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Positron lifetime in vacancy-impurity complexes (*)

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Résumé. — Nous avons calculé la durée de vie du positon dans des complexes lacune-solutés (Na, Mg, Zn) dans l'aluminium. La durée de vie dépend du complexe et augmente en présence de ces solutés. Dans certains cas, la durée de vie est même voisine de celle de la bilacune. Ces résultats montrent que la distinction des complexes lacunaires de complexes de type monolacune-solutés est difficile par une mesure de temps de vie du positon.

Abstract. — We calculate the positron lifetime in vacancy-solute (Na, Mg, Zn) complexes in Al. The lifetime depends on the solute atom and increases with these solute. The variation is in some cases of the same order as for the divacancy. These results indicate that vacancy complexes and vacancy-impurity complexes are not easily distinguished by lifetime measurements.

1. Introduction. — The positron annihilation spectroscopy [1] can be fruitfully exploited to study vacancy-type defects in metals [2] since the positrons are trapped there and annihilate in the trapped state. A number of investigations on the properties of vacancies [3], especially the recovery, have already been performed by this technique during the last few years. Recently however, the interest has shifted to the case of doped metals [4]. If solute atoms are to play a role in the recovery of vacancies, then clearly the structure of secondary defects in doped metals must be different from that in pure metals. One hopes that the positron is sensitive enough to the variations of the electron density that one can distinguish the complexes of the type $L_n Y_m$ with n vacancies and m solute atoms (L stands here for a vacancy and Y for a solute atom) from clusters of vacancies L_k , with k vacancies.

Several theoretical calculations exist in the literature on the characteristics of a positron annihilating in monovacancies. Hautojärvi *et al.* [5] have estimated the positron lifetime in a cluster of N vacancies by considering the cluster as a spherical void. However, to our knowledge, no calculations exist to date for the positron lifetime in complexes $L_n Y_m$. In these calculations the presence of the iron-cores cannot be

neglected. As a result these calculations cannot be performed in the jellium model or its extensions [6] commonly used for vacancies in simple metals where the main contribution to the annihilation rate of the positron comes from valence electrons and the contribution of the core electrons is either completely neglected or accounted for in an *ad hoc* manner. An augmented plane wave (APW)-type calculation used by Gupta [7] for a monovacancy would be better for this problem. But for this, exact spatial structure and geometry of the complex must be known and these are not always well defined. We have therefore used a simple model to estimate the annihilation parameters for a positron trapped in a complex in Al.

This note gives the calculation of the lifetimes in the complex LY_m with the following structure :

— a vacancy (L) is taken as the centre of the complex,

— m solute atoms (Y), supposed to be in substitutional positions, surround the vacancy in the first nearest neighbour configuration.

For an fcc lattice this restricts $m \leq 12$. The cases $m = 0$ and $m = 1$ correspond respectively to a single vacancy and to the simplest complex LY formed by a vacancy with a first neighbour solute atom.

2. Model. — In the independent particle model (IPM) the annihilation rate λ^i of a positron is pro-

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portional to the overlap of the electron and positron densities ρ_-, ρ_+ ,

$$\lambda^i = \pi r_e^2 c \int d^3r \rho_-(\mathbf{r}) \rho_+(\mathbf{r}).$$

Here r_e is the classical electron radius (2.81×10^{-15} m) and c the velocity of light.

Because of the polarization of the electron density by the positron, the electron density around the positron is enhanced. Several theoretical investigations suggest that the core electron density enhancement is not the same as the one for valence electrons and have therefore introduced two enhancement factors; ε_c for the core electrons and ε_v for the valence electrons. The positron lifetime $\tau = (\lambda^{-1})$, λ is the total annihilation rate) can then be written as

$$\tau^{-1} = \varepsilon_c \lambda_c^i + \varepsilon_v \lambda_v^i = \tau_c^{-1} + \tau_v^{-1}.$$

For the valence electrons we choose the Brandt-Reinheimer formula [8] for enhancement factor $\varepsilon_v = 8/3 + 2/\lambda_v^i$. For the core electrons in Al a value of $\varepsilon_c = 1.6$ may be taken [7].

2.1 ELECTRON DENSITY. — In our model the electron density ρ_- is taken as the superposition of the neutral atom charge densities ρ_v situated at sites R_v . The latter is calculated selfconsistently in the Hartree-Fock-Slater scheme with exchange parameter $\alpha = 1$ in the manner of Herman-Skillman [9]. Taking the vacancy at the origin

$$\rho_-(\mathbf{r}) = \sum'_v \rho_v(\mathbf{r} - \mathbf{R}_v)$$

where the prime on the summation excludes the $v = 0$ term. In this fashion the solute is represented by a simple replacement of the atomic charge density of the host by that of the solute at that position. Admittedly, this procedure ignores the screening associated with the solute which is however not easy to account for in any case.

