Depolarization mechanisms for muonium in germanium

A. Weidinger, G. Balzer,* H. Graf, E. Recknagel, and Th. Wichert
Physics Department, University of Konstanz, 7750 Konstanz, West Germany
(Received 10 June 1981; revised manuscript received 28 August 1981)

The depolarization rate λ of normal and anomalous muonium in Ge was studied as a function of temperature and doping. Evidence for three different depolarization mechanisms is presented: (i) exchange interaction with paramagnetic doping atoms ($T < 20$ K); (ii) Korringa scattering of impurity charge carriers ($20 < T < 50$ K); and (iii) an intrinsic process, either a chemical reaction or a spin-lattice relaxation ($T > 50$ K).

Muonium is a hydrogenlike atom consisting of a positive muon and a bond electron. In Ge, as in Si, two such states are known[1–4]: normal muonium (μ^+e^-) with an isotropic and anomalous muonium ($\mu^+e^-)^*$ with an anisotropic hyperfine interaction. The muon spin rotation (μSR) data received considerable interest since muonium can be considered as a light isotope of hydrogen. Surprisingly, very little is known about hydrogen in Si and Ge, in particular, no NMR or ESR signals were observed which could be attributed to hydrogen in defect-free surroundings in Si or Ge. On the other hand, in a theoretical paper on the hydrogen interstitial impurity in germanium[5] a 1s-like deep donor state was found for hydrogen in Ge. The authors suggest that this deep donor state is the one probed by μ^+SR experiments, but that pure paramagnetic hydrogen is not observed since H migrates to impurities or defects or forms molecular hydrogen rather than remaining as an isolated interstitial impurity. In this context the muon may play an important role since the interaction of a bare positive charge with the crystal lattice can apparently not be studied in hydrogen experiments. So far, most μSR work in this field was concerned with the hyperfine interaction of muonium, but recently the depolarization rate gained considerable interest also.[4,6–9] It was found that this quantity depends strongly on various external parameters, but no comprehensive data set was presented yet. The present paper gives a systematic survey of the relaxation rate for Ge with the intention to clear up the relaxation mechanisms.

The experiment was performed at the Swiss Institute of Nuclear Research (SIN) in a transverse magnetic field. The muon spin precession and relaxation was measured via the anisotropic decay of the polarized muons. (For a general review see Ref. 10.) The bare muon and the two muonium states were distinguished by their characteristic precession frequencies. The undoped Ge crystal used in the experiment had an electrically active impurity concentration of less than 1.5×10^{10} cm^{-3}. The other samples were doped either with Sb (n type) or Ga (p type). In all cases the magnetic field was applied parallel to a crystallographic $\langle 110 \rangle$ direction. The field strengths were 0.006 T for normal, and 0.3 and 0.4 T for anomalous muonium, respectively.

Below 80 K the amplitudes of the different muon or muonium states in Ge are roughly independent of temperature and doping of the crystals. The largest fraction of stopped muons (60–70%) forms normal muonium; the rest goes into anomalous muonium and the diamagnetic muon state.

Figure 1 shows the spin depolarization rate λ of muons in the normal muonium state (μ^+e^-). The undoped crystal exhibits a smooth and continuous increase of λ with temperature. The data are excellent-

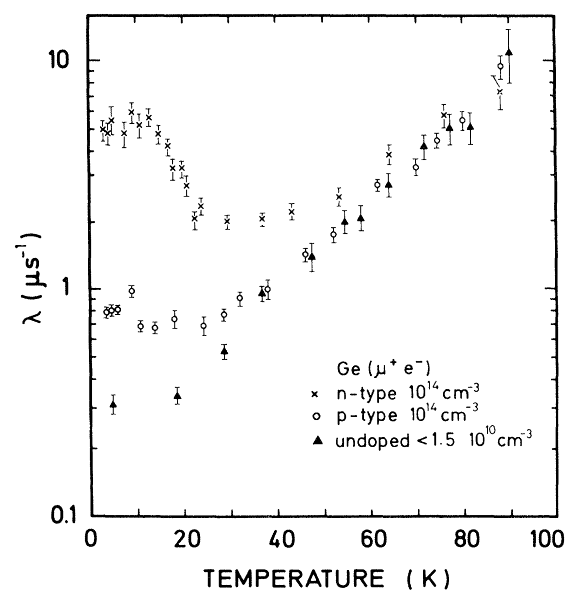

FIG. 1. Depolarization rate λ or muons for normal muonium (μ^+e^-) in Ge as a function of temperature. An undoped, a p-type (Ga) and a n-type (Sb) sample were investigated.

