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Calculation of positron lifetimes for bulk and vacancy-type defects in Ga simple metal

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Abstract. Theoretical calculations have been carried out in order to extract the annihilation characteristics of bulk and mono- and divacancies expected in electron-irradiated Ga simple metal. From these theoretical results it is inferred that the positron signal reported in electron-irradiated Ga at 77 K in a previous work is due to divacancies present in the metal at 77 K.

1. Introduction

In a previous paper [1] we showed evidence of positron trapping at monovacancies at 77 K in electron-irradiated Ga and concluded that the lack of trapping signal in isothermal positron measurements in Ga is due to the low vacancy concentration even close to the melting point in this metal. In this paper we present theoretical lifetime calculations for bulk Ga and vacancy defects and reinterpret the experimental measurements in the light of these results. We have used a reliable method for the lifetime calculations in metals based on the atomistic model of Puska and Nieminen [2]. Even though this method is a simple one that makes use of non-self-consistent electronic densities, it gives good values for lifetimes in simple metals [3, 4]. The good agreement between the experimental and theoretical lifetimes is mainly due to the fact that the positron annihilation rate is obtained as an integral over the product of positron and electron densities and the positron density relaxes following the electron charge transfer, keeping the value of the overlap integral constant.

In section 2 we present the calculation method for the positron states and the theoretical results; section 3 is devoted to the discussion and reinterpretation of the experimental results of [1] and in section 4 we give the conclusions of the present work.

2. Calculation of positron states

The potential seen by the positron, its wave function, and annihilation characteristics in a given atomic arrangement have been calculated using the atomistic model of Puska and Nieminen [2], which retains the three-dimensional character of the atomic arrangement. This method is based on non-self-consistent electron structures, which is much less time consuming than the simultaneous self-consistent calculation of the electron density and the positron state. Model calculations made by the two-component density-functional theory

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[5, 6] for the positron localized in a metal vacancy [7, 8] show that the self-consistency of the electron structure changes the positron annihilation rate only slightly. For a non-localized positron in a perfect metal host the non-self-consistent superposition of free atoms used in the present method reproduces rather well the electron density and the open volume in the interstitial regions [4]. In the case of a perfect host these are indeed the most important aspects of the positron state calculations, because, due to the strong repulsion from the ion cores, the positron wave function is mainly localized in the interstitial regions between the ions.

The three-dimensional potential $V_+(\boldsymbol{r})$ sensed by the positron [2] is constructed as a sum of an electrostatic potential $V_H(\boldsymbol{r})$ due to nuclei and electrons, and of a correlation potential V_{corr} describing the electron–positron correlation effects:

$$V_+(\boldsymbol{r}) = V_H(\boldsymbol{r}) + V_{corr}(\boldsymbol{r}). \tag{1}$$

Thus, the correlation potential depends on the local electron density unperturbed by the positron and it corresponds to the low-density limit of the positron in two-component density-functional theory. Therefore, within the local-density approximation (LDA) the correlation energy $V_{corr}(\boldsymbol{r})$ is given by the electron–positron correlation energy of one positron in an electron gas of density $n(\boldsymbol{r})$. The density dependence of V_{corr} is obtained using the parametrization of Boronski and Nieminen [8]. The charge density $n(\boldsymbol{r})$ and the electrostatic potential $V_H(\boldsymbol{r})$ in the crystal (either perfect or defected) is approximated by superimposing atomic charge densities:

$$n(\boldsymbol{r}) = \sum_i n_{at}(|\boldsymbol{r} - \boldsymbol{R}_i|) \qquad V_H(\boldsymbol{r}) = \sum_i V_{at}(|\boldsymbol{r} - \boldsymbol{R}_i|) \tag{2}$$

where $\boldsymbol{R}_i$ runs over the occupied atomic sites, and n_{at} and V_{at} are the atomic electron density and Coulomb potential, respectively. The Ga free-atom results needed for this construction are calculated by a self-consistent density-functional program with the local-spin-density approximation [9] using the exchange–correlation potential of Ceperly and Alder [10].

