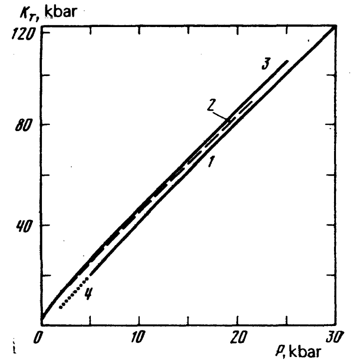

FIG. 2. Dependence of the isothermal modulus of hydrostatic compression of n-H_2 on pressure. 1—77 K, present data; 2—$T=4.2$ K (Ref. 1); 3—$T=4.2$ K (Ref. 2); 4—$T=77$ K (Ref. 14), fluid phase.

Table II gives the values of the molar volume and of the isothermal modulus of hydrostatic compression $K_T = (-V\partial P/\partial V)_T$, calculated from Eq. (2). The same values are shown in Figs. 1 and 2 together with the data obtained by others.

On the basis of the $P-V$ dependence at 77 K and of the data of Anderson and Swenson[2] at 4.2 K we can estimate the average isobaric coefficient of volume expansion $\overline{\alpha}_P = (\Delta V/V\Delta T)_P$. The coefficient $\overline{\alpha}_P$ decreases rapidly ($\overline{\alpha}_P \sim 1/P$) with increasing pressure: $\overline{\alpha}_P \approx 7.5\cdot 10^{-4}$ deg^{-1} at 5 kbar and $\overline{\alpha}_P \approx 1.4\cdot 10^{-4}$ deg^{-1} at 25 kbar. The contribution made to the pressure by the lattice thermal vibrations (the Mie–Grüneisen thermal pressure), defined as $\Delta P_T(V) = P_T(V) - P_0(V)$, amounts to ~1.1 kbar and is practically independent of volume (it decreases slightly with decreasing volume).

In conclusion, the authors thank L. Z. Ponizovskiĭ for help with the experiment, and O. K. Revyakin and V. M. Blinov for supplying the steels with optimal structure.

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Positive-muon depolarization in weakly doped silicon single crystals

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Results are reported of experiments devoted to a study of the depolarization of positive muons in silicon single crystals of both types of conductivity. The existence in silicon of two paramagnetic states and one diamagnetic state that include muons is confirmed. The dependence of the relative fraction of the diamagnetic state on the temperature is investigated. The results are discussed under the assumption that a chemical bond exists between the muon and the crystal lattice of the semiconductor.

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1. INTRODUCTION

The investigation of the interactions of positive muons with the crystal lattice of a semiconductor (the most thoroughly investigated were silicon and germanium) has led to the observation of many phenomeno hitherto not revealed by the method of observing the muon spin precession (rotation) (μSR) for other media. These include the noticeable decrease of the frequency of the hyperfine splitting of muonium atoms imbedded in the crystal lattice of a semiconductor, compared with the vacuum value,[1-3] as well as the presence in silicon, simultaneously with the isotropic "normal" muonium, of a muon + electron system with a substantially smaller hyperfine-interaction constant, having a clarly pronounced anisotropy with respect to the symmetry axes of the single crystal.[4] The obtained preliminary data point also to a probable existence of an analogous system—"anomalous" muonium—ingermanium.[5] Thus, positive muons can be situated in semiconductors in three states that manifest themselves differently in experiment. Following the established terminology, we henceforth designate by μ^+ the state when the muonium is an a dynamic environment, i.e., it precesses in a magnetic field perpendicular to the initial direction of the particle spin at the meson precession frequency $\omega_\mu = g_\mu H$, where $g_\mu \approx 8.5 \times 10$ $\sec^{-1}G^{-1}$ is the gyromagnetic ratio of the muon and H is the field intensity. The second state (Mu) is "isotropic" muonium, which precesses in the magnetic field at the muonium precession frequency $\omega_{\mathrm{Mu}} \approx 103\ \omega_\mu$. The frequency $\omega_0 = (1980 \pm 90)$ MHz of the ω_{Mu} hyperfine splitting in silicon was measured[3] by observing two-frequency precession of the muonium. The third state (Mu*) is anomalous muonium, whose energy-level structure was investigated in detail in Refs. 6 and 7. The frequencies of the hyperfine interaction in this system can be directly observed in experiment and were investigated in Ref. 4.

