Low-temperature positron diffusion in GaAs

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Positron diffusion has been measured in undoped and Si doped ($n=2\times10^{18}\,\mathrm{cm}^{-3}$) GaAs in the temperature range 20–290 K using the positron-beam technique. The experimental diffusion length values are strongly influenced by positron trapping at vacancies and negative ions existing in the samples. After subtraction of the trapping effects the diffusion coefficient for free positrons in the GaAs lattice is obtained. The diffusion coefficient is $14\pm2\,\mathrm{cm}^2\,\mathrm{s}^{-1}$ at 20 K and $1.6\pm0.2\,\mathrm{cm}^2\,\mathrm{s}^{-1}$ at 295 K. Below 80 K it follows the $T^{-1/2}$ law due to scattering from acoustic phonons. From 80 to 300 K the diffusion coefficient decreases strongly with increasing temperature meaning that positron scattering from polar-optical phonons is switched on.

I. INTRODUCTION

Positrons in semiconductors are analogous to free carriers. Their transport is described by drift-diffusion equation including annihilation and trapping at defects.[1] Due to strong repulsion between positrons and positive-ion cores the diffusion is limited by scattering from phonons.

The slow-positron beam is a practical tool for experimental investigation of positron diffusion.[2] It has several times been applied to GaAs.[3–5] Saarinen *et al.*[3] found that in liquid-encapsulated Czochralski-grown (LEC) GaAs the diffusion length is strongly reduced below 300 K and they attributed it to positron trapping at Rydberg states around negative ions. According to Soininen *et al.*[4] the diffusion coefficient from 300 to 800 K depends only weakly on temperature due to positron scattering from both acoustic and polar-optical phonons. In the recent experiments of Shan *et al.*[5] the positron diffusion coefficient shows a strong temperature dependence below 300 K and the authors attributed it to optical phonons.

The motivation of this work is to clarify the role of acoustic and polar-optical phonons in positron diffusion. Our experimental data from 20 to 300 K show that positron motion is strongly affected by trapping at native vacancies and negative ions which seem to be unavoidable in GaAs material. After careful subtraction of the trapping effects the diffusion of free positrons can be deduced. The diffusion is limited only by acoustic phonons below 80 K. From 80 to 300 K the diffusion coefficient strongly decreases due to the onset of scattering from polar-optical phonons.

II. EXPERIMENTAL

Two different GaAs materials were used. The Si-doped GaAs sample ($n=2\times10^{18}\,\mathrm{cm}^{-3}$) was grown by molecular-beam epitaxy (MBE) on a (100)-oriented GaAs substrate at 900 K (sample ''*L*'' in Ref. 6). The epilayer was 2 μm thick and electrically fully activated. The undoped LEC GaAs was semi-insulating with a resistivity of $10^8\ \Omega$ cm. As the reference sample for positron annihilation in the bulk lattice we used a Zn-doped LEC GaAs sample ($p=10^{18}\,\mathrm{cm}^{-3}$). The Si-doped sample was measured in the as-received state whereas the undoped and Zn-doped samples were first etched with the H_2SO_4:H_2O_2:H_2O (3:1:1) solution.

Positron annihilation spectra were measured using the low-energy positron beam technique.[2,7] The annihilation radiation was monitored by a high-purity Ge detector with a resolution of 1.4 keV. The shape of the 511-keV annihilation line was described by the conventional low and high electron-momentum parameters *S* and *W*. The *S* parameter was defined as the ratio of annihilation events in the gamma-ray energy range $E_\gamma=511\pm0.95$ keV around the centroid of the peak to the total number of events in the annihilation line. It represents the electron-positron pairs with a longitudinal momentum component of $p_L/m_0c<3.7\times10^{-3}$, where m_0 is the electron mass and *c* the velocity of light. The core annihilation parameter *W* was defined as the fraction of the annihilation events in the tail of the peak, $2.9\,\mathrm{keV}<|E_\gamma-511\,\mathrm{keV}|<7.3\,\mathrm{keV}$ (or $11\times10^{-3}<p_L/m_0c<28\times10^{-3}$). When positrons annihilate at vacancies or surfaces, the *S* parameter increases and the *W* parameter decreases, since a larger fraction of annihilations takes place with low-momentum valence electrons.

