Characteristics states of muonium (μ^+meson) in silicon in longitudinal magnetic fields

S. G. Barsov, A. L. Getalov, V. A. Gordeev, R. F. Konopleva, S. P. Kruglov, V. I. Kudinov,
L. A. Kuz'min, S. M. Mikritych'yants, E. V. Minaĭchev, G. G. Myasishcheva, Yu. V. Obukhov,
G. I. Savel'ev, V. G. Firsov, and G. V. Shcherbakov

Institute of Theoretical and Experimental Physics
and Leningrad Institute of Nuclear Physics, USSR Academy of Sciences
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Experimental results are presented of an investigation of the depolarization of positive muons in silicon in longitudinal magnetic fields. It is shown that the mesic component of the polarization is due to thermal ionization of the "anomalous" muonium system. The data are discussed with allowance for the possible chemical bonding of the muonium atoms with the crystal lattice.

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INTRODUCTION

It is known from experiments with polarized positive muons (the μSR method) that silicon contains, besides muonium atoms, also a bound muon–electron system in which the hyperfine interaction is anisotropic. It follows from Ref. 1 that to describe the properties of such a system, called "anomalous" muonium, it is necessary to introduce two constants, $A_\perp$ and $A_\parallel$, in the interaction Hamiltonian

$$\hat{\mathscr{H}}=A_\perp(\sigma_{ex}\sigma_{\mu x}+\sigma_{ey}\sigma_{\mu y})+A_\parallel\sigma_{ez}\sigma_{\mu z}+\hat{\mathscr{H}}_{\text{ext}},$$

where $\hat{\mathscr{H}}_{\text{ext}}$ are the terms that take into account the interaction with the external magnetic field, and the subscripts $\perp$ and $\parallel$ indicate the relative positions of the coordinate axes and of the principal diagonal of the single crystal, the Z axis being aligned with the $\langle 111\rangle$ axis; σ_{ei} and $\sigma_{\mu i}$ are the components of the spin vectors of the electron and muon, respectively. Both constants were measured by separating the Fourier components of the distribution of the μe-decay times in an external magnetic field, and it was shown that the aggregate of the experimental data agrees well with the calculation. The constants turned out to equal $A_\perp/h=92,1\pm 0,3$ and $A_\parallel/h=17,1\pm 0,3$ MHz, i.e., much less than the frequency of the hyperfine splitting of muonium in silicon, $\omega_0(\mathrm{Si})=1980\pm 90$ MHz.[2] The experiments have shown that both types of muonium exist in the silicon simultaneously,[3] i.e., part of the muons are contained in the anomalous muonium, and another part in the normal muonium atoms. In addition, there is a third group of muons that precess in a direction perpendicular to the vector of the initial polarization, in an external magnetic field at the Larmor precession frequency of the free muon. We shall use hereafter the notation established in the papers dealing with this problem, and refer to the foregoing states as the Mu*, Mu, and μ^+ components, respectively.

The possible physical causes of the appearance of the anomalous muon precession frequencies in silicon were discussed in the literature many times.[1,2,4-6] These include, for example, a model according to which the muon can have in the silicon crystal lattice two equilibrium positions. In one of them—the octapore of the single crystal—the spherical symmetry of the hyperfine interaction is disturbed.[5,6] The other scheme, proposed by us earlier,[4] is based on the assumption that a diamagnetic chemical bond exists between the muonium and the lattice. The anisotropic hyperfine interaction between the muon and the electron occurs if the latter is localized on one of the silicon atoms adjacent to a muon.[4]

Experimental data favoring one or the other mechanism for the presence of three muon states in silicon can be obtained by studying the temperature dependences of the population of each state in a wide temperature interval, inasmuch as experiment shows that in this case it is possible to investigate the characteristics of the transitions between the various components.[3,4,7,8] However, the determination of these dependences, based on separation of the corresponding Fourier components in the distribution of the μe-decay times, is made complicated by a number of factors that lead to rapid relaxation of the precession of Mu* and Mu. First among them are the spin-exchange and dipole-dipole interactions between the electron of the Mu* and Mu states and the free carriers of the semiconductors; second, a jumplike change of the precession frequency takes place in a perpendicular external field in the presence of transitions between the different states; under certain relations between the transition probability and the precession frequency this change also depolarizes the muons. In the present paper, whose main purpose is a quantitative determination of the amplitude of each of the three muon states in silicon at various temperatures, we use therefore longitudinal magnetic fields in addition to the perpendicular ones. Under conditions of rapid relaxation of the muonium spin system, experiments performed in this manner yield more information, since the longitudinal magnetic field hinders to a considerable degree the depolarizing action of the aforementioned factors.[9] The results reported below are analyzed with the earlier work taken into account.[4,10,11]

EXPERIMENTAL RESULTS

The experiments were performed in the muon channel of the synchrocyclotron of the Leningrad Institute of Nuclear Physics of the USSR Academy of Sciences.

