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# Shallow acceptor centres in silicon studied by means of spin rotation of negative muons

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**Abstract.** The residual polarization of negative muons has been studied for phosphorus-doped ([P]:  $1.6 \times 10^{13}$  cm<sup>-3</sup>) and antimony-doped ([Sb]:  $2 \times 10^{18}$  cm<sup>-3</sup>) silicon crystals. The measurements were carried out in a transverse magnetic field of 0.1 T over the temperature region 4 K–300 K. The ionized and neutral states of the  $\mu$  Al pseudo-acceptor were observed in antimony-doped silicon for the first time. The rate of transition from the neutral to the ionized state of the acceptor was found to be equal to  $1.2 \times 10^{6}$  s<sup>-1</sup> over the temperature range 4 K–12 K. The estimated rates of relaxation of the magnetic moment of the acceptor-centre electron shell are  $5 \times 10^{10}$  s<sup>-1</sup> and  $1.6 \times 10^{12}$  s<sup>-1</sup> in phosphorus-doped silicon and  $6 \times 10^{11}$  s<sup>-1</sup> and  $6.7 \times 10^{12}$  s<sup>-1</sup> in antimony-doped silicon at 4 K and 15 K respectively. The experimental results obtained are interpreted in terms of spin–lattice relaxation of the acceptor magnetic moment and of the acceptor–donor pair formation.

#### 1. Introduction

From the standpoint of Coulomb interaction the negative muon  $\mu^-$  is an analogue of the electron. The muon possess a spin of 1/2, its mass is about 207 times larger than that of the electron and its electric charge is equal to the electron charge. The muon is an unstable particle decaying via the mode  $\mu^- \rightarrow e^- + \nu_{\mu} + \bar{\nu}_e$ . The mean lifetime of free muons is  $\tau_{\mu}^{\text{free}} \approx 2.2 \times 10^{-6} \text{ s.}$ 

Beams of polarized muons with kinetic energy up to 125 MeV are produced at proton accelerators in several scientific centres of the world. Interaction of accelerated protons with nuclei results in  $\pi$ -mesons. The mean lifetime of  $\pi$ -mesons is shorter than that of the muon and the main decay channel is  $\pi^- \rightarrow \mu^- + \bar{\nu}_{\mu}$ . Due to nonconservation of space parity, muons that arise from the decay of  $\pi$ -mesons are longitudinally polarized and the spatial distribution of electrons from the  $\mu^-$  decay, with respect to the  $\mu^-$  spin, is asymmetrical. In muon spin-rotation ( $\mu^-$ SR) experiments, the decay asymmetry is the measured quantity that can be obtained from the count rates of the electron counters.

In condensed matter, negative muons with the energy of tens of MeV lose the initial kinetic energy within  $\sim 10^{-11}$  s and are captured to highly excited levels by matter atoms due to Coulomb interaction. Then, after a cascade of radiative and Auger transitions, the muon reaches the 1S state at the muonic atom. The muon polarization remains practically unchanged during the slowing down. However, substantial loss of polarization takes place at the cascade

transitions. As the muon reaches the 1S state, its polarization is only 15–20% of the initial one [1], which is still large enough for solid-state spin-rotation experiments.

Since the muon is heavier than the electron, its orbital radius in the 1S state of the muonic atom is 207 times smaller than that of the K electron. The muon effectively screens the nuclear charge for the electrons by one unit; therefore the muonic atom can be regarded as an atom consisting of a pseudonucleus  $(Z + \mu^{-})$  and Z - 1 electrons.

Variation in the polarization of the muon during its lifetime can be experimentally followed by detecting the time-dependent decay asymmetry and thus interaction between the magnetic moment of the muon and the electron shell of the atom (hyperfine interaction), and interaction between the electron shell and the matter can be investigated.

Since, apart from the decay, the muon can be captured by the nucleus from the 1S state, and the capture rate depends on the nuclear charge, the mean muon lifetimes in the 1S state of different atoms are appreciably different, varying from  $\sim$ 2200 ns for hydrogen to 70 ns for heavy nuclei [2].