ly reproduced by an exponential temperature dependence. However, in the analysis only data above 30 K will be considered since the lowest λ values ($\lambda \approx 0.3\ \mu s^{-1}$) might be affected by inhomogeneities of the external field. In contrast to the undoped crystal, the doped samples show a strongly structured T dependence of λ. They all converge to the data of the undoped crystal above 50 K but at lower temperatures drastic differences are present. For the n-type crystal in Fig. 1, λ passes through a minimum around 30 K and increases to a plateau of $\lambda = 5.3\ \mu s^{-1}$ below 12 K. The depolarization rate for the p-type sample falls between the two other curves. Experiments on more heavily doped p-type samples show the same behavior of λ as the n-type sample in Fig. 1. The drop of λ between 12 and 20 K coincides with the ionization of the dopant atoms.

Figure 2 shows the doping dependence of the depolarization at 4.3 and 30 K. These temperatures are characteristic for the plateau and minimum region in Fig. 1, respectively. The strong increase of λ with the doping concentration and the large difference between p- and n-type doping are obvious from Fig. 2. The straight lines in Fig. 2 were obtained from a fit to the depolarization rates after subtracting the λ values of the undoped sample ($\lambda = 0.35\ \mu s^{-1}$ at 4.3 K and $\lambda = 0.55\ \mu s^{-1}$ at 30 K). The fit yields $\lambda \sim c^{0.7 \pm 0.2}$ at 4.3 K and $\lambda \sim c^{0.95 \pm 0.20}$ at 30 K, where c is the dopant concentration. The functional dependence is the same for n- and p-type crystals, but the strength differs by a factor of 30.

The depolarization rate λ of anomalous muonium (Fig. 3) shows a quite different behavior. The most striking feature is the almost zero depolarization below 12 K for all three samples. Between 12 and 20 K a strong increase of λ occurs for the two doped samples and above 20 K the $(\mu^+e^-)^*$ signal disappears completely. For the undoped sample no change of λ is observed up to 70 K. Above this temperature the signal becomes very weak either due to a decrease of the amplitude or due to an increase of λ. No distinction between these two effects was possible.

On the basis of the presented data three temperature regions can be distinguished in which different depolarization mechanisms are effective.

Region I ($T < 12$ K). The decrease of λ between 12 and 20 K for normal muonium strongly suggests that the depolarization below 12 K is connected with the *paramagnetic* state of the dopant atoms. We propose an interaction of the type: $H_1 = -2J_{12}\vec{S}_1 \cdot \vec{S}_2$ where $\vec{S}_1$ is the spin of the unpaired electron of the dopant, $\vec{S}_2$ is the spin of the muonium electron, and J_{12} is the exchange interaction of the two electrons. The large Bohr radius of shallow donors and acceptors [e.g., $a_0 \approx 80$ Å for Sb in Ge (Ref. 11)] causes a nonvanishing overlap of the dopant electron with the

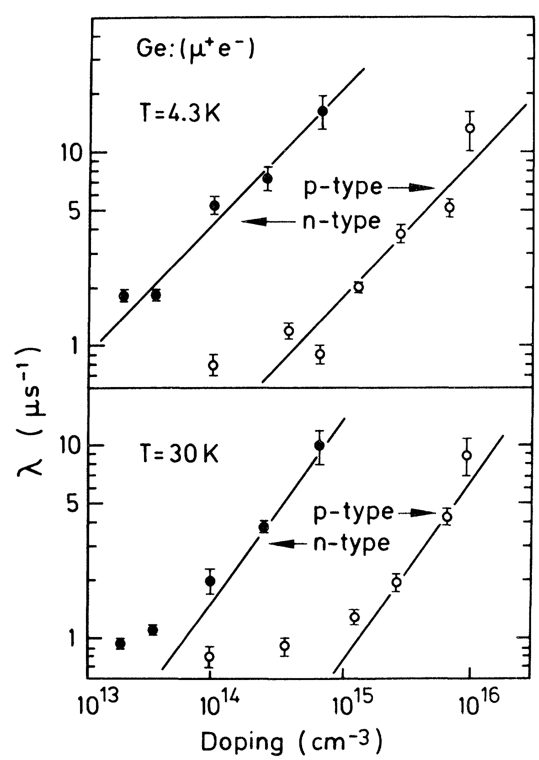

FIG. 2. Depolarization rate λ of muons for normal muonium (μ^+e^-) in Ge as function of the dopant concentration c. The straight lines ($\lambda \sim c^{0.7}$ in the upper and $\lambda \sim c^{0.95}$ in the lower part of the figure) correspond to a fit to the corrected data for which the λ values of the undoped sample are subtracted.