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The characteristics of the channel and the parameters of the recording apparatus are given in Refs. 4 and 8. Two p- and n- type silicon samples with impurity-atom concentrations $\sim 5\times 10^{12}$ cm^{-3} and 1.6×10^{13} cm^{-3} respectively were investigated in the temperature interval 80–730 K. Both samples were so oriented that the direction of one of the axes $\langle 111\rangle$ of the single crystals coincided with the direction of the muon beam and with the axis of the telescope that recorded the decay positrons. The sample thickness was about 4 g/cm^2, and the counting rate of the muon stopping in the target was $\sim 8\times 10^3$ sec^{-1}. To maintain the specified temperature accurate to ±1° we used thermostats of two types for the low- and high-temperature regions, respectively. The external magnetic fields were produced by two pairs of Helmholtz coils. The maximum field intensities reached 470 Oe and 200–270 Oe in the perpendicular and longitudinal directions, respectively.

Since the experiments were not intended to determine the muonium precession frequencies, the width of one channel of the recording apparatus, chosen for the time averaging of the frequencies, was quite large, 20–40 nsec in the different experimental runs (the maximum event registration time interval reached ~8 μsec). The distribution of the μe-decay times, obtained in a longitudinal magnetic field with a positron telescope placed at an angle ~0° relative to the direction of the muon beam, provides most information on the time dependence of the polarization, but not the absolute values of the polarization. The normalization needed to make the calculations definite was carried out by comparing the results of two experiments, the second of which was performed in the perpendicular field. To decrease the possible statistical fluctuations of the registration efficiency, the measurements in the longitudinal and perpendicular fields were alternated. The distributions obtained in perpendicular fields were computer-reduced by the maximum-likelihood method in accordance with the expression

$$N_i(\perp)=N_0[1+a(R)Pe^{-\lambda t_i}\cos(\omega t_i+\delta)]. \tag{1}$$

To simplify the exposition, we have omitted from (1) the background of the random coincidences and the exponential factor that takes the muon decay into account, i.e., $N_i(\perp)$ is the number of counts in the i-th analyzer channel corresponding to the time t_i, with the background subtracted, multiplied by the muon-decay exponential factor; N_0 is a quantity whose meaning on the plot is the average precession line and depends on the summary statistics of the experiment; a (R) is the μe-decay asymmetry coefficient, measured in control experiments with a graphite target thickness R g/cm^2 chosen equal to the thickness of the investigated target; P and λ are respectively the initial polarization and the relaxation rate of the mesic component; ω and δ are the frequency and the initial phase of the mesic precession.

The distributions obtained under these conditions, but in longitudinal magnetic fields, can be expressed in the form

$$N_i(\|)=N_0[1+a(R)P(t_i)]. \tag{2}$$

Here $P(t_i)$ is the time-dependent polarization in the longitudinal magnetic field and is calculated by introducing the parameter N_0 determined from the results of the experiment in a perpendicular field.

The program of the experiments with silicon was based on the data of our preceding work,[4] in which we investigated the temperature dependence of the amplitude of the μ^+ component in perpendicular fields, and on the work of Patterson *et al.*,[1] from which we took, for our calculations, the measured constants of the hyperfine interaction of the Mu* state. The maximum longitudinal magnetic field intensity $H\approx 270$ Oe is sufficient to prevent almost completely the Mu* depolarization, and at the same time does not influence substantially the hyperfine interaction in the Mu state (the field intensity produced by the muon magnetic moment at a distance equal to the electron orbit is respectively ~30 and ~700 Oe for the Mu* and Mu states).