The muonic atom resulting from the capture of a negative muon by the silicon atom is an analogue of the aluminium atom. The experimental data known today (see for example [3]) do not contradict the assumption that in condensed matter capture of a negative muon by a host atom does not lead to its displacement from the node of the crystal lattice. An aluminium atom in a node of a silicon crystal lattice is a shallow acceptor centre. Thus, investigating the behaviour of the polarization of the negative muon, one can obtain information on the interaction of isolated acceptor centres with the matter.

The temperature dependence of the relaxation rate and the precession-frequency shift of negative muon spin in n- and p-type silicon at temperatures below 30 K were observed in previous  $\mu^{-}$ SR experiments [4–6].

The relaxation of the muon spin-precession signal in diamagnetic silicon was explained by paramagnetism of the electron shell of the muonic atom formed as a result of negative muon capture by silicon atoms.

In contrast to that of donor impurities, the behaviours of acceptors in semiconductors with diamond crystal structure (silicon, germanium, diamond) have been studied only incompletely by traditional methods (EPR, ENDOR etc). The study of shallow acceptor centres is limited by the large spin–lattice relaxation rate and random internal crystal strains. Therefore, attempts to observe the EPR signal of acceptor centres in these semiconductors were unsuccessful [7–9].

The first observation of the EPR signal from the boron atom in a uniaxially stressed silicon crystal was reported in 1960 [7]. To the best of our knowledge, only one paper was published, in which EPR spectra of boron atoms in silicon crystals in the absence of applied external stress were reported [10]. As was noted by the author of that paper, some phenomena had no reasonable explanations: (i) the temperature dependence (in the range from 4.2 K to 1.2 K) of the 'fine' structure of the EPR spectra; (ii) decrease of the signal amplitude with decreasing thickness of the samples; and (iii) twofold broadening of the line after finely grinding the samples.

The present paper contains the results on the behaviour of the residual polarization of negative muons in silicon doped with phosphorus and antimony, as well as in graphite and germanium. The impurity concentrations in the phosphorus- and antimony-doped silicon samples were  $[P] = 1.6 \times 10^{13} \text{ cm}^{-3}$  and  $[Sb] = 2 \times 10^{18} \text{ cm}^{-3}$ , respectively. To clarify the effect of exchange scattering of charge carriers on the relaxation of the magnetic moment of the acceptor centre, silicon samples with low  $(1.6 \times 10^{13} \text{ cm}^{-3})$  and high  $(2 \times 10^{18} \text{ cm}^{-3})$  concentrations of n-type impurity were investigated. (As follows from the temperature dependence of the concentration of free charge carriers in silicon with arsenic impurity [11], a concentration of such an order does not lead to the predominance of metal-like conductivity.)

A graphite sample served as a reference to determine the muon-beam polarization and the set-up parameters. The measurements on germanium were carried out to verify the total loss of negative muon polarization observed in [12] under unfavourable conditions. Germanium, like silicon, has diamond crystal structure and investigation of the behaviour of the Ga acceptor in germanium is interesting from the same point of view as that of the Al acceptor in silicon.

#### 2. Experimental set-up

The measurements were performed on the 'Stuttgart LFQ Spectrometer' [13] in the  $\mu$ E4 decay channel beamline of the Paul Scherrer Institut (PSI, Villigen, Switzerland). The magnetic field of 0.1 T was produced by Helmholtz coils transverse to muon spin polarization. The sample thickness along the beam direction was 2.6, 3.8, 2.4, 2.1 g cm<sup>-2</sup> for the phosphorus-doped silicon, antimony-doped silicon, graphite and germanium samples, respectively. The samples had approximately equal areas of about 7 cm<sup>2</sup>. To cool the sample, vapour flow of cold helium was used. The sample temperature was stabilized with an accuracy of 0.1 K. The spectrometer time bin was 0.625 ns; the total number of channels in the spectrum was 16 000. Electrons from stopped muons were registered by counters placed in front of and behind the sample. Thus, two spectra from the decay electrons, moving forward (FW) and backward (BW) relative to the muon beam direction, were stored simultaneously. The muon stop rate in the sample was about 10<sup>4</sup> per second. The beam momentum adjusted for the best signal-to-background ratio was 60 MeV/*c* in chromatic mode.