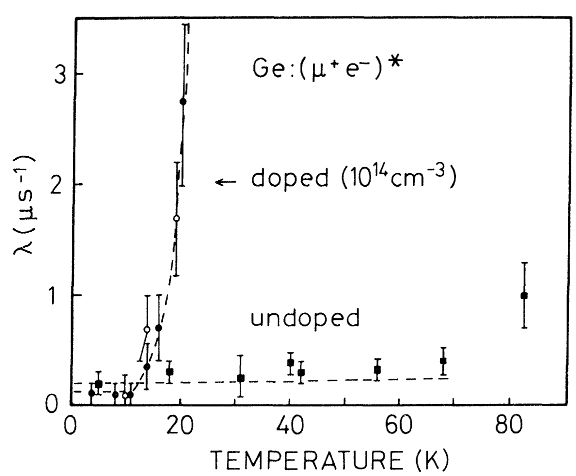

FIG. 3. Depolarization rate λ of muons for anomalous muonium $(\mu^+e^-)^*$ in Ge as function of temperature. Different symbols are used for the different samples (○, Ga-doped, ●, Sb-doped, and □, undoped). The applied magnetic field was 0.4 T below 40 K and 0.3 T above 40 K.

muonium even at rather low doping concentrations. A rough estimate of the exchange integral can be obtained by assuming 1*s*-like wave functions with $a_0 = 80$ Å for the donor and $a_0 = 0.53$ Å for muonium. In this way we obtain $J_{12}/\hbar \approx 1$ μs^{-1} assuming a separation of 500 Å. This value shows the right order of magnitude, although more detailed calculations are necessary for a quantitative comparison. Strong support for the proposed depolarization mechanism is provided by the different behavior of (μ^+e^-) and $(\mu^+e^-)^*$. The difference is apparently due to the fact that (μ^+e^-) is observed in the Zeeman region whereas $(\mu^+e^-)^*$—because of the smaller hyperfine constant—is measured in the Paschen-Back region. In the strong field limit the muon and electron spins are decoupled and an interaction of the type H_1 has no effect on the muon spin in agreement with the small depolarization rate observed for $(\mu^+e^-)^*$. In the Zeeman region, which is the relevant case for normal muonium, the muon and electron spin are strongly coupled and the interaction H_1 causes a splitting of the μSR frequencies by $\Delta\omega = J_{12}/\hbar$. Because of the variation of the distance between muonium and the dopant, not a splitting but only a distribution of frequencies is predicted. The expected depolarization rate is $\lambda \approx \Delta\omega = J_{12}/\hbar$, where J_{12} is an average value. The observed differences between *p*- and *n*-type doping is probably due to the different local densities of the unpaired electrons from donors and acceptors, respectively, at the muonium site. It is expected that donor electrons have a larger overlap with muonium at an interstitial site than unpaired electrons from an acceptor. Thus a larger exchange integral with muonium for donors than for acceptors is predicted.

Region II $(20 < T < 50\ K)$. In this temperature region practically all dopants are ionized in Ge, whereas intrinsic electrons or holes are still not present. Thus the number of charge carriers is essentially constant and equal to the dopant concentration *c*. Moreover, the spins of the electrons in the conduction band flip very rapidly so that the paramagnetic depolarization is reduced. This effect explains the decrease of λ when the dopants are ionized around 15 K. We suggest that the depolarization in region II is governed by the spin exchange scattering of conduction electrons (or holes) with the muonium electron (this process is called Korringa relaxation in analogy to the nuclear relaxation by conduction electrons[12]). The model predicts $\lambda \sim c$ (*c*, the doping concentration) in excellent agreement with the data. The predicted temperature dependence[13] $(\lambda \sim \sqrt{T})$ is consistent with the experimental results although the data are not very precise in this respect. In contrast to the static paramagnetic depolariztion (T_2 process) discussed for region I, the electron spin flip described here (T_1 process) causes a relaxation for normal as well as for anomalous muonium.[14] The effect on $(\mu^+e^-)^*$ is observed experimentally to be even stronger. This might be explained by a larger spatial extension of $(\mu^+e^-)^*$ compared to (μ^+e^-).