The *S* and *W* parameters were measured as functions of the positron energy *E* from 0 to 25 keV at various temperatures between 20 and 290 K. Below 150 K there were experimental difficulties, because the *S* and *W* parameters at low energies became time dependent due to slow adsorption of residual gases on the sample surface. The problem was eliminated by annealing the sample at 150 K before each measured point. Using this procedure we were able to measure reversible $S(E)$ and $W(E)$ curves even at 20 K.

Positron lifetime spectra were also measured in the undoped and Zn-doped GaAs samples between 80 and 290 K. The average positron lifetime τ is more accurate than the Doppler parameter *S* or *W*, but we could measure it only in the two bulk samples.

III. RESULTS ON DIFFUSION LENGTH

The $S(E)$ curves for the Si-doped sample at 20 and 290 K are shown in Fig. 1. We can see that the half-width of the $S(E)$ curve is smaller at 20 than at 290 K. Similar kinds of curves can be drawn for the *W* parameter. In undoped GaAs the width of the $S(E)$ curve is rather independent of temperature.

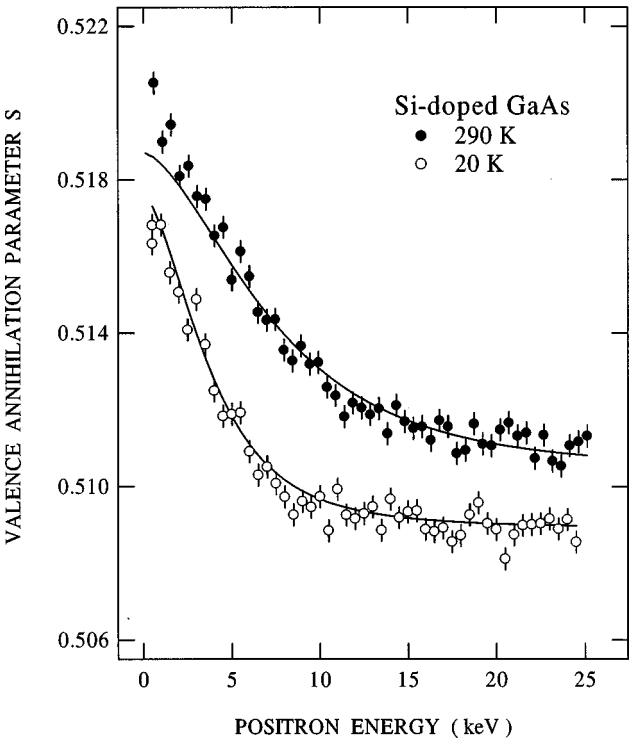

FIG. 1. The low electron-momentum parameter S as a function of positron energy measured at two different temperatures in the Si-doped GaAs layer with $[\mathrm{Si}]=2\times10^{18}\,\mathrm{cm}^{-3}$.

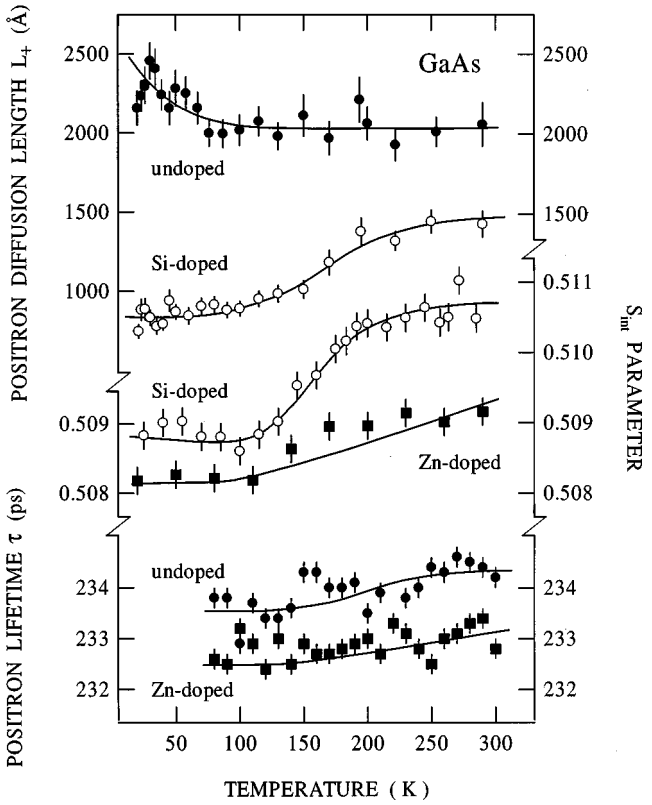

FIG. 2. Positron diffusion length L_+, the S_{int} parameter in the sample interior, and positron lifetime τ in the GaAs samples. The solid lines for $S_{\mathrm{int}}(T)$ and $\tau(T)$ curves are the results of the trapping model fits.