## 3. Results

Besides stopping in the sample, some of muons stop in the cryostat walls and in the scintillation counters and contribute to the spectrum as copper and carbon components respectively. Since the negative muon lifetime in the 1S state depends on the nuclear charge, the time distribution of electrons from the  $\mu^- \rightarrow e^- + \nu_{\mu} + \bar{\nu}_e$  decay can be written as

$$f(t) = \sum_{X} N_{X} e^{-t/\tau_{X}} \left[ 1 + \alpha p_{X}(t) \right] + B(t)$$
(1)

where  $N_X$ ,  $\tau_X$ ,  $p_X$  are respectively the count rate at t = 0, the muon lifetime and the projection of the polarization onto the observation direction for the muon captured by the atom X;  $\alpha$  is the asymmetry coefficient in the space distribution of decay electrons assuming the detection solid angle; *B* is the background from accidental coincidences.

From the experimental data one can find the quantity  $a_X(t) = \alpha p_X(t)$ , where  $\alpha$  is a constant. This means that the time dependences of  $a_X(t)$  and of  $p_X(t)$  are the same.

The background was modulated with the frequency  $v_{hf} = 50.6328$  MHz due to the proton beam microstructure. It was found from the preliminary Fourier analysis that the spectra contained higher harmonics of  $v_{hf}$  as well. As a result, *B* was considered as ( $\omega_{hf} = 2\pi v_{hf}$ )

$$B = b_0 + \sum_{n=1}^{2} b_n \cos(n\omega_{\rm hf}t + \phi_n).$$
(2)

The experimental data were fitted by the weighted least-squares method. The negative muon lifetimes were kept constant at the mean values of the experimental data given in [2] (2030 ns, 760 ns, 167 ns and 163 ns for carbon, silicon, germanium and copper, respectively).

# 3.1. Copper

To determine  $a_{Cu}(t)$  a measurement with a copper sample was carried out. This experiment was necessary for correct consideration of the copper contribution in the processing of the experimental data for different samples. The spectra contained two components with the negative muon lifetimes  $\tau_C$  in carbon and  $\tau_{Cu}$  in copper. Since copper atoms possess nonzero nuclear spin, there is additional depolarization of the muon caused by hyperfine interaction between the magnetic moments of the nucleus and muon [14, 15]. Analysis of the data shows that  $a_{\pm}$ , the amplitudes of the muon spin precession at frequencies  $\omega_{\pm} = \gamma_{\pm}H$  corresponding to the hyperfine states with total angular momentum  $F_{\pm} = I \pm S_{\mu}$ , are equal to zero within experimental errors:  $a_{\pm} = (0.2 \pm 0.3)\%$ . The present result agrees with earlier data [16, 17].

From the experimental data on copper and germanium the polarization function  $a_{\rm C}(t)$  corresponding to muons stopped in the scintillation counters was obtained. The contribution from the counters depends on the sample thickness and appears to be 7%, 4% and 1.5% for copper, silicon and germanium samples, respectively (in the 'forward' spectrum). The contribution is well described by the function  $a_{\rm C}(t) = a_0 \cos(\omega t + \phi)$ , where  $a_0 = 0.009 \pm 0.002$  and  $\omega$  is the precession frequency of the muon spin in the external magnetic field. For further analysis it was supposed that  $a_{\rm Cu}(t) = 0$  and  $a_{\rm C}(t) = 0.009 \cos(\omega t + \phi)$ .

#### 3.2. Graphite

In the processing of the graphite sample data, the copper (the cryostat walls) contribution was taken into account. It was supposed that  $a_{\rm C}(t) = a_0 \cos(\omega t + \phi)$  for graphite.