The material obtained after reducing the experimental data is shown in Figs. 1–3 and contains dependences of three types. Figure 1 shows the decrease of the muon polarization in longitudinal magnetic fields, which takes place during the observation time for n-silicon at a sample temperature 78 K. It is seen from the figure that the polarization has two components, one with a relaxation rate on the order of 10^6 sec^{-1}, and the other practically independent of time. A similar picture was observed for the same sample at 204 K. With increasing field intensity, the initial summary (both components) polarization P_0 increases, and the relaxation slows down. The character of these changes is shown in greater detail in Fig. 2, which shows plots of P_0 and λ against the longitudinal field intensity. The measurements with n-silicon were made at three temperatures, 78, 204, and 293 K, while those with p-silicon at 78 K. In the latter case no muon depolarization was discerned during the observation time ($\lambda < 1\times 10^5$ sec^{-1}). It follows from Fig. 2a that the law governing the increase of the initial polarization with increasing longitudinal magnetic-field intensity is practically independent of the type of sample conductivity and agrees qualitatively with the value of the critical field intensity for the Mu* component, i.e., is of the order of 30 Oe. However, a comparison of the data obtained for the n-sample at 78 and 204 K (Fig. 2) points to the presence of other muon depolarization mechanisms in weak

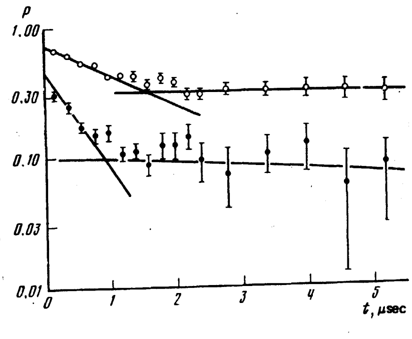

FIG. 1. Time dependence of muon polarization in n-silicon at 78 k. Ordinates—polarization P (in logarithmic scale). Circles—longitudinal magnetic field intensity 240 Oe (○) and 20 Oe (●).

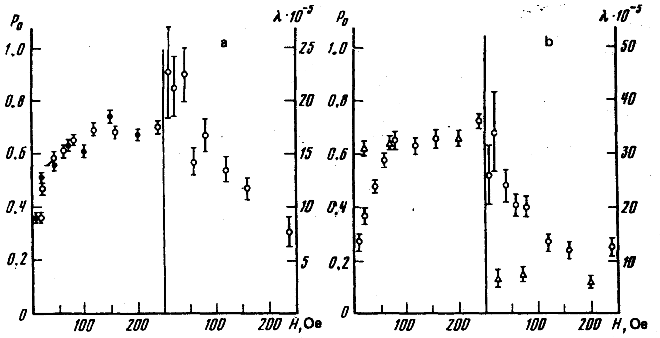

FIG. 2. Dependence of initial muon polarization P_0 (left) and of its relaxation rate λ (right) on the longitudinal field intensity: a) in n-silicon (○) and p-silicon (●) at 78 K; b) in n-silicon at temperatures 204 K (○) and 293 K (△). Ordinates—values of P_0 and of the relaxation rate (in units of 10^5 sec^{-1}). Values of λ less than 1×10^5 sec^{-1} are not shown in the figure for p silicon.

longitudinal fields. These mechanisms are apparently due to interactions of the electrons of the Mu* and Mu states with the free carriers, interactions that are more intense at 204 K. This conclusion is confirmed by results of direct measurements of the relaxation rates of the muonium precessions in perpendicular fields[4] (for the same samples), which exceed approximately by one order of magnitude the values shown in Fig. 2. At higher temperatures ($T = 293$ K, Fig. 2b), the $P_0(H)$ and $\lambda(H)$ dependences point to a qualitatively different character of the muon depolarization processes compared with the low temperature, a fact particularly noticeable at a longitudinal field intensity $H \approx 20$ Oe.