Further progress towards the construction of the real scheme of the interaction of positive muons with semiconductors necessitates not only a determination of the physical characteristics of each of the states, but also a study of the possible transitions between them. For example, for muonium in germanium it was found, by measuring the phase shift of the mesic precession, that Mu $\to \mu^+$ transitions exist, with a rate that depends exponentially on the sample temperature and with an energy-barrier height (0.18 ± 0.02) eV.[8] In silicon, the dependence of the relative fraction of each of the states on the temperature and concentration of the doping impurity was investigated in Ref. 9. Three temperatures were used, 25, 80, and 295 K, and it was established that the states Mu and Mu* are observed at low temperatures but not at room temperature. In this paper we consider the results of experiments with silicon samples whose temperature was varied smoothly with an aim and finding and investigating in greater detail those temperature regions in which changes occur between the state amplitudes.

2. EXPERIMENTAL DATA

The experiments were performed with the separated beam of positive muons of the meson channel of the synchrocyclotron of the Leningrad Institute of Nuclear Physics. Muons with momenta ~100 MeV/c were stopped in the investigated silicon sample, which was placed in a thermal bath. A multichannel analyzer was used to study the distribution of the times of the μe decays at 0° to the beam in a perpendicular or longitudinal external magnetic field. The block diagram of the recording apparatus was similar to that used by us earlier.[10]

The main characteristics of the experiments were the following: the stopped-muon counting rate was up to ~10^4 $\sec^{-1}$, the initial beam polarization was ~0.85, the maximum luminosity of the decay registration was ~10%, the width of the range curve at half height was ~3.5 g/cm², the range of employed analyzer channel widths was 1.2–1.8 nsec, the resolution time was ±1.5 nsec, the intensity of the external magnetic fields produced by two pairs of Helmholtz coils was up to 500 G, and the temperature was kept constant in the target working volume within ±2°.

The experimentally obtained distributions of the μe-decay times were reduced with a computer to determine the characteristics of the muon depolarization process. The search was by the maximum likelihood method in accord with the expression

$$N_i(t) = N_0 e^{-t/\tau}[1 + a(R) P_0 e^{-\lambda t} \cos(\omega t + \delta)] + \Phi, \qquad (1)$$

where $N_i(t)$ are the numbers of counts in the i-th channel of the analyzer, and depend on the time, i.e., on the channel number; N_0 is a factor determined by the summary statistics of the experiment; the first exponential factor takes into account the muon decay; τ is the muon lifetime; $a(R)$ is the asymmetry coefficient of the μe decay and depends weakly on the effective target thickness R; in our set of experiments this coefficient was determined by control experiments with graphite targets of varying thickness ($a \approx 0.29$ at $R = 3.0$ g/cm²); P_0 is the polarization extrapolated to the instant of stopping of the muon in the target; λ is the rate of depolarization of the muons in the course of the observation; ω and δ are respectively the frequency and initial phase of the spin precession; Φ is the random-coincidence background ($\Phi/N_0 < 1\%$).

The program of the experiments with silicon was aimed at searching and studying, at various temperatures, the following precession characteristics of the three states Mu, Mu* and μ^+: the relative fraction of the polarized mesons in each state; the rates of the particle depolarization that occurs during the observation time. Since the earlier studies[9,11] precessions with frequencies corresponding to paramagnetic Mu and Mu* states were observed in semiconductors with impurity carrier densities on the order of 10^{12}–10^{14} cm^{-3} and were not observed in materials with higher impurity concentration, we used in our experiments samples of weakly doped silicon. The targets were mosaics made up of equally oriented single crystals, whose electric conductivity and Hall constant were measured beforehand. The sample with n-type conductivity (n-sample) had a conduction-electron density in the range (1.3–1.8) $\times 10^{13}$ cm^{-3} (for individual single crystals), which depended little on the temperature in the interval from 78 K to the start of the intrinsic conductivity, and with a symmetry axis [111] oriented along the beam direction. The p-type sample had a hole density (5–8)$\times 10^{12}$ cm^{-3}, which also depended weakly on the temperature, and the principal diagonal of the crystal coincided with the direction of the intensity vector perpendicular to the beam of the external magnetic field. Measurements at a given temperature were made at several values of field intensity and several analyzer channel widths, since the muonium (Mu and Mu*) and the mesic precessions differ substantially in frequency and in relaxation rate. The muon-