For the graphite sample, measurements were performed at temperatures 4 K, 20 K and 300 K. It was found that the function of polarization  $a_{\rm C}(t)$  and the precession frequency  $\omega_{\rm C}$  do not depend on temperature in the range 4 K–300 K. The averaged values of  $a_{\rm C}(t = 0)$  and  $\omega_{\rm C}$  are respectively  $(4.16 \pm 0.07) \times 10^{-2}$  and  $(85.113 \pm 0.008)$  rad  $\mu {\rm s}^{-1}$ . For a single measurement  $\sigma(\omega_{\rm C})/\omega_{\rm C}$ , where  $\sigma(\omega_{\rm C})$  is one standard deviation of  $\omega_{\rm C}$ , is approximately equal to  $2 \times 10^{-4}$ . The deviation from the averaged value is less than the errors for the single measurements. This means that the precision of  $\omega_{\rm C}$  is limited by the statistics. Therefore, the stability of the external magnetic field was not worse than  $2 \times 10^{-4}$  and the spectrometer allows one to obtain the muon spin-precession frequency with the same accuracy.

#### 3.3. Germanium

The negative muon lifetimes in germanium and copper are close to each other. For this reason the contributions from the sample and the cryostat walls were described by a single exponential term in (1), the lifetime being considered as a free parameter.

Figure 1 shows an example of the  $\mu^{-}$ SR spectrum for the germanium sample at 300 K. The precession frequency is equal to the free-muon spin-precession frequency within errors and corresponds to the peak on the Fourier image of the spectrum.

It was assumed that muons captured by copper atoms are totally depolarized within a short time that is inaccessible to observation and that the fraction of muons stopped in the cryostat walls depends only on the effective stopping thickness of the sample. To determine the dependence of the copper contribution on the sample thickness, the corresponding measurement on silicon were carried out.

After correction to the copper contribution,  $a_{\text{Ge}}(t = 0)$  in germanium was found to be  $(2.2 \pm 0.2)\%$ ,  $(2.4 \pm 0.2)\%$ ,  $(2.3 \pm 0.2)\%$  and  $(2.7 \pm 0.2)\%$  at 4.5 K, 30 K, 100 K and 290 K, respectively.



Figure 1. An example of the  $\mu^{-}$ SR spectrum for the germanium sample after background subtraction and correction for the negative muon lifetime at T = 300 K. The lower picture is its Fourier image.

Systematic errors, which arise from the correction for the copper contribution, were not taken into account. It follows from the present data that muon polarization in the 1S state of germanium is only 30–40% less than in carbon. Probably, there is a slight decrease in the polarization with decreasing temperature. Muon spin relaxation in germanium within errors was not observed. Averaged over 4.5 K, 30 K, 100 K and 300 K, the muon spin-precession frequency for the germanium sample is  $(84.9 \pm 0.4)$  rad  $\mu s^{-1}$ .

#### 3.4. Phosphorus-doped silicon

The  $\mu$ SR spectrum of silicon contains contributions from copper (the cryostat walls), silicon and carbon (the scintillation counters). Accordingly, the function describing the experimental data, besides the background, contains three terms. Considering muon spin relaxation at low temperatures, the polarization function for silicon was expressed as (here and further on the

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subscript 'Si' in the formulae for a(t) is omitted for simplicity)

$$a(t) = a_0 e^{-\lambda t} \cos(\omega t + \phi)$$
(3)

where  $\lambda$  is the muon spin-relaxation rate. The functions for the copper and carbon contributions were presented above.

Examples of  $\mu^{-}$ SR spectra for the phosphorus-doped silicon sample are shown in figure 2. The background, copper and carbon contributions are subtracted. For visual presentation the data are multiplied by  $\exp(t/\tau_{\rm Si})$ . It is clearly seen that muon spin relaxation takes place at low temperatures.



