. Secondly, in the temperature range which we have investigated, the product $\nu\tau$ remains approximately constant although one of the factors (τ) changes by more than an order of magnitude. Thirdly, we note that the parameters calculated for the two specimens are very similar although the impurity-atom concentrations differ by a factor of 15.

CALCULATION OF THE BARRIER HEIGHT

The values of τ given in Table II at different temperatures can be used to determine the height ΔE of the energy barrier for the transition of the muonium atoms to the state with diamagnetic properties if we write the probability of such transitions in the form

$$1/\tau=Z\exp(-\Delta E/kT), \quad (2)$$

TABLE II. Calculated parameters of the muonium depolarization stage.

T, K	$\tau\cdot10^9$, sec	$\nu\tau$	β	χ^2
Ge(As–$1\cdot10^{14}$ cm^{-3})				
161±1	17±3	0.7±0.2	0.12±0.02	3.5 (7) *
177±1	7±2	0.6±0.2	0.13±0.02	7.9 (9)
187±1	2.9±0.4	1.1±0.2	0.13±0.02	3.8 (9)
198±2	1.4±0.5	1.2±0.2	0.15±0.03	9.0 (9)
Ge(As–$1.5\cdot10^{15}$ cm^{-3})				
160±1	24±5	2.0±0.5	0.08±0.02	9.6 (9)
173±1	6.2±1.4	1.2±0.2	0.06±0.02	2.6 (9)
192±1	2.2±0.4	1.6±0.3	0.06±0.02	3.6 (7)

*Numbers in parentheses indicate the number of experimental points.

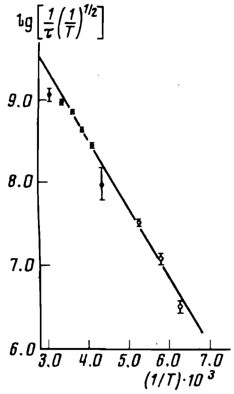

FIG. 3. Duration of the muonium stage in the Ge specimen with 1.5×10^{15} cm^{-3} As as a function of temperature. The abscissa axis indicates values of the reciprocal of the temperature, and the ordinate axis gives the logarithm of the rate of interaction of muonium with germanium, including the factor $T^{1/2}$: o—value of τ determined by the least-squares method from experimental data, ●—based on experimental values of polarization. The calculation of τ was made assuming $\beta = 0.07$, $\nu\tau = 1.5$.

where $Z \sim T^{1/2}$ is the frequency of collisions between muonium and the crystal-lattice atoms or free electrons (both germanium specimens are nondegenerate in the temperature range which we considered). These calculations yielded $\Delta E = 0.17 \pm 0.02$ eV for Ge containing 10^{14} cm^{-3} As and $\Delta E = 0.20 \pm 0.02$ eV for Ge containing 1.5×10^{15} cm^{-3} As. The collision frequency Z can be obtained only very approximately because the barrier height is large in comparison with kT. Thus, 3×10^{12} sec$^{-1} < Z < 4\times10^{13}$ sec^{-1} for the first specimen and 3×10^{13} sec$^{-1} < Z < 4\times10^{14}$ sec^{-1} for the second. These estimates were made for the temperature of 175 °K.

The statistical precision of the calculations of barrier height and collision frequency can be substantially increased by using the experimental data obtained at higher temperatures. Since the phase shift was not measured in this case, certain additional assumptions are necessary in the calculations. Bearing in mind the rapid temperature dependence of the duration of the muonium depolarization stage as compared with the other quantities in Table II, we assume that the product $\nu\tau$ and the parameter β remain constant and are given by averaging the data in Table II:

$$\nu\tau=0.9\pm0.2,\quad \beta=0.13\pm0.01 \quad \text{for Ge(As} - 1\cdot10^{14}\,\text{cm}^{-3}),$$
$$\nu\tau=1.5\pm0.4,\quad \beta=0.07\pm0.01 \quad \text{for Ge(As} - 1.5\cdot10^{15}\,\text{cm}^{-3}).$$

In that case, τ can be calculated unambiguously from the experimental polarization P in a transverse field of ~ 50 Oe, provided the rigorous expressions are used[13] because the approximation $\omega_0\tau \gg 1$ is then inadequate.

It is clear that a further check on the agreement between experimental data and the proposed picture of interactions between positive muons and germanium can be obtained by introducing into the calculation the hyperfine splitting frequency ω_0, which is measured under totally different conditions and was not previously used.