The experimental data were analyzed in the conventional way, see, e.g., Soininen *et al.*[4] After implantation and rapid thermalization, positrons start to diffuse. They annihilate in the sample interior, or diffuse back and annihilate on the entrance surface. The surface was intentionally covered with thin oxide to prevent positron or positronium emission into vacuum. No positron drift effect was taken into account. The high carrier concentration in the Si-doped sample screens out any surface electric field at a distance of 100 Å. In undoped GaAs the Fermi-level pins close to the midgap which together with the very low carrier concentration leads to a negligible electric field.

The fraction of the annihilations on the surface $f(E)$ is determined by the implantation depth profile $P(E,x)$ and the diffusion length L_+. The experimental Doppler parameter $S(E)$ measured as a function of positron incident energy E is a superposition of the different S-parameter values for the annihilations in the interior (S_{int}) and on the sample surface (S_{surf}):

$$S(E)=f(E)S_{\mathrm{surf}}+[1-f(E)]S_{\mathrm{int}}, \tag{1}$$

where

$$f(E)=\int_0^\infty P(E,x)e^{-x/L_+}dx. \tag{2}$$

For the positron implantation profile $P(E,x)$ we used the Makhovian profile with the parameters $m=1.8$, $n=1.60$, and $A_0=70$ Å.[4]

The experimental $S(E)$ curves were fitted to Eq. (1) with three free parameters S_{surf}, S_{int}, and L_+. The energy range $E_{\min}-23\,\mathrm{keV}$ was used, and small energies ($E_{\min}=0,...,3\,\mathrm{keV}$) were skipped due to weak (<10%) positronium formation and epithermal positron emission. The solid lines in Fig. 1 show fitted curves and the fitted values for L_+ are presented in the upper part of Fig. 2.

The diffusion length in undoped GaAs decreases from 2500 to 2000 Å, suggesting reduced phonon scattering for free positrons at low temperatures. In the Si-doped GaAs sample L_+ is only 800 Å at 20 K and increases to 1500 Å at 290 K. The big differences between the samples point out to different amount of defects trapping positrons during diffusion. Indeed the S values in the Si-doped sample and the τ values in the undoped sample are higher than the corresponding values measured in the Zn-doped GaAs sample which according to earlier lifetime experiments[8] serves as the reference for free positron annihilation in the lattice.

The presence of negative ions and vacancies are clearly manifested by the shape of the $S_{\mathrm{int}}(T)$ curve in the Si-doped GaAs.[6] Negative ions are able to bind positrons in competition with vacancies only below 200 K. Above 100 K S_{int} increases when positrons start to escape from the ions and a larger fraction of them annihilate at vacancies. The solid line for the Zn-doped sample represents purely the lattice expansion effects on the free positron annihilation parameter $S_b(T)$.

IV. RESULTS ON DIFFUSION COEFFICIENT

The diffusion coefficient for free positrons is obtained from the diffusion length by[3,7]

$$D_+ = L_+^2/\tau_{\text{eff}}, \tag{3}$$

where the effective lifetime τ_{eff} is the mean residence time of positrons in the freely diffusing state. In the presence of trapping at vacancies (rate κ_v), trapping at shallow Rydberg states around negative ions (rate κ_{ion}), and detrapping from ions (rate δ_{ion}) we have

$$\tau_{\text{eff}}^{-1} = \tau_b^{-1} + \kappa_v + \frac{\kappa_{\text{ion}}}{1+\tau_{\text{ion}}\delta_{\text{ion}}}. \tag{4}$$

Here τ_b and τ_{ion} denote the positron lifetime in the bulk lattice and around the negative ions, respectively. In practice they are equal, i.e., $\tau_{\text{ion}} = \tau_b$.