The potential of equation (1) is calculated at the nodes of a three-dimensional mesh that forms an orthorhombic Bravais lattice. The Schrödinger equation is discretized, and the positron wave function and its energy eigenvalue are solved iteratively at the mesh points by a numerical relaxation method [11]. The boundary conditions used for the trapped states are the vanishing of the wave function on the surface of a large enough symmetric polyhedron enclosing the trap. In practice, this polyhedron has been chosen so that its surface lies at least 1.5 lattice constants away from the centre of the trap in each direction of the orthorhombic Bravais lattice. The boundary condition chosen for the positron wave function in a perfect lattice of Ga is the vanishing of its normal derivative on the surface of the orthorhombic Bravais lattice.

The solution of the three-dimensional Schrödinger equation gives the energy eigenvalue and the wave function $\Psi_+(\boldsymbol{r})$ for the positron. From the wave function we calculate the positron annihilation rate using the LDA

$$\lambda_{LDA} = \int d\boldsymbol{r}\, |\Psi_+(\boldsymbol{r})|^2 \Gamma[n(\boldsymbol{r})] \tag{3}$$

where $\Gamma[n(\boldsymbol{r})]$ is the annihilation rate of a positron in an electron gas of density n. This calculation was performed by using the LDA for core and valence electrons [3], and for $\Gamma(n)$ the following expression [8] was used:

$$\Gamma[n(\boldsymbol{r})] = \pi r_0^2 cn(1 + 1.23r_s + 0.8295r_s^{3/2} - 1.26r_s^2 + 0.3286r_s^{5/2} + r_s^3/6) \tag{4}$$

where r_0 is the classical electron radius, c is the velocity of light and $r_s = (3/4\pi n)^{1/3}$.

It has to be mentioned that even though the use of non-self-consistent electron densities gives smaller positron binding energies to defects than the use of self-consistent ones [4], this calculation method has proved to give theoretical positron lifetimes in simple metals, either perfect or defected, that agree very reliably with the experimental values [3, 4].

The positron wave function calculated for the orthorhombic structure of Ga [12] is shown in figure 1. Due to the complex atomic configuration of the Ga structure two types of divacancies have been chosen to perform the theoretical calculations. These two configurations are shown in the schematic representation of the orthorhombic structure of Ga in figure 2: configuration (a) corresponds to diatomic vacancies in the diatomic unit and configuration (b) to two adjacent atomic positions belonging to two different diatomic units (see the figure captions for a better description of the configuration). The positron wave functions calculated for monovacancies and divacancies belonging to the configuration (a) are shown in figure 3. The values of the lifetimes $\tau_{LDA} = 1/\lambda_{LDA}$ calculated for Ga bulk, monovacancies and divacancies with configurations (a) and (b) as well as the positron binding energies to the different defects are given in table 1.

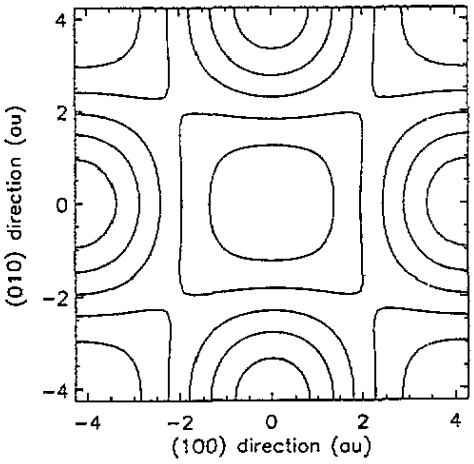

Figure 1. Positron wave function contour plot for Ga

Table 1. Calculated positron lifetimes τ and positron binding energies E_b in bulk and different vacancy-type defects in Ga.

	τ (ps)	E_b (eV)
Bulk	196	—
Monovacancy	240	0.44
Divacancy (a)	294	1.50
Divacancy (b)	268	1.18

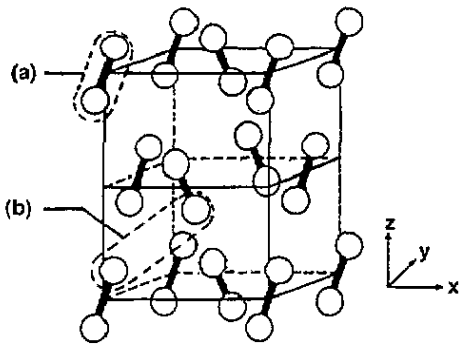

Figure 2. Schematic structure of the orthorhombic Ga structure, showing the diatomic units lying on the face centres and on the corners of the unit cell. The two atomic positions taken to form the two divacancy configurations studied in this work have been marked as (a) and (b). The (a) configuration corresponds to the diatomic unit; the (b) configuration corresponds to two adjacent atomic positions belonging to two different diatomic units (highlighted in the figure).

