μ^- SR IN SEMICONDUCTORS

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μ^- SR experiments have been performed on Si between room temperature and 6 K. The amplitude of the muon spin precession signal in an applied magnetic field of 0.04 T decreased below 30 K. A zero-field measurement at 6 K revealed a μ^- spin precession frequency of 650 MHz. The muonic atom represents an aluminium acceptor in the silicon matrix, its electronic state is responsible for the μ SR signal. A possible influence of the γ recoil produced by the X-ray cascade is discussed.

1. Introduction and general background

During the past two decades μ^+ SR has become a well-established technique in solid-state physics and chemistry. After the positive muons have been slowed down in the sample they behave similarly to hydrogen nuclei and occupy interstitial sites in the crystal lattice. In metals the μ^+ allow us to investigate local magnetic fields, diffusion of light particles with unit positive charge, and low concentrations of lattice defects. In semiconductors and insulators the study of two types of muonium (Mu, Mu*) has led to a better understanding of the behaviour of hydrogen in these materials.

The behaviour of *negative muons* implanted into condensed matter is radically different from that of μ^+ [1,2]. The electrostatic attraction between the nuclei and the μ^- leads to the incorporation of the μ^- in 1s atomic orbits. Owing to the large mass of the μ^- – which in this context may be considered as “heavy electrons” – the spatial extent of the muonic 1s orbital is very small compared with that of the electronic 1s orbitals. This means that in its ground state a “muonic atom” of charge number Z appears to the electronic system like an “ordinary atom” of charge number $Z - 1$.

The present paper is concerned with the $Z - 1$ effect in semiconductors. In the case of Si ($Z = 14$) capture of μ^- in 1s orbitals leads to the formation of quasi-aluminium ($Z = 13$). The particular interest of this is that the introduction of the quasi-Al impurity takes place almost suddenly and that its effects on the electronic system may be studied on the μ s time scale.

Fig. 1. Characteristic muonic X-rays as measured with a 4×1 inch NaI scintillator. For charge numbers $Z < 20$ the energy resolution is good enough to distinguish between different elements. The energy spectrum shows a large X-ray line at 76 keV, emitted by the muonic transition from $n = 3$ to $n = 2$ in Si and a smaller peak at 403 keV due to the transition $n = 2$ to $n = 1$. The peaks at 34 keV and 171 keV are caused by muons stopped in the beryllium moderator and by the capture of muons at fluorine in the BaF_2 stop detector, respectively.

When an incoming μ^- has been slowed down to kinetic energies in the eV range, the formation of a “muonic atom” starts with the capture of a μ^- in a high-lying excited muonic orbital. This process is accompanied by the ejection of Auger electrons. Subsequently, the μ^- cascades down to its 1s orbital, strongly disturbing the electronic shells. Depolarization of the μ^- takes place due to spin-orbit coupling and due to spin-flip transitions during the cascade [3]. Typically only about 18% of the initial polarization remains when the muon has reached its ground state [4,5]. The entire rearrangement process takes place in a time that is short compared with the typical μ SR timescale. Above $Z \approx 7$, the lifetime of μ^- in muonic atoms decreases rapidly with increasing Z , thus giving a signature of the element involved. In Si ($Z = 14$) the lifetime is $8 \cdot 10^{-7}$ s [6]. Another signature is provided by characteristic X-rays emitted during the cascade (fig. 1). By means of μ^- SR experiments in coincidence with X-ray measurements the local magnetic fields near the nuclei of each element in alloys and compounds (“X-ray triggered μ^- SR”) may be measured [9].

In a semiconductor, the replacement of a lattice atom of charge number Z by a muonic atom of effective charge number $Z - 1$ leads to the introduction of an

acceptor. The acceptor level close to the edge of the valence band ($E_A = 57$ meV for Al in Si) disappears again when the captured muon disappears. During the μ^- lifetime the acceptor level may be occupied by an electron. This process can be followed quantitatively by observing the interaction between the magnetic moment of the unpaired electron and that of the μ^- by means of μ^- SR.

We must keep in mind, however, that there is no guarantee that the muonic atom remains on its lattice site. About 20 eV are necessary to produce a Frenkel pair by irradiation. It is true that the γ -recoil energy does not suffice to displace the atoms permanently in a perfect environment. (In Si, the emission of a 411 keV photon results in a recoil energy of 3.1 eV.) This may be different during the transit of the μ^- to its 1s orbital when the electronic structure and hence the bounding of the atom is strongly disturbed.

2. Experimental techniques

The experiments were performed on three different silicon crystals between room temperature and 6 K, viz n-doped Si (Si n 1: $2.5 \cdot 10^{14} \text{ cm}^{-3}$ phosphorus), p-doped Si (Si p 1: $1.4 \cdot 10^{14} \text{ cm}^{-3}$ boron), and intrinsic high purity Si (Si int: room temperature resistivity $2 \cdot 10^3 \Omega \text{ cm}$). Natural Si possesses a 95% abundance of isotopes with zero nuclear magnetic moment, so depolarization due to dipolar interaction is negligible.

Two types of experiments were performed using the μE1 beam at the Paul Scherrer Institut (Villigen, Switzerland), viz μ^- SR experiments in an applied field $B_{\text{appl}} = 40$ mT and zero-field measurements. For the latter an apparatus with high time resolution was constructed. We used pilot-U plastic scintillators coupled directly to XP-2020 phototubes. By means of constant-fraction discriminators acting on the fast-rising signal of a dynode a time resolution of 130 ps (FWHM) was achieved for prompt events from beam electrons.