Figure 2. The phosphorus-doped silicon sample.  $\mu^{-}$ SR spectra after background subtraction and correction for the negative muon lifetime.

Figure 3 gives the temperature dependence of the relaxation rate and precession-frequency shift  $\delta\omega/\omega_0$ , where  $\omega_0$  is the precession angular frequency at room temperature. The temperature dependence of the relaxation rate is well approximated by the function  $\lambda = dT^{-q}$ , where  $d = (4.0 \pm 0.7) \times 10^3 \ \mu \text{s}^{-1}$ ,  $q = 2.73 \pm 0.06$  and the temperature is given in K. In the present study the value of the parameter q was determined four times more precisely than in previous work [5, 6]. The data given in figure 3 confirm the conclusion [6] that at T < 50 K there is a shift of the muon spin-precession frequency. The uncertainty of the  $\delta\omega/\omega_0$  measurement is half of that of the previous experiment [6].

#### 3.5. Antimony-doped silicon

Preliminary data processing showed that function (3) was not suitable for describing the lowtemperature experimental data for the antimony-doped silicon sample. After correction for



**Figure 3.** The phosphorus-doped silicon sample. The temperature dependence of the relaxation rate (top) and the frequency shift (bottom).

the muon lifetime the presence of both damped and undamped polarization components in the spectra is evident (see figure 4). The term 'undamped' means that the possible relaxation rate does not exceed 0.05  $\mu$ s<sup>-1</sup>.

Therefore, for further data processing the polarization function a(t) was taken as

$$a(t) = a_1 e^{-\lambda t} \cos(\omega_1 t + \phi_1) + a_2 \cos(\omega_2 t + \phi_2).$$
(4)

The behaviour of negative muon-polarization in this sample differs considerably from that in the other silicon samples investigated with different impurity types and concentrations [4–6]. The function describing muon spin polarization contains both damped and undamped components. It was found that the sum of the amplitudes of the components  $(a_1 + a_2)$  is independent of temperature within the experimental errors and equal to the muon spin-precession amplitude observed at room temperature. The temperature dependence of  $a_1$  and  $a_2$  is shown in figure 5.

The amplitude of the damped component  $(a_1)$  at 4.1 K is about 85% of the sum. With increasing temperature,  $a_1$  decreases and at 17 K  $a_1 \approx a_2$ . The muon spin-precession frequencies for the damped and undamped components differ from each other. For the undamped component it corresponds to the free-muon spin-precession frequency, whereas for the damped component a frequency shift is observed at temperatures lower than 20 K. The mean value of the frequency shift  $\delta\omega/\omega_0$  in the temperature range 8–20 K is  $(8.0 \pm 2.2) \times 10^{-3}$ .



Figure 4. The antimony-doped silicon sample. The  $\mu^-SR$  spectrum at 6 K in different time windows.



Figure 5. The antimony-doped silicon sample. The temperature dependence of the relative amplitudes of the damped  $(a_1)$  and undamped  $(a_2)$  components of the negative muon polarization  $(a_1 + a_2 = 1).$ 

# 4. Discussion

Observation of the damped and undamped components of the residual polarization of negative muons in the antimony-doped Si indicates that during the observation time the acceptor centre

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can be found in both ionized  ${}_{\mu}Al^{-}$  and neutral  ${}_{\mu}Al^{0}$  states. Therefore, it is necessary to assume that either muonic atoms are originally (for a time shorter than  $1/\Omega_{hf}$ ) formed in both ionized and neutral states or there is a transition between the states with a rate comparable to  $1/\tau_{si}$ .