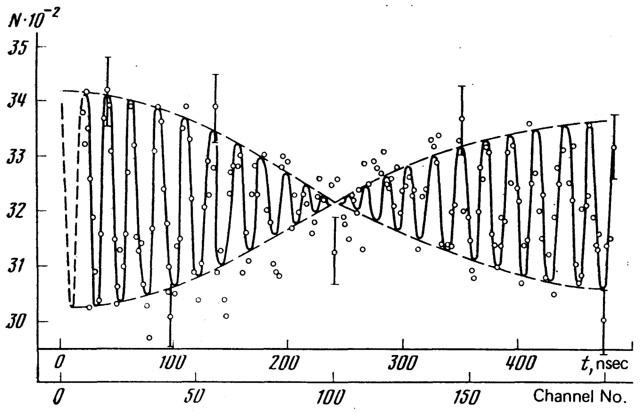

FIG. 1. Two-frequency precession of muonium in *p*-silicon. The abscissas are the time and the analyzer channel numbers, while the ordinates are the counts in the channel, corrected to allow for the background of the random coincidences and for the exponential decay of the mesons. The curves were drawn in accord with the results of the computer reduction of the spectrum. The sample temperature is 78 K.

ium precessions were investigated in fields of intensity 32, 47 and 157 G, while the mesic precession was observed in fields 120–470 G. In the latter case, the field intensity was varied with an aim at finding the shift produced in the initial precession phase by transitions between paramagnetic and diamagnetic states.[8] Experiments were also performed at a temperature of 78 K and at a number of temperatures between 120 and 720 K.

Figure 1 shows a picture of two-frequency precession of "normal" muonium in *p*-Si at 78 K and a perpendicular-field intensity 32 G. The calculated precession parameters are the following: the initial amplitude corresponds to a polarization P (Mu) = 0.26 ± 0.03; the relaxation time $\tau(\mathrm{Mu}) > 600$ nsec, apparently the largest value observed to date in silicon; the hyperfine splitting frequency $\omega_0 = 1910 \pm 60$ MHz agrees well with the results of Refs. 1 and 3.

Under the same conditions, but in a field of 157 G, we observed two-frequency precession with a carrier frequency $\omega = 44 \pm 1$ MHz and a modulation frequency $\Omega = 2.6 \pm 0.5$ MHz, due to the Mu* state (the precession frequency of "normal" muonium in such a field is ~220 MHz), for which $P_0(\mathrm{Mu}^*) = 0.37 \pm 0.04$. The depolarization of the muons contained in the anomalous muonium is faster than in the ordinary triplet state of muonium, $\tau(\mathrm{Mu}^*) = 240 \pm 50$ nsec. In the *n*-type sample at 78 K, we observed only one precession frequency, corresponding to the Mu state. The parameters measured in a field 32 G were $P_0(\mathrm{Mu}) = 0.49 \pm 0.19$ and $\tau(\mathrm{Mu}) = 80 \pm 30$ nsec. It is obvious that in view of the high relaxation rate of the precession, in the latter case we measured not the frequency doublet but the mean value. With increasing temperature, the relaxation times of the normal and anomalous muonium decrease. At a temperature above 170 K no precession with muonium frequencies were observed in either *n*-Si or *p*-Si. The maximum relaxation rate corresponding to the experimental limit of the observation of the precession is in the foregoing experiments of the order of $(3-5)\times 10^7$ sec^{-1}.

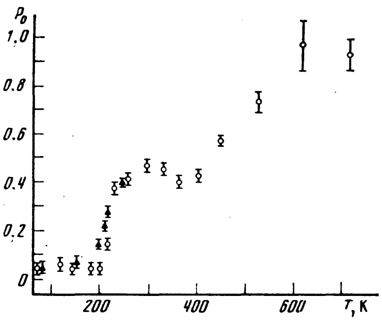

FIG. 2. Temperature dependence of the residual polarization of muons in silicon. Abscissas—sample temperature, ordinates—polarization. The measurements were made in a perpendicular magnetic field, and the precession with mesic frequency was investigated. The light circles and dark triangles correspond to measurements with *n*- and *p*-samples, respectively.