Figure 3 shows the calculated values of τ in the temperature range 160–350 °K for the Ge specimen with 1.5×10^{15} cm^{-3} As. It shows the logarithm of the Mu $\to \mu^+$ transition frequency (including the factor $T^{1/2}$) as a function of the reciprocal of temperature which, according to (2), should be a straight line. The three points on the right-hand side correspond to the values of τ given in Table II. The points on the left-hand side were calculated from the general formulas given in[13], using the measurement results shown in Fig. 2. The following values were used in the calculation: $\omega_0 = 2\pi\times2580$ MHz,[3] $\nu\tau = 1.5$, and $\beta = 0.07$ (Table II). The energy-barrier height ΔE and collision frequency Z (at 175 °K) were determined from the straight-line equation by the least-squares method. The results are:

$$\Delta E=0.166\pm0.005\,\text{eV},\quad 0.8\cdot10^{13}\,\text{sec}^{-1}<Z<2\cdot10^{13}\,\text{sec}^{-1};$$

where $\chi^2 = 6.5$ and the analysis was based on seven experimental points with points corresponding to the highest temperatures excluded.

Similar calculations performed for the Ge specimen with 10^{14} cm^{-3} As, $\nu\tau = 0.9$, and $\beta = 0.13$ yielded

$$\Delta E=0.184\pm0.007\,\text{eV},\quad 1\cdot10^{13}\,\text{sec}^{-1}<Z<5\cdot10^{13}\,\text{sec}^{-1};$$

with $\chi^2 = 12$ (10 experimental points).

DISCUSSION OF RESULTS

Although the choice of the particular variant of the theory used in the analysis of the results and the method employed to calculate ΔE and Z are not totally undisputed, let us consider some of the consequences of the foregoing material.

We note to begin with that, under the conditions of the above experiments, the electron density in the conduction band of the semiconductor is practically constant and equal to the concentration of the impurity atoms. The estimated collision frequency between muonium and free electrons is $Z = \sigma n v \approx 3\times10^7$ sec^{-1} for $\sigma \approx 10^{-15}$ cm^2, density $n = 1.5\times10^{15}$ cm^{-3}, and electron velocity $v \approx 2\times10^7$ cm·sec^{-1}, i.e., it is much less than the result calculated from the experimental data. On the other hand, the number of collisions between muonium and crystal-lattice atoms $Z = v/l$ ($v \approx 5\times10^5$ cm·sec^{-1} is the thermal velocity of the muonium and $l \approx 10^{-8} - 3\times10^{-8}$ cm is the mean distance between neighboring atoms) is of the same order (10^{13} sec^{-1}) as the calculated result.

The predominant contribution of the lattice to the Mu $\to \mu^+$ process is also confirmed by the weak dependence of the rate of the process on the density of conduction electrons. On the other hand, muonium depolarization at low temperatures, the rate of which is inversely proportional to the muonium-precession relaxation time and is very dependent on the amount of impurity, can be explained in terms of interactions between muonium and free electrons. The measured relaxation time is then found to correspond to a cross section of the order of 10^{-14} cm^2 (this is the upper limit because the possible contribution of subbarrier and dipole-dipole interactions has been ignored).

The assumption that the product $\nu\tau$ is contant whilst the duration of the muonium stage changes by more than two orders of magnitude means that we are regarding $Mu \to \mu^+$ transitions and reversals of the direction of electron spin in muonium as different consequences of the same process. This process can be regarded, for example, as the transfer of an electron from a local donor level, formed as a result of the presence of a muonium atom between the crystal lattice sites, to the conduction band. The character of the final state of the positive muon depends on the direction of the spin of the delocalized electron relative to the spin of the positive muon.

CONCLUSIONS

The experimental results reported in this paper suggest, in our view, that further studies of the properties of muonium in the crystal lattices of semiconductors should yield useful results. Elucidation of the actual energy level scheme that appears during the interaction between muonium electron shells and the neighboring lattice atoms is an important problem, the solution of which should be helpful in the understanding of the formation of deep donor levels in semiconducting materials.

We emphasize the satisfactory agreement between the calculated barrier heights corresponding to narrow and broad temperature ranges. This may be regarded as evidence for the fact that the hyperfine splitting frequency of muonium in germanium remains constant in the temperature and dopant density ranges we have investigated.

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A soliton model of particles of the ψ-boson type

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Coherent states (condensates) constructed on the basis of classical soliton-like solutions of a one-dimensional field equation are considered. The narrowness of the ψ boson is explained by the smallness of the amplitude for the transition of the condensate into a state with definite number of particles, much smaller than the average number. The ψ' boson is interpreted as a condensate with a mean field which differs little from the mean field of the ψ state, and it is proved that the $\psi' \to \psi$ transition is not suppressed.

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INTRODUCTION

The suppression of the number of pion channels characteristic of the ψ particles manifests itself particularly clearly in comparing the decays of the ψ' into 3π and $2\pi\psi$. The three-particle Lorentz invariant phase space for the first of these decays is approximately 260 times larger than for the second mode, whereas the rates are comparable (the $2\pi\psi$ channel is responsible for about 30% of the width of the ψ'). Our fundamental idea