A similar conclusion can be drawn also on the basis of an analysis of the temperature dependences of $P_0(T)$ and $\lambda(T)$ shown in Fig. 3 and obtained in a longitudinal magnetic field of intensity $H_{\parallel} = 200$ Oe and in a perpendicular field $H_{\perp} = 420$ Oe. It is seen that the polarization of the mesic component in the temperature range 200–240 K increases sharply with increasing temperature, whereas in a longitudinal field the polarization is approximately constant. This fact can be interpreted as the onset of a $\mathrm{Mu}^* \to \mu^+$ transition at the indicated temperature, with a rate $\alpha(\mathrm{Mu}^* \to \mu^+) < \tilde{\omega}(\mathrm{Mu}^*)$ at $T < 200$ K, where $\tilde{\omega}(\mathrm{Mu}^*)$ denotes the rates corresponding to the set of precession frequencies of the Mu* state. At $T > 240$ K, on the contrary, $\alpha(\mathrm{Mu}^* \to \mu^+) \gg \tilde{\omega}(\mathrm{Mu}^*)$. Obviously, the polarization "lost" in the longitudinal magnetic field corresponds to muonium atoms with a quantum number $m = 0$, whose fraction depends weakly on the sample temperature all the way to $T \approx 400$ K, above which the intensity of the relaxation processes increases sharply and hinders substantially the quantitative measurements. In perpendicular magnetic fields and in this temperature region (p-silicon, $T \approx 380$ K), the preliminary data have pointed to the presence of a shift of the initial meson precession phase ($-\tan\varphi \approx 0.20$–$0.30$), indicating apparently the presence of transitions between the states Mu and μ^+. However, in view of the high relaxation rates of the mesic precessions, the statistical accuracy of the experiments must be improved if precision results are to be obtained.

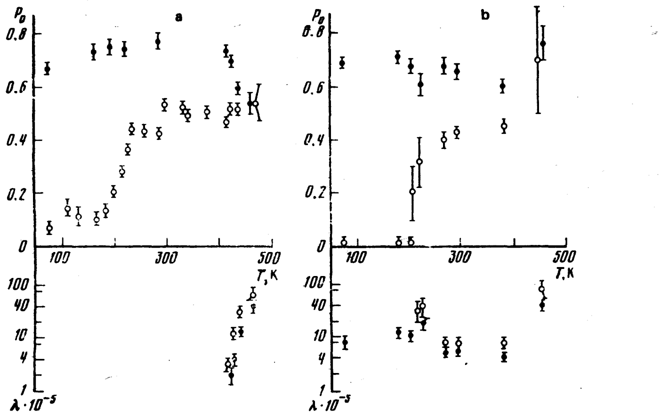

FIG. 3. Temperature dependence of the initial muon polarization P_0 (top) and of the relaxation rate λ (bottom) in a perpendicular magnetic field ~420 Oe (○) and in a longitudinal field ~200 Oe (●): a) p-silicon, b) n-silicon. Below 400 K, $\lambda < 1 \times 10^5$ sec^{-1}.

DISCUSSION OF RESULTS

It is useful in this section to estimate the amplitude of each state at different temperatures and to assess the possibilities and causes of the existence of transitions between them. We formulate first the principal facts that can be deduced from the experimental material.

1. The fraction $\beta(\mathrm{Mu})$ of the Mu component depends little on temperature in both samples. The largest spread of the experimental points (Fig. 3, longitudinal magnetic field) is not monotonic and allows us to assume that $\beta(\mathrm{Mu})$ is constant in the interval $78\text{ K} < T < 400$ K. Averaging of the results yields $P_0 = 0.73 \pm 0.02$ (the error takes into account the spread of the points) for p-silicon and $P_0 = 0.67 \pm 0.03$ for n-silicon in a longitudinal field $H = 200$ Oe. The amplitude of the Mu component can be obtained from the given values of the polarization using the known formulas of Ref. 12 and assuming low rates of the spin-exchange (frequency ν) and chemical (characteristic time of muonium stage τ) interactions compared with the hyperfine interaction frequency ω_0, i.e., $\nu \ll \omega_0$ and $\omega_0 \tau \gg 1$. The validity of this assumption follows from the presence of long-lived muonium at low temperatures and from the constancy of P_0 in a longitudinal field in the temperature interval indicated above. Using, according to the data of Ref. 2, for muonium in silicon a critical field intensity $H_{\mathrm{crit}}(\mathrm{Si}) = 704 \pm 35$ Oe, we have respectively $\beta(\mathrm{Mu}) = 2.08\,(1 - P_{0\parallel}) = 0.56 \pm 0.05$ and $\beta(\mathrm{Mu}) = 0.69 \pm 0.06$ for p- and n-silicon.