From the *x*- and *y*-components of the muon polarization one can define a complex variable according to  $p = p_x + ip_y$ . It is evident that the modulus of  $p_1 (p_2)$  in the paramagnetic (diamagnetic) state of the acceptor decreases (increases) by  $v_{12}p_1 dt$  in time dt via the transition  $(1 \rightarrow 2)$  from the paramagnetic to the diamagnetic state with the rate  $v_{12}$ . At the same time  $p_1 (p_2)$  increases (decreases) by  $v_{21}p_2 dt$  via the  $2 \rightarrow 1$  transition with the rate  $v_{21}$ . In the paramagnetic state 1 there is a loss of muon polarization by  $\lambda p_1 dt$  due to the interaction between the magnetic moment of the muon and the magnetic moment of the electron shell of the acceptor. In the external magnetic field parallel to the *z*-axis the time development of *p* can be described by the following system of differential equations:

$$\frac{d}{dt}p_1 = (i\omega_1 - \lambda - \nu_{12})p_1 + \nu_{21}p_2$$

$$\frac{d}{dt}p_2 = \nu_{12}p_1 + (i\omega_2 - \nu_{21})p_2$$
(5)

where indices 1 and 2 denote the neutral (paramagnetic) and the ionized (diamagnetic) states of the acceptor centre, respectively. The terms  $i\omega_1$  and  $i\omega_2$  describe rotation of the polarization vectors  $p_1$  and  $p_2$  in the external magnetic field transverse to the *xy*-plane. The experimentally observed value is  $p_x = \text{Re } p$ , where *p* is the total polarization  $p_1 + p_2$ .

The solution of the set of equations (5) in the general case (without limitations on the values of the parameters  $v_{12}$  and  $v_{21}$ ) will be discussed in a separate paper. Here we confine ourselves to considering solutions of the set of equations (5) for  $v_{21} = 0$ , which leads to the polarization function of form (4). It should be mentioned that the polarization function, which is a solution of (5), for  $v_{21} \neq 0$  does not involve the undamped component.

Under the initial conditions p(0) = 1 and in the absence of the transition  $2 \rightarrow 1$  ( $\nu_{21} = 0$ ) the solution of the set of equations (5) has the form

$$p_{x}(t) = C_{1} e^{-(\lambda+\nu)t} \cos(\omega_{1}t+\varphi_{1}) + C_{2} \cos(\omega_{2}t+\varphi_{2})$$

$$C_{1} = \frac{n_{1}\sqrt{\lambda^{2}+\delta^{2}}}{\sqrt{(\lambda+\nu)^{2}+\delta^{2}}} \qquad C_{2} = \frac{\sqrt{(n_{2}\lambda+\nu)^{2}+(n_{2}\delta)^{2}}}{\sqrt{(\lambda+\nu)^{2}+\delta^{2}}} \qquad (6)$$

$$\tan\varphi_{1} = -\frac{\nu\delta}{\lambda(\lambda+\nu)+\delta^{2}} \qquad \tan\varphi_{2} = \frac{n_{1}\nu\delta}{(n_{2}\lambda+\nu)(\lambda+\nu)+n_{2}\delta^{2}}$$

where  $n_1$  and  $n_2$  are the initial populations of states 1 and 2 ( $n_1 + n_2 = 1$ ),  $\delta = \omega_1 - \omega_2$ ,  $\nu = \nu_{12}$ .

As is evident from (6), the polarization function will have two terms (damped and undamped) in two cases: (a) in the absence of the transition  $1 \rightarrow 2$  ( $\nu = \nu_{12} = 0$ ,  $n_1 \neq 0$  and  $n_2 \neq 0$ ); (b) for  $\nu \neq 0$  and  $n_1 \neq 0$ , including the case with  $n_1 = 1$  ( $n_2 = 0$ ). Obviously, at  $n_1 = 0$ , function (6) will consist of the undamped term alone for all  $\nu$ .

The kinetic calculation of the muonic atom formation [18] shows that in silicon the muonic atom is formed in the neutral state  ${}_{\mu}Al^0$  in  $10^{-9}$  s, which corresponds to the case with  $n_2 = 0$ .

The results of the fit to the experimental data for the antimony-doped silicon sample using function (6) (under the condition  $n_2 = 0$  and  $\delta = 0$  in the expressions for  $C_1$  and  $C_2$ ) are shown in figure 6. As is seen, the transition rate  $\nu$  is practically constant in the temperature range 4 K–12 K and approximately equal to  $1.2 \times 10^6$  s<sup>-1</sup>. Above 13 K the transition rate decreases with increasing temperature.