We analyzed the $S_{\text{int}}(T)$ curve of the Si-doped sample and the $\tau(T)$ curve of the undoped sample using the model, including positron trapping at vacancies and negative ions and detrapping from ions.[6,9,10] In the case of the undoped sample the trapping effect is so weak that it is reliably deduced only from the lifetime data. The annihilation parameter S_{int} (and τ) is a superposition of different annihilation states:

$$S_{\text{int}} = f_b S_b + f_{\text{ion}} S_{\text{ion}} + f_v S_v \tag{5}$$

where the annihilation fractions are

$$f_b = 1 - f_{\text{ion}} - f_v,$$

$$f_{\text{ion}} = \frac{\kappa_{\text{ion}}}{(1+\delta_{\text{ion}}/\lambda_{\text{ion}})\left(\lambda_b + \kappa_v + \dfrac{\kappa_{\text{ion}}}{1+\delta_{\text{ion}}/\lambda_{\text{ion}}}\right)}, \tag{6}$$

$$f_v = \frac{\kappa_v}{\lambda_b + \kappa_v + \dfrac{\kappa_{\text{ion}}}{1+\delta_{\text{ion}}/\lambda_{\text{ion}}}}.$$

Based on our earlier lifetime- and Doppler-broadening results on vacancies in GaAs,[6,10,11] we used $S_v = 1.015 S_b$ and for the ions $S_{\text{ion}} = S_b$. The positron trapping rate at a defect is proportional to the defect concentration, i.e., $\kappa_v = \mu_v c_v$ and $\kappa_{\text{ion}} = \mu_{\text{ion}} c_{\text{ion}}$. The trapping coefficient μ_v is T-dependent [Eq. (3) in Ref. 10] and has a value of $1.4\times10^{15}\,\text{at}\,\text{s}^{-1}$ at 300 K. For ions $\mu_{\text{ion}} = 4.5\times10^{16}\,(\text{T/K})^{-1/2}\,\text{at}\,\text{s}^{-1}$ was used. The detrapping rate from ions is $\delta_{\text{ion}} \propto \exp(-E_b/kT)$ where $E_b \approx 40$ meV is the positron binding energy to the ion.

The fitting to the experiments (solid lines shown in Fig. 2) gave the following concentrations: $c_v = 2.7\times10^{16}\,\text{cm}^{-3}$ and $c_{\text{ion}} = 6.6\times10^{17}\,\text{cm}^{-3}$ in Si-doped GaAs, and $c_v = 5\times10^{15}\,\text{cm}^{-3}$ and $c_{\text{ion}} = 1.2\times10^{17}\,\text{cm}^{-3}$ in undoped GaAs. These concentrations are typically those found earlier in GaAs. The vacancies have been earlier identified as negative Ga vacancies and the negative ions as acceptor impurities or Ga antisites.[6,9–11]

We calculated the diffusion coefficient from the trapping and detrapping rates using Eqs. (3) and (4). The results are plotted in Fig. 3 together with the earlier high-temperature values of Soininen *et al.*[4] One can see that the present D_+ values from both samples fall around the same line. Further, they join smoothly with the high-temperature values. The value of D_+ at 290 K is $1.6\pm0.2\,\text{cm}^2\,\text{s}^{-1}$ and it increases to $14\pm2\,\text{cm}^2\,\text{s}^{-1}$ when the temperature drops down to 20 K.

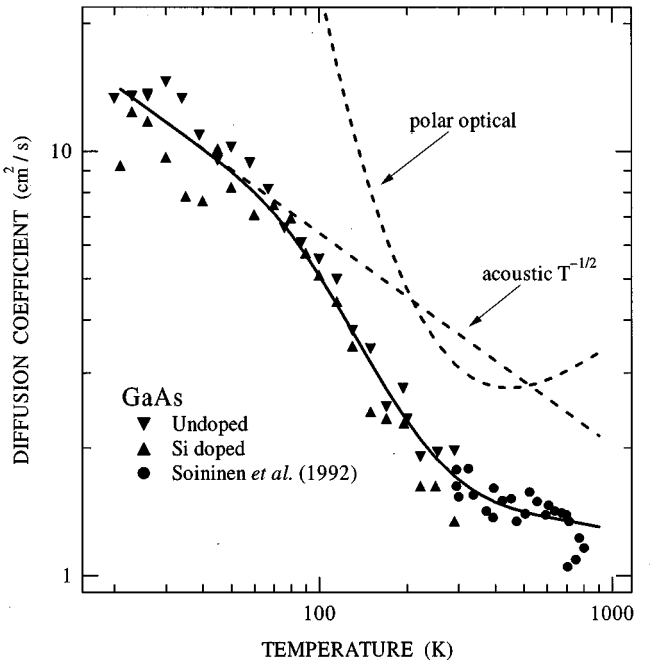

FIG. 3. Positron diffusion coefficient in GaAs deduced from two different samples. The points above 300 K are taken from Soininen *et al.* (Ref. 4). The broken lines show the theoretical contributions of the longitudinal-acoustic and polar-optical phonons. The solid curve is the sum of the both contributions.