Region III $(T > 50,\ T > 30\ K$ *for undoped Ge).* Two different processes were considered for the doping independent depolarization: (i) A chemical reaction: $(\mu^+e^-) \rightarrow \mu^+$ (diamagnetic) and (ii) a spin-lattice relaxation (Raman process). Within the accuracy of the present data no distinction can be made between these two processes on the basis of the temperature dependence. The analysis of the (μ^+e^-) data with an Arrhenius function yields an activation energy of $E_a = 12 \pm 3$ meV. No microscopic models can be offered at present.

CONCLUSION

There is strong evidence that the depolarization at low temperature $(T < 12$ K) is caused by a spin-spin interaction between the muonium electron and the paramagnetic dopants. The arguments for this assignment are (i) strong doping dependence of λ, (ii) no effect on anomalous muonium, and (iii) abrupt change of λ when the dopants are ionized. The depolarization in region II $(20 < T < 50$ K) is attributed to a Korringa process and in region III $(T > 50$ K) to a chemical reaction or to spin-lattice relaxation. No detailed models were developed for the latter mechanisms.

ACKNOWLEDGMENTS

The financial support of the Bundesminister für Forschung und Technologie is gratefully acknowledged. We thank Mr. A. Möslang for the help in the experimental work.

---

*Present address: Physics Department, University of Dortmund, 4600 Dortmund, Germany.

[1] I. I. Gurevich, I. G. Ivanter, E. A. Meleshko, B. A. Nikol'skii, V. S. Roganov, V. I. Selivanov, V. P. Smilga, B. A. Sokolov, and V. D. Shestakov, Zh. Eksp. Teor. Fiz. 60, 471 (1971) [Sov. Phys. JETP 33, 253 (1971)].

[2] J. H. Brewer, K. M. Crowe, F. N. Gygax, R. F. Johnson, B. Patterson, D. G. Fleming, and A. Schenck, Phys. Rev. Lett. 31, 143 (1973).

[3] B. D. Patterson, W. Kündig, P. F. Meier, F. Waldner, H. Graf, E. Recknagel, A. Weidinger, Th. Wichert, and A. Hintermann, Phys. Rev. Lett. 40, 1347 (1978).

[4]E. Holzschuh, H. Graf, E. Recknagel, A. Weidinger, Th. Wichert, and P. F. Meier, Phys. Rev. B 20, 4391 (1979).
[5]W. E. Pickett, M. L. Cohen, and C. Kittel, Phys. Rev. B 20, 5050 (1979).
[6]C. Bucci, R. De Renzi, G. Guidi, P. Podini, R. Tedeschi, and L. O. Norlin, Hyper. Inter. 8, 385 (1981).
[7]C. Boekema, E. Holzschuh, W. Küdig, P. F. Meier, B. D. Patterson, W. Reichart, and K. Rüegg, Hyper. Inter. 8, 401 (1981).
[8]C. W. Clawson, E. E. Haller, K. M. Crowe, S. S. Rosenblum, and J. H. Brewer, Hyper. Inter. 8, 417 (1981).
[9]K. W. Blazey, J. A. Brown, D. W. Cooke, S. A. Dodds, T. L. Estle, R. H. Heffner, M. Leon, and D. A. Vanderwater, Phys. Rev. B 23, 5316 (1981).
[10]J. H. Brewer, K. M. Crowe, F. N. Gygax, and A. Schenck, in *Muon Physics,* edited by V. W. Hughes and C. S. Wu (Academic, New York, 1975), Vol. III.
[11]C. Kittel, *Introduction to Solid State Physics,* 5th ed. (Wiley, New York, 1976), p. 233.
[12]J. Kondo, in *Solid State Physics,* edited by F. Seitz, D. Turnbull and H. Ehrenreich (Academic, New York, 1969), Vol. 23, p. 183.
[13]A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961), p. 390.
[14]I. G. Ivanter and V. P. Smilga, Zh. Eksp. Teor. Fiz. 54, 559 (1968) [Sov. Phys. JETP 27, 301 (1968)].