Observation of the undamped component in the residual polarization of negative muons for the antimony-doped  $(2 \times 10^{18} \text{ cm}^{-3})$  silicon sample at 4 K may be qualitatively understood



Figure 6. The temperature dependence of the muon spin relaxation  $\lambda$  and transition  $(1 \rightarrow 2)$  rates  $\nu$  for the antimony-doped silicon sample.

on the basis of the known mechanism of acceptor–donor pair formation in a semiconductor with n- and p-type impurities [19–21]. It was found that at concentrations of about  $10^{16}$  cm<sup>-3</sup> of donor and acceptor impurities in silicon, acceptor–donor pairs are formed. The ground state of this system is  $A^-D^+$  ( $A^-$  and  $D^+$  are the ionized acceptor and donor respectively). When the distance between an acceptor and a donor is larger than 50 Å the process of acceptor–donor pair transition from the state  $A^0D^0$  to the ground state is well described within the framework of the model developed in [19].

Unfortunately, a quantitative comparison of the present experimental data with the calculations by the means of the model [19] is not possible, because in our case an average distance between a neutral Sb donor and a neutral  $_{\mu}$ Al acceptor, formed via  $\mu^{-}$  capture, is about 50 Å. In this case in approximately 17% of events the wave functions of the acceptor and the donor overlap and the consideration given in [19] no longer holds.

Formation of the  $A^-D^+$  pair within a time comparable to or shorter than the mean lifetime of a negative muon in silicon would result in an undamped component of the residual polarization of the muon. Observation of the undamped component for the sample with antimony (2×10<sup>18</sup> cm<sup>-3</sup>) impurity and its absence in the silicon samples doped with phosphorus (1.6 × 10<sup>13</sup> cm<sup>-3</sup>) and with boron (2 × 10<sup>13</sup> cm<sup>-3</sup>) [4] do not contradict the idea of acceptor–donor pair formation in the first sample. Further experiments on n- and p-type silicon with different concentrations of impurities will make it possible to prove the suggested hypothesis.

Muon spin relaxation at a frequency close to that of free-muon spin is due to relaxation of the magnetic moment of the electron shell of the muonic atom, which is the acceptor impurity in semiconductors. In the hydrogen-like atom approximation, which is often used for describing an acceptor centre + hole system, the relaxation rate of the muon spin can be expressed as [22]

$$\lambda = \Omega_{\rm hf}^2 / 4\nu_{\rm r} \tag{7}$$

where  $\nu_r$  is the relaxation rate of the magnetic moment of the electron shell of the acceptor centre,  $\Omega_{hf}$  is the angular frequency of hyperfine interaction between the magnetic moments of the muon and the electron shell of the acceptor centre.

Using the value  $\Omega_{\rm hf}/2\pi = 650$  MHz from [23] and the value of  $\lambda$  obtained for the phosphorus-doped silicon, we found that  $\nu_{\rm r} \approx 1.6 \times 10^{12} \, {\rm s}^{-1}$  at T = 15 K. Extrapolating according to the dependence  $\lambda = dT^{-q}$  to 4 K, the value of  $\lambda$  leads to  $\nu_{\rm r} \approx 5 \times 10^{10} \, {\rm s}^{-1}$ . For the antimony-doped sample the value of  $\nu_{\rm r}$  was estimated as  $6.7 \times 10^{12} \, {\rm s}^{-1}$  and  $6 \times 10^{11} \, {\rm s}^{-1}$ 

at 15 K and 4 K respectively. The estimated values of  $v_r$  do not contradict the results of the EPR measurements carried out on a silicon crystal under uniaxial stress [8].