2.2 POSITRON DENSITY. — The potential for the trapped positron in the vacancy is approximated by a spherical square potential well, the radius R_0 being equal to the Wigner-Seitz radius R_{ws} . The depth V_0 is taken as the average depth of the potential obtained in the APW calculation [7] :

$$V_0 = \left(\frac{4\pi}{3} R_0^3 \right)^{-1} \int_0^{R_0} 4\pi r^2 V(r) dr$$

where $V(r)$ is the APW potential for the positron (relative to the Bloch state). With a lattice constant $a = 4.04$ Å we find $V_0 = 8.14$ eV, a value intermediate between the two values given by Hodges [10].

For the positron trapped in the vacancy with a binding energy E ($E < 0$) the following transcendental equation is obtained :

$$\tan [R_0(V_0 + E)^{1/2}] = - \left(\frac{V_0 + E}{E} \right)^{1/2}.$$

The positron density is then given by

$$\rho_+(r) = C_1 \sin^2 w_1 r \quad r < R_0$$

$$\rho_+(r) = C_2 \exp(-2 w_2 r) \quad r > R_0$$

with

$$w_1 = (V_0 + E)^{1/2} \quad w_2 = (-E)^{1/2}$$

$$C_1 = -\sin(2 w_1 R_0)/4 w_1 + C^2 \exp(-2 w_2 R_0)/2 w_2$$

$$C_2 = C^2 C_1 \quad C^2 = \sin^2 w_1 R_0 \exp(-2 w_2 R_0).$$

2.3 LIFETIME OF THE POSITRON IN THE VACANCY AND THE COMPLEX LY_m . — The electron and positron densities calculated above are used to calculate the positron lifetime in the monovacancy and the vacancy-solute atom complex. The IPM annihilation rate λ^i is decomposed into contributions $\lambda_{nl}^i(R_v)$ from different atomic (n, l) shells and sites ($\mathbf{R}_v$) :

$$\lambda_{nl}^i(\mathbf{R}_v) = \pi r_e^2 c \int d^3r \rho_+(\mathbf{r}) \rho_{v,nl}(\mathbf{r} - \mathbf{R}_v).$$

Since the positron density is spherically symmetric with respect to the vacancy site, only the spherical component of $\rho_{v,nl}$ projected at the vacancy site contributes; the latter can be obtained from Lowdin's formula [11]

$$\rho_{v,nl}(r, R_v) = \frac{1}{2 R_v r} \int_{|R_v-r|}^{|R_v+r|} r \rho_{v,nl}(r) dr.$$

We considered fourteen shells of atoms surrounding a vacancy, so that

$$\lambda^i = \sum_{nl} \sum_{v=1}^{14} N_v \lambda_{nl}^i(R_v)$$

where N_v is the number of atoms at distance R_v from the vacancy.

The IPM rate of annihilation Λ^i in a complex LY_m is calculated from λ^i according to the following expression :

$$\Lambda^i = \lambda^i - m \sum_{n'l'} \lambda_{n'l'}^{\text{Al}}(R_1) + m \sum_{nl} \lambda_{nl}^{\text{Y}}(R_1)$$

where $\lambda_{nl}^{\text{Al}}(R_1)$ and $\lambda_{nl}^{\text{Y}}(R_1)$ are respectively the annihilation rates of an (n, l) shell of an Al and a solute atom and m is the number of solute atoms in the nearest neighbour position to a vacancy.

3. Results and discussion. — **3.1 LIFETIME IN A MONOVACANCY.** — With this method a value of $\tau_v = 258$ ps for the positron lifetime in a vacancy is obtained. This is equal to the value found in the jellium model [13], and agrees quite well with the available experimental values 243 ps [12], 246 ± 4 ps [13]. This shows that reasonable values of the positron life-

time in complexes can be obtained with this method in spite of its simplicity.

The APW calculated positron and valence electron density yield $\tau_v = 236$ ps [7]. If we use the APW valence electron density in conjunction with our positron density a value of $\tau_v = 240$ ps is obtained, which justifies the choice and the construction of our positron potential in the vacancy.

In our model it is difficult to introduce explicitly the contribution from core electrons, and hence in these calculations the core contribution is not included. The difficulty arises from the fact that our positron wavefunction is constructed from the potential well at the vacancy site and does not include the strong repulsion felt by the positron in the core region of the surrounding atomic sites. The neglect of this repulsion has little effect on the positron density at the vacancy site but it is extremely serious for the calculation of the core annihilation rates. Fortunately, in the case of Al the core contribution is quite negligible; its neglect increases τ_v by only 5 ps. However, this does restrict the applicability of our model :

- (i) the model cannot be used for transition metals without an appropriate scheme for core corrections,
- (ii) the core contribution expected from solute atoms must be very small as compared to the valence electron