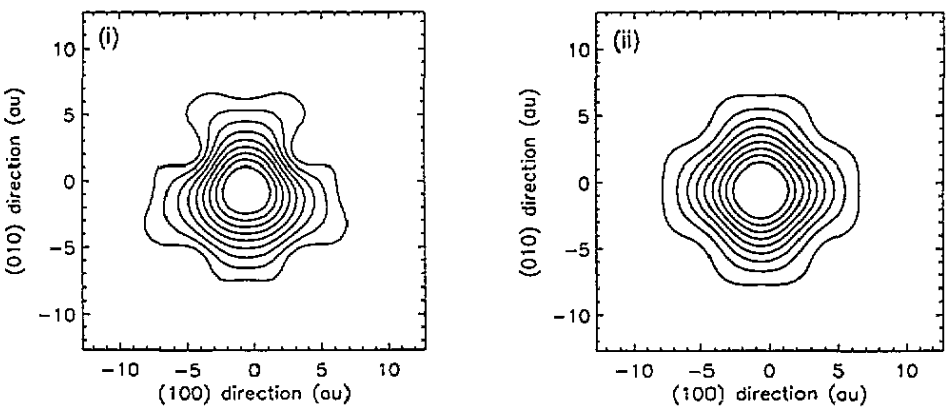

Figure 3. Positron wave function contour plot in a Ga matrix for (i) a monovacancy and (ii) a divacancy in configuration (a).

3. Discussion

The calculated lifetime value for the defect-free material presented in table 1, $\tau = 196$ ps, is in agreement with our experimental value [1] and the former experimental data [13, 14]. In the as-irradiated sample we found a long component, $\tau = 303$ ps, which we attributed to monovacancies. However, by inspection of table 1, we observe that the experimental value is closer to that calculated for a divacancy in the configuration labelled as (a), which has also a higher binding energy for the positron than the monovacancy and the divacancy in configuration (b) (see figure 2 and table 1). According to these data and recalling that the experimental data reported in [1] satisfy the one-trap model, we conclude that the trapping signal observed in electron-irradiated Ga at 77 K is due to divacancies in the configuration

labelled (a) in figure 2.

The question is to elucidate whether monovacancies are formed in Ga by electron irradiation and migrate below 77 K giving rise to divacancies, or whether the only stable vacancy-type defects in Ga are divacancies in the diatomic unit. A third hypothesis of monovacancies stable above 100 K is to be excluded, since the positron–monovacancy binding energy is high enough to originate a positron bound state at that temperature and the associated lifetime should be easily resolved from the experimental spectra, giving rise to a component of $\tau = 240$ ps (see table 1), which is not experimentally observed. It is to be remarked that the previous interpretation concerning the absence of positron trapping at thermal vacancies is still valid and can be interpreted in terms of a low density of vacancy-type defects close to the melting point below the detection limit of the positron technique. If the first hypothesis holds, the estimated activation energy for the annealing stage between 77 and 125 K, $E = 0.4$ eV [1], should then correspond to the divacancy binding energy in Ga and the monovacancy migration energy would be lower than 0.4 eV. According to the self-diffusion data [15], the monovacancy concentration close to the melting point would be even lower than the value estimated in our previous work [1] as the monovacancy formation energy would be higher than 1 eV. In the second case no isolated monovacancies could form and the reasoning previously applied for monovacancies [1] would hold for divacancies.

The recovery behaviour below 50 K reported by Myhra and Gardiner [16] in electron-irradiated Ga has been interpreted as interstitial migration, although the migration of very unstable monovacancies recombinating with moving interstitials cannot be excluded. It is not very unlikely that a divacancy should be more stable than a monovacancy, considering the orthorhombic Ga structure [12] (see figure 2), which can be considered as an assembly of diatomic molecules. It would be highly desirable to study by positron annihilation in the temperature range 20–50 K the electron damage in Ga to understand the point-defect kinetics, which would help to clarify the diffusion mechanism in this metal.

4. Conclusions

The main conclusion of the present work is that the trapping signal observed in irradiated Ga at 77 K is due to divacancies and not to monovacancies as it had been interpreted in a previous paper.

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