According to the consideration given in [22] the temperature dependence of the frequency shift of the muon spin precession can be expressed as

$$\frac{\delta\omega}{\omega_0} \sim \frac{g_{\rm eff}}{2} \frac{m_\mu}{m_e} \frac{\hbar\Omega_{\rm hf}}{T} \sim \frac{1}{T}$$
(8)

where  $g_{\text{eff}}$  is the Landé factor for the acceptor,  $m_{\mu}$  and  $m_e$  are the muon and the electron mass, respectively.

The data on  $\delta\omega/\omega_0$  obtained for the phosphorus-doped silicon sample do not contradict the 1/T dependence. Unfortunately, formula (8) gives only the type of the dependence of the frequency shift on temperature and determination of the hyperfine frequency on the basis of (8) is not possible.

In thermodynamical equilibrium in n-type silicon, where the concentration of donors substantially exceeds the concentration of acceptors, in the temperature region considered, the acceptor centres are ionized with high probability. The fastest process which results in the ionization of the acceptor at low temperatures is the radiative recombination of the hole trapped on the acceptor with the electron trapped on the donor (see for example [19, 20]), the rate in silicon being not higher than  $10^5 \text{ s}^{-1}$  at 4.2 K at concentrations of the dominant donor impurity up to  $10^{17} \text{ cm}^{-3}$  [19]. The observation of the frequency shift of the muon spin precession (see figure 3) is evidence that in the phosphorus-doped silicon sample the probability of the transition from the neutral to the ionized state of the  $_{\mu}$ Al pseudo-acceptor within the muon lifetime is negligible at temperatures of 50 K and below.

Relaxation of the magnetic moment of the acceptor centre in a semiconductor can be due to: (a) the exchange scattering of charge carriers on the acceptor and (b) spin–lattice interaction. Theoretical calculations [24–26] and experimental investigations on EPR of shallow acceptors in silicon under external uniaxial stress [7,8] show that the high relaxation rate of the magnetic moment the acceptor centre at low temperatures is due to spin–lattice interaction and that the contribution of the exchange scattering process to the relaxation is negligible.

The results of the present experiment also confirm the dominant role of the phonon process. These are: (a) rather weak dependence of the relaxation rate on temperature (much weaker than the dependence of the free-charge-carrier concentration on temperature); (b) weak dependence of the relaxation rate on the impurity concentration (the concentration in the phosphorusand antimony-doped samples differs by five orders of magnitude, while the difference in the relaxation rate is by a factor of about three).

The high spin-lattice relaxation rate of the magnetic moment of the acceptor in silicon is due to the structure of the valence band of the silicon crystal. The band structure is such that in an undeformed crystal the ground energy level of an acceptor is fourfold degenerate (see for example [27]).

As shown in [25, 26], the following processes can give a contribution to spin–lattice relaxation of shallow acceptor centres in silicon: the direct (one-phonon) process, the Raman process and the resonance fluorescence (Orbach) process. As follows from [25, 26], the relaxation rate of the magnetic moment of the acceptor is dependent on the external conditions. Thus in an undeformed silicon crystal in external magnetic field H = 30 kOe the Raman scattering of phonons dominates in the temperature range 10 K–100 K and the relaxation rate of the magnetic moment of the acceptor is proportional to  $T^5$  [26]. If a strong uniaxial stress is applied to the crystal, the Orbach process becomes the dominant one at temperatures higher than 13 K with a temperature dependence of the relaxation rate like  $v_r \sim \exp(-\Delta/T)$ . As is shown in [22], taking into account the random strains in an undeformed crystal may result in a power dependence of the relaxation rate on temperature different to  $T^5$ . At temperatures

higher then 10 K a dependence like  $v_r \sim T^2$  is possible. Thus the difference in the dependence of the relaxation rate of the muon spin on temperature in this experiment  $\lambda \sim v_r^{-1} \sim T^{-q}$  $(q = 2.73 \pm 0.06$  for the phosphorus-doped sample) from the  $T^5$ -dependence of  $v_r$  for an ideal crystal [26] is probably due to the random crystal strains.

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