2. The character of the "restoration" of the muon polarization in the Mu* state by a longitudinal magnetic field agrees with the conclusions of Refs. 1, 5, and 6. Experiment indicates that the Mu* component exists in the temperature interval 78–204 K, and that at higher temperatures it goes over into the μ^+ component. The fraction $\beta(\mathrm{Mu}^*) = 0.39 \pm 0.03$ of the Mu* component was calculated in accord with the results of Refs. 1, 5, and 6 using for the calculations the experimental data for p-silicon at 78 K, i.e. under the conditions when the precession frequencies of Mu* were observed in experiment.[4]

3. At low temperatures the mesic component of the polarization in n-silicon is practically zero and its fraction in p-silicon is $\beta(\mu^+) = 0.11 \pm 0.03$. At the same time, at temperatures 300–400 K the fraction of the mesic component is $\beta(\mu^+) = 0.50 \pm 0.02$ and $\beta(\mu^+) = 0.42 \pm 0.03$ for p- and n-silicon, respectively. These values, compared with the values given above for $\beta(\mathrm{Mu})$, show that the μ^+ state has evolved from the Mu^* component. The errors in the considered scheme can be assessed from the criterion of the summary polarization $\beta(\mu^+) + \beta(\mathrm{Mu}) = 1.06 \pm 0.06$ and 1.11 ± 0.07 for p- and n-silicon, or slightly more than unity.

4. In the temperature region $T > 400$ K, the polarization relaxation rate in both silicon samples increases both in the perpendicular and in the longitudinal magnetic field. This means that at $T > 400$ K the mesic component becomes unstable. A comparison of the polarization relaxation rates in the longitudinal and perpendicular field (the longitudinal field does not stop the relaxation, but decreases its rate by a factor 1.5–3) indicates that the products of the decay of the mesic components are muonium atoms, and that we have both a chain of successive transitions $\mu^+ \to \mathrm{Mu} \to \mu^+$ etc. and spin-exchange interactions between the muonium electron and the conduction electrons of the medium. The validity of this conclusion is confirmed, first, by the fact that in n-silicon the relaxation is observed also at lower temperatures, in contrast to p-silicon; second, the used silicon samples go over into the region of intrinsic conductivity at $T > 400$ K (see, e.g., Ref. 13).

On the basis of the foregoing, we can represent the picture of the interactions of the positive muons with the crystal lattice of silicon in the following fashion. At low temperature, after the nonequilibrium carriers produced at the end of the muon track melt away, two states are produced, Mu^* and Mu. The mesic component in p-silicon is the consequence of the formation of part of the Mu^* in a polarized state. In this case the energy level of the Mu^* system is close to the bottom of the conduction band and cannot be filled because in contrast to n silicon, the concentration of the free electrons in the band is negligibly small.

At temperatures 200–240 K thermal ionization of the Mu^* system and the transition $\mathrm{Mu}^* \to \mu^+$ take place. For a mesic component to be produced in a perpendicular field the rate V of the ionization process must exceed the maximum precession frequency of the Mu^* state, i.e., $V = Z \exp(-E/kT) \gg 3.10^8\ \mathrm{sec}^{-1}$, where E is the electron binding energy. Assuming the pre-exponential factor to be of the order of 10^{12}–10^{13} sec^{-1}, we obtain a binding energy $E = 0.1$–0.2 eV. The mesic component is either the free μ^* meson particle (in which case the energy level of the Mu component is in the forbidden band of silicon, for otherwise the free muon would capture very rapidly a valence-band electron), or a muon that forms a chemical bond,[4] with the crystal lattice of silicon for example of the $[\mu^+ e^- e^- \mathrm{Si}^+]$ type.

Taking the foregoing material into account, the choice between the two models mentioned in the Introduction, anomalous muonium in a single-crystal octapore or normal muonium in a tetrapore and the $[\mu^+ e^- e^- \mathrm{Si}^+]$ system, depends on the possible existence of an ionization potential on the order of 0.1–0.2 eV for the Mu^* and Mu atoms introduced into the lattice. This conclusion is valid for the latter, as follows from the $\mathrm{Mu} \to \mu^+$ transitions observed in the present study at high temperatures. According to the premises of Ref. 14, it can be concluded from the measured hyperfine interaction constant of Mu that its energy level is lower than the forbidden band of silicon. Thus, the inclusion of a diamagnetic chemical bond in the scheme of muon interactions with the crystal lattice is fully justified.

It should be noted in conclusion that the use of a combination of longitudinal and perpendicular magnetic fields makes it possible to distinguish clearly between the muon depolarization processes, and to identify the processes connected with the participation of various states of the muonium (μ^+ meson). The acquisition of such data is necessary to obtain a complete picture of the interactions of muons with the crystal lattices of solids, including semiconductors.

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