3. Results and discussion

In each run of the measurements with $B_{\text{appl}} = 0.04$ T more than $6 \cdot 10^7$ events were accumulated in the four electron-counter telescopes surrounding the sample. The count rates measured in telescope no. i were fitted to

$$N^i(t) = N_1^i \exp(-t/\tau_1) \left[1 + A_1^i e^{-\lambda_1 t} (\cos \omega_1 t + \phi_1^i) \right] \\ + N_2^i \exp(-t/\tau_2) \left[1 + A_2^i e^{-\lambda_2 t} (\cos \omega_2 t + \phi_2^i) \right] + B_s^i (\cos \omega_s t + \phi_s^i) + U^i,$$

where τ_1 denotes the decay time of the μ^- in the sample and A_1^i , λ_1 , ω_1 , and ϕ_1^i are the asymmetry, the damping rate, the circular frequency, and the phase of that signal. The subscript 2 refers to muons stopped in the moderator. B_s^i , ω_s , ϕ_s^i

Fig. 2. μ^- SR signal of n-doped silicon (sample Si n 1, $2.5 \cdot 10^{14} \text{ cm}^{-3}$ phosphorous) in an applied magnetic field of 0.04 T at room temperature (a) and at 6 K (b) after background subtraction and correction for the finite muon lifetime as measured in four electron-counter telescopes surrounding the sample.

Fig. 3. Arrhenius plot of the average asymmetries A_1 measured in an external magnetic field of 0.04 T on samples Si p 1 (p-doped, $1.4 \cdot 10^{14} \text{ cm}^{-3}$ boron), Si n 1 (n-doped, $2.5 \cdot 10^{14} \text{ cm}^{-3}$ phosphorous), and Si int (intrinsic, $2 \cdot 10^3 \Omega \text{ cm}$). The straight line fitted to the data below 30 K corresponds to an activation enthalpy of $\approx 1 \text{ meV}$.

account for the background caused by beam electrons, and U' represents a time-independent background.

Fig. 2 shows the spin precession signal of μ^- in Si as measured in the four counter telescopes at room temperature (a) and 6 K (b) with $B_{\text{appl}} = 0.04 \text{ T}$. The damping rate λ_1 is less than 100 kHz in the entire temperature range. In fig. 3 the temperature dependence of the weighted average A_1 of the asymmetries A_1^i is shown for all experiments with $B_{\text{appl}} = 0.04 \text{ T}$. The type of doping seems to have no significant influence on A_1 . An Arrhenius fit to the data below 30 K (the straight line in fig. 3) yields a very low activation enthalpy of about 1 meV. From the fact that in spite of the decrease of the asymmetry A_1 at temperatures below 30 K no significant change in the damping rate λ_1 is observed we conclude that the process responsible for the variation of A_1 is terminated after less than 10^{-9} s .

At 6 K a first measurement in zero applied field revealed a peak in the Fourier spectrum at 650 MHz (fig. 4). At room temperature no frequency could be resolved. From the hyperfine splitting in EPR measurements on Al-doped silicon we obtain a hyperfine constant of 1.3 GHz [7]. This value, scaled with the ratio between the magnetic moment of the muon and the nuclear moment of aluminium, leads to a hyperfine constant of 3.2 GHz. However, the “nucleus” of the muonic

Fig. 4. Fourier-spectrum of the μ^- SR signal measured in zero magnetic field at 6 K in intrinsic silicon (sample Si int).

quasi-Al is significantly larger than the aluminium nucleus, so the lower frequency observed in our experiment is also reasonable.

The simplest explanation of the variation of A_1 with temperature would be the temperature dependence of the acceptor ionisation, but with the small activation enthalpy deduced from fig. 3 we do not get a reasonable value for the energy of the Al level in the band gap. This is obvious in the case of n-doped Si where the Fermi level is above the aluminium acceptor level at all temperatures. However, there is a substantial difference between μ^- SR at muonic Al acceptors and classical experiments in Al-doped silicon crystals: in contrast to the classical methods, the μ^- SR technique starts to be sensitive immediately after production of the Al impurity.

Al on interstitial sites may be responsible for the temperature dependence of the μ^- SR amplitude. At interstitial sites the Al will have Al^{2+} configuration with an unpaired electron. When the muon cascades down to the ground state, a 411 keV γ -quantum is emitted. Under normal conditions the recoil energy of 3.1 eV would not be sufficient to displace Si atoms from their lattice sites. As most of the outer electrons are removed by the muon capture the threshold energy for displacement is presumably lowered to such an extent that quasi-Al interstitials are produced. From the experiments we deduce that only below 30 K are the Frenkel-pair configurations sufficiently long-lived to give rise to a detectable μ^- signal.

In order to exploit the full potential of the present technique further experiments are necessary, e.g. measurements on more heavily doped Si crystals. Experiments with pairs of aligned γ detectors in order to investigate the anisotropy of the displacement process appear to be particularly promising.

Acknowledgements

The authors are indebted to F.N. Gyax, A. Schenck (both ETH Zürich) and E. Roduner (Universität Zürich) for lending us their μ SR apparatus. We would

like to thank L. Schimmele for useful discussions. The work was supported by the Bundesministerium für Forschung und Technologie, Bonn, Fed. Rep. of Germany, under contract no. 03-SE2STU-0.

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