We consider now the experimental results obtained in the investigation of the precessions with mesic frequency (Figs. 2 and 3). Some of the data pertaining to high temperatures were taken from our earlier paper.[12] The character of the change of the polarization $P_0(\mu^+)$ with increasing temperature allows us to single out four regions:

I. 78–200 K; the polarization values are approximately constant; $P_0 \approx \mathrm{const} = 0.05$;

II. 200–250 K; a sharp increase of the polarization to a level $P_0 \approx 0.42$;

III. 250–430 K; $P_0 \approx \mathrm{const} = 0.42$;

IV. $T > 430$ K; further increase of the polarization, at a rate slower than in region (II), with P_0 approaching unity.

The increase in the fraction of the mesic component of the polarization in the regions (II) and (IV) occurs apparently as a result of the presence of $\mathrm{Mu}^* \rightarrow \mu^+$ and $\mathrm{Mu} \rightarrow \mu^+$ transitions, whose probability depends substantially on the silicon temperature and does not depend (in region II) on the type of conductivity of the single crystals. The admixture of the unaccounted-for muon depolarization channels is determined by the relation

$$P(x) = 1 - 2P_0(\mathrm{Mu}) - P_0(\mathrm{Mu}^*) - P_0(\mu^+), \tag{2}$$

since the employed magnetic fields are weak for Mu and

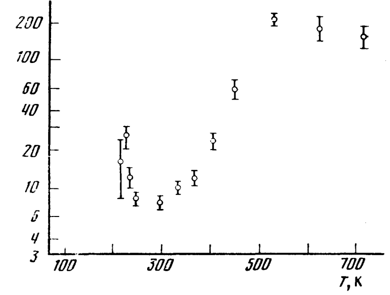

FIG. 3. Dependence of the relaxation rate of precession with mesic frequency in *n*-Si on the temperature. Abscissas—temperature, ordinates—relaxation rate in logarithmic scale. No values of λ below 200 K are available because of the smallness of P_0 (see Fig. 2).

strong for Mu* (Refs. 3, 4); $P(x) = 0.06 \pm 0.10$ for the p-sample.

Figure 3 shows the dependence of the rate of relaxation of the mesic precession on the temperature for n silicon. The positions of the extrema on the figure correspond to the maximum rates of change of the polarization on Fig. 2. For p-silicon no relaxation was observed ($\lambda < 1 \times 10^5$ sec^{-1}). It is known that slow relaxation of the muon spin can be due to the interaction of the magnetic moments of the muon and of the surrounding nuclei.[13] However, in our experiments the contribution of these interactions to the relaxation processes is neglibible. This conclusion follows, first, from a comparison of the data for the n- and p-samples, in which the relaxation rates differ greatly. Second, the measured relaxation rates at the extrema of the function $\lambda(T)$ greatly exceed the analogous values for media in which there is no electronic paramagnetism.[13,14] Therefore the main cause of the relaxation of the mesic precession in silicon must be taken to be the instability of the produced diamagnetic states of the muon; the transitions $\mathrm{Mu} \rightleftharpoons \mu^+$ and $\mathrm{Mu}^* \rightleftharpoons \mu^+$ are reversible, and the rates of the reverse transitions depends both on the temperature and on the state of the conduction band of the semiconductor.

3. DISCUSSION OF RESULTS

We consider now one of the possible schemes of the interaction of positive muons with the crystal lattice of silicon, a scheme based on formation of a chemical bond between the muon and one of the lattice atoms $[\mu^+e^-e^-\mathrm{Si}^+]$, and which makes it possible to explain satisfactorily, in our opinion, most of the experimental results reported above. Since the magnetic moments of the bond-producing electrons are mutually compensated, the muon precesses in the external magnetic field like a free particle (μ^+ state). The need for introducing a chemical bond in the interaction scheme is dictated by two considerations. First, the material presented above indicates that the $\mathrm{Mu} \to \mu^+$ transition in silicon is a thermal process, i.e., the energy barrier for the considered transition is comparable with kT. On the basis of an analysis of experimental data, a similar conclusion was drawn in Ref. 8 for the nearest analog of silicon-germanium. Second, the measured frequency of the hyperfine interaction in silicon (and germanium) indicates that the ionization potential of muonium in semiconductors, on the contrary, greatly exceeds kT, as confirmed by model calculations.[15]