Below 80 K the slope is close to $T^{-1/2}$ which is the prediction based on scattering off acoustic phonons.[12]

The recent value of Shan *et al.*[5] at 290 K is $1.6\pm0.5\,\text{cm}^2\,\text{s}^{-1}$ which is in good agreement with the present one. They also found the strong increase of D_+ with a slope of $T^{-1.5}$ when temperature decreased from 290 to 50 K. This slope is slightly steeper than that of our results and their values overshoot our values below 100 K. They failed to observe the change of the slope to $T^{-1/2}$ because they did not measure at low enough temperatures. Taking into account all the difficulties in determining the absolute diffusion coefficient values we consider that the agreement of our results and and those of Shan *et al.*[5] is good.

V. MODELING OF THE DIFFUSION COEFFICIENT

In this section we extend down to 20 K the earlier calculations of the theoretical positron diffusion coefficient in GaAs by Soininen *et al.*[4] The contributions of longitudinal acoustic (LA) and polar-optical phonon (LO) scattering are summed up as

$$D_+^{-1} = D_{\text{LA}}^{-1} + D_{\text{LO}}^{-1}. \tag{7}$$

The relaxation time approximation is used for D_{LA} with the earlier parameter values for the deformation potential $E_d = -5.83$ eV,[13] for the average elastic constant for longitudinal phonons modes $c_L = 14.1\times10^{10}\,\text{Nm}^{-2}$, and for the positron effective mass $m^* = 1.3 m_0$. Please note that m^* was the only adjustable parameter by Soininen *et al.*,[4] the value of which was obtained by fitting the theoretical D_+ to the experiments between 300 and 600 K.

The scattering from polar-optical phonons is both inelastic and unisotropic, and therefore a universal relaxation time cannot be defined. We follow the calculation of Fletcher and Butcher[14] which is based on an exact solution of the linearized Boltzmann equation. This calculation yields the mobility which gives the diffusion coefficient via the Einstein relation. The following material parameters are used: the phonon frequency $h\omega = 35.3\,\mathrm{meV}$, and the static and high-frequency dielectric constants $\varepsilon(0) = 12.85$ and $\varepsilon(\infty) = 10.89$, respectively.

The contributions of D_{LA} and D_{LO} are plotted separately as broken lines in Fig. 3. The acoustic phonons give the $T^{-1/2}$ dependence. As can be seen, it fits well the experimental diffusion coefficient from 20 to 80 K. The contribution of polar-optical phonons is switched on around 100 K explaining the rapid decrease of the experimental diffusion coefficient.

The total diffusion coefficient from Eq. (3) is shown as the solid line in Fig. 3. A good agreement between the experiments and theory is achieved in the whole temperature range from 20 to 700 K. The present experimental data indicate that above 100 K positron diffusion is limited both by longitudinal-acoustic and polar-optical phonons.

It is interesting to note that no contribution of charged-impurity scattering is seen in the experimental results of the Si-doped GaAs sample although it contains $2\times10^{18}\,\mathrm{cm}^{-3}$ ionized Si^{+} impurities. The reason is that the critical donor concentration for the semiconductor-metal (Mott) transition is only $5\times10^{16}\,\mathrm{cm}^{-3}$ in GaAs. Thus our sample is metallic and the ionized Si^{+} impurities stay well screened by the degenerate free electrons even at low temperatures.

VI. SUMMARY

We have reported on a positron-beam study of GaAs where positron diffusion coefficient was experimentally determined in the temperature range 20–290 K. We have seen that the positron diffusion length shortens at low temperatures when the sample contains defects like vacancies and negative ions. The diffusion coefficient can be deduced by taking into account the defect trapping effects. The diffusion coefficient of free positrons follow the $T^{-1/2}$ law in the temperature range from 20 to 80 K due to scattering from acoustic phonons. From 80 to 290 K the diffusion coefficients strongly decreases because of the onset of scattering from the polar-optical phonons. Above 300 K the diffusion coefficient is rather flat due to the presence of both types of phonons.

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