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Positron trapping at vacancies in Ga

N de Diego†, J del Rio†, P Moser‡ and F Plazaola§

† Départamento de Física de Materiales, Universidad Complutense, 28040 Madrid, Spain
‡ Centre d'Etudes Nucléaires de Grenoble, DRF/SPh/MP, 85X, 38041 Grenoble Cédex, France
§ Elektrika eta Elektronika Saila, Euskalherriko Unibertsitatea, PK 644, 48080, Bilbo, Spain

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Abstract. Gallium is one of the very few elements in which positron trapping at vacancies *under equilibrium conditions has not been experimentally observed. We present clear* evidence of positron trapping in electron-irradiated Ga at 77 K. The recovery of the vacancies produced by irradiation as shown by the recovery of both the lifetime and the Doppler parameter is observed at around 125 K.

1. Introduction

It is a well established fact that positron-bound states are present in most metals. However, positron trapping at vacancies has not been found in Ga under equilibrium conditions [1] and only the effects of thermal expansion are observed up to the melting point [2]. It has been argued that a strong relaxation around vacancies could be responsible for the absence of positron-bound states in an unusual crystal structure such as Ga [3]. A strong relaxation could indicate a small open volume associated with the vacancy and thus a low positron–defect binding energy; in such case the effects of thermal detrapping could be noticeable and could prevent observation of trapping effects even close to the melting point.

With the aim of understanding the absence of trapping effects in isothermal measurements in Ga, we have performed positron measurements on Ga samples irradiated at low temperatures in order to create a high density of vacancies in such a condition that no detrapping effects are expected.

2. Experimental details

Well annealed high-purity Ga samples (purity, 99.9999%) were electron irradiated at 20 K in a Van de Graaff accelerator at a dose of 2×10^{18} electrons cm^{-2}, with an electron energy of 3 MeV.

Positron lifetime measurements were performed with a fast system having a resolution of 250 ps (FWHM); simultaneously, Doppler-broadening spectra were recorded using a Ge(Li) detector having a linewidth of 1.2 keV (FWHM) at the annihilation peak.

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Table 1. Lifetimes of the well annealed, the quenched and the irradiated samples. T_M is the measurement temperature.

	T_M (K)	τ_1 (ps)	τ_2 (ps)	I_2 (%)
Well annealed Ga	77	195±2		
Well annealed Ga	275	198±2		
Quenched Ga	77	195±2		
As-irradiated Ga	77	181±4	303±12	23±5

The annealing curve was obtained by measurements in the temperature range 77–275 K; the temperature was raised to the required value and maintained for 5 h, whereafter the sample was cooled to the measurement reference temperature of 77 K. A measurement was also performed at each annealing temperature.

Lifetime measurements were also made at 77 and 275 K in a well annealed sample in order to extract the bulk lifetime. An additional measurement was carried out at 77 K in a sample quenched from 300 K to liquid-nitrogen temperature.

The lifetime spectra were analysed using one- or two-component decomposition after subtracting the source contributions.

The Doppler-broadening spectra were analysed in terms of the parameter D defined as $D = S/W$, where S and W represent respectively the central area and the area of two symmetric wing segments of the Doppler spectrum. The annealing curves were characterized by D/D_0, where D_0 is the value of the parameter D in the well annealed sample.

3. Results

The lifetime values for the well annealed, the quenched and the as-irradiated samples are shown in table 1. For the defect-free and quenched samples, one-component analyses gave satisfactory fits, whereas two-component decomposition was necessary in the as-irradiated sample. The annealing curves for both the mean lifetime $\bar{\tau}$ and the parameter D are plotted as a function of the temperature in figures 1(a) and (b), respectively.

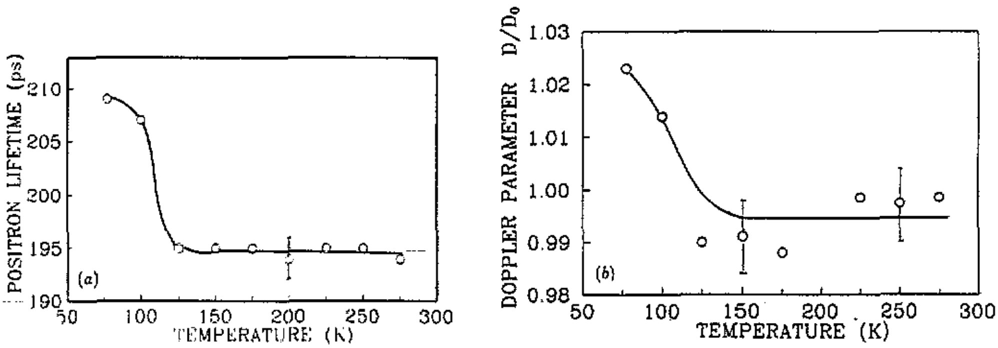

Figure 1. The evolution of (a) the average lifetime $\bar{\tau}$ and (b) D/D_0 as functions of the annealing temperature in an electron-irradiated Ga sample.

The mean lifetime $\bar{\tau}$ measured at 77 K ranges from 209 ps for the as-irradiated sample to 195 ps, which is achieved at 150 K and corresponds to the measured bulk value (see figure 1(*a*)). It should be remarked that two-component analyses gave a better fit for the samples annealed at 100 K. However, the statistical scatter was too high to consider the analyses reliable. The $\bar{\tau}$ behaviour of the samples measured at the annealing temperature is the same as its evolution for the samples measured at 77 K up to 175 K. From this temperature a linear increase in $\bar{\tau}$ is observed up to 275 K where the value measured in the well annealed sample, $\tau = 198$ ps, is attained. In both the $\bar{\tau}$ and the D curves (figures 1(*a*) and (*b*)) it can be seen that the damage produced by the electrons is completely recovered at 150 K.

4. Discussion

The lifetime values obtained for well annealed Ga samples agree with previously measured values [4, 5]. The difference between the values measured at 77 K and those measured at 275 K can be attributed to thermal expansion effects, as reported for the Doppler measurements by Segers *et al* [2].

The increase observed in the average lifetime $\bar{\tau}$ and in D after electron irradiation clearly indicates trapping at vacancy-type defects. The threshold displacement energy for Ga has been found to be $E_D = 12$ eV [6]; thus each primary electron with an energy of 3 MeV is expected to displace about eight atoms, meaning that vacancies and probably very small vacancy clusters such as divacancies or trivacancies might be present in the electron-irradiated samples studied.

A measure of the effect of trapping by monovacancies on the lifetime is usually given by the parameter

$$\Delta\tau/\tau_b = (\tau_{1v} - \tau_b)/\tau_b$$

where τ_b is the lifetime in the defect-free sample and τ_{1v} the value associated with a monovacancy. According to the data collected from the literature [7], $\Delta\tau/\tau_b$ ranges between 0.1 and 0.8 excluding the values for the alkali metals, which are extremely low. In the case of Ga and by assuming that $\tau_{1v} = 300$ ps, $\Delta\tau/\tau_b$ yields a value of 0.54 within the data in the literature for monovacancies. Seeger and Banhart [7] define a dimensionless parameter δ_{jv} as a measure of the lifetime for vacancy-type defects. According to their definition, δ_{jv} decreases for increasing number of vacancies in the cluster. We have calculated the δ_{1v} value for Ga by assuming that $\tau_{1v} = 300$ ps and it yields a value of 0.66, slightly below but very close to the lowest value obtained from the literature, which is 0.69. Moreover, it should be remarked that at 77 K the simple trapping model yields a value for τ_1 in accordance with that found experimentally.

Therefore, we can conclude that the long component present in the irradiated samples at 77 K can be unambiguously assigned to monovacancies produced by the electron damage, showing thus that vacancies are able to trap positrons in Ga.

The damage produced is recovered at 150 K (see figures 1(*a*) and (*b*)) where the lifetime and D achieve their bulk values. We can assign this stage to vacancy migration with an activation energy of 0.4 eV as deduced from the experimental data. The temperature dependence shown by the lifetime when measuring at the annealing temperature would be again associated with thermal expansion effects [2].

From self-diffusion data on Ga polycrystals the self-diffusion energy has a value of approximately 1.4 eV [8]; if we assume from our results that 0.4 eV is the vacancy migration energy, we can obtain a value of 1 eV for the vacancy formation energy. With this value we can roughly estimate the vacancy concentration close to the melting point. According to the data in the literature [9] the vibrational entropy in FCC metals gives a contribution of a factor of about 10 to the vacancy concentration; to the present authors' knowledge no data on the vibrational entropy for Ga are available in the literature, but its order of magnitude is not expected to differ very much from that of the Boltzmann constant; thus, even close to the melting point the vacancy concentration would be as low as 10^{-15}. These arguments could explain the lack of a vacancy trapping signal in isothermal positron measurements in Ga. This effect has also been observed in Sb [10] and it has also been explained in terms of a low vacancy concentration well below the sensitivity of the positron technique.

The measurement made at 77 K in a sample quenched from 300 to 77 K supports the above hypothesis. We can assume that, after quenching, almost all the vacancies at the equilibrium temperature are retained in the specimen. This means that, by measuring at 77 K immediately after quenching, we should have a vacancy concentration close to that at 300 K, since vacancies or vacancy clusters are stable at 77 K, as proved by the isochronal measurements for the electron-irradiated sample. Nevertheless, the value obtained for the quenched specimen reproduces the bulk value, in agreement with the previous hypothesis of a vacancy concentration close to the melting point (about 300 K), below the detection limit of the positron technique.

5. Conclusions

Clear evidence of positron trapping has been observed in electron-irradiated Ga at 77 K. In the as-irradiated sample a long component with a lifetime τ of 300 ps is measured, indicating that vacancies are trapping centres for positrons. A rough estimation of the vacancy concentration close to the melting point, based on the self-diffusion data in the literature and our measurements, yields an extremely low value, well below the sensitivity of positrons. In the light of these results, the absence of trapping by thermal vacancies cannot be explained by the lack of positron-bound states in vacancies; it can rather be attributed to a low vacancy concentration in Ga in the whole temperature range up to the melting point.

References

[1] Szymanski C, Chabik S, Pajak J and Rozenfeld B 1980 *Phys. Status Solidi* a **60** 375
[2] Segers D, Dauwe C, Dorikens M and Dorikens-Vanpraet L 1977 *Phys. Status Solidi* a **40** 153
[3] Seeger A 1973 *J. Phys. F: Met. Phys.* **3** 248
[4] Weisberg H and Berko S 1967 *Phys. Rev.* **154** 249
[5] Brandt W and Waung H F 1968 *Phys. Lett.* **27A** 700
[6] Myhra S and Gardiner R B 1975 *Radiat. Eff.* **27** 35
[7] Seeger A and Banhart F 1987 *Phys. Status Solidi* a **102** 171
[8] Carter A C and Wilson C G 1968 *J. Phys. D: Appl. Phys.* **1** 515
[9] Seeger A 1973 *Cryst. Latt. Defects* **4** 221
[10] de Diego N, Hidalgo C and Moser P 1987 *Mater. Sci. Forum* **15–18** 193