The formation of the system $[\mu^+e^-e^-\mathrm{Si}^+]$ in which the silicon atom is bound, naturally, also to three neighboring lattice atoms can be represented as the result of the transfer, into the conduction band, of an electron from a local donor level produced when the lattice is perturbed by the presence of the muonium atom in the interstice. The appearance of the chemical bond of the muon with the lattice causes one of the valent bonds of another (neighboring) atom to be broken. The silicon atom, deprived of one of its valence bonds has apparently an electron affinity comparable in order of magnitude with the ionization potential of the impurity elements of group V of the periodic system, i.e., it is a "shallow" donor. The suggested picture of the interaction of muons with silicon can be represented for the sake of clarity in the form of the schemes

$$\mu^+ + e^- \text{ (from the valence band)} \to \mathrm{Mu}$$

$$\mathrm{Mu} + [\mathrm{Si}^+e^-e^-\mathrm{Si}^+] \to [\mu^+e^-e^-\mathrm{Si}^+]\mathrm{Si}^+ + e^- \text{ (to the conduction band)}$$

$$[\mu^+e^-e^-\mathrm{Si}^+]\mathrm{Si}^+ + e^- \text{ (from the valence band)} \rightleftharpoons [\mu^+e^-e^-\mathrm{Si}^+](\mathrm{Si}^+e^-).$$

The process (a) is the production of muonium and is energywise always possible if the ionization potential of muonium exceeds the semiconductor band gap. The process (b) is interpreted as the $\mathrm{Mu} \to \mu^+$ transition; the activation barrier in germanium is 0.18 ± 0.02 eV.[8] The process (c) corresponds to the transitions $\mu^+ \rightleftharpoons \mathrm{Mu}^*$, i.e., the symbol Mu* stands for a system in which the electron is localized on one of the lattice atoms that is adjacent to the muon. It is impossible to predict beforehand which of the quantities, the binding energy of the localized electron (c) or the height of the barrier in process (b), is higher, but experiment shows that it is precisely the transitions $\mathrm{Mu}^* \to \mu^+$ which cause the increase of the polarization $P_0(\mu^+)$ in the temperature region 200–250 K, while the $\mathrm{Mu} \to \mu^+$ transitions cause the increase at > 430 K (see Fig. 2). The depolarization of muons in strong longitudinal magnetic field were investigated in Ref. 1 for the same sample as in the present study. The measurements were made at 300–330 K, i.e., in region (III), and have shown that the growth of the initial polarization, which proceeds with an increase of the field intensity, corresponds to normal muonium. This means that the $\mathrm{Mu}^* \to \mu^+$ transition takes place in the region of lower temperatures.

The model considered here does not explain the relations between the initial populations of each of the states, relations determined in many respects by the conditions for the restoration of the radiation damage in the crystal at the end of the muon track. Partial information on these processes can apparently be obtained from a comparison of the experimental data for silicon and germanium, although for the latter there are still no detailed data on the anomalous muonium. However, the transition $\mathrm{Mu} \to \mu^+$ in germanium has been investigated in sufficient detail.[8] It takes place in the temperature region 170–250 K, i.e., at lower temperatures than in silicon. One can expect that the $\mathrm{Mu}^* \to \mu^+$ transition, if it does occur in germanium, should do so at a temperature lower than 78 K. An indirect confirmation of this assumption is contained in Ref. 5, where anomalous precession frequencies were observed at 10 K.

The described mechanism of interactions of muons with silicon is, of course, not the only one possible. As noted above, observation of a muonium-like system with an anisotropic hyperfine interaction has stimulated attempts to find an explanation of this phenomenon. A detailed analysis of the influence of the crystal-field anisotropy of single crystals of the diamond type on the properties of the muonium produced in a crystal lattice is contained in Ref. 6, where the existence of two types of muonium atoms is considered. One type is located in the tetrapore of the crystal (Mu), and the other (Mu*) in the octapore. The conclusion drawn in this paper concerning the dependence of the anomalous frequencies on the intensity of the external magnetic field and the mutual orientation of the field-intensity vector and the crystal symmetry axes agree with experimental results.[4] The factors, however, that determine the number of muons that enter in a particular state, and the dependence of the relative fraction of each state on the temperature, have been analyzed to a much lesser degree. The mathematical formalism developed in Ref. 6 for "anisotropic" muonium can apparently be applied also to the proposed model since, first, the wave functions of the electrons of the impurity atoms in silicon and germanium are anisotropic,[16] and second, the muon in the system $[\mu^+e^-e^-\text{Si}^+](\text{Si}^+e^-)$ is displaced away from the center of the orbit of the electron localized on the neighboring silicon atom. The advantage of the considered model over the anisotropic muonium lies, in our opinion, in the fact that the explanation of the existence of a mesic component of the polarization and of its temperature dependence does not require the introduction of any additional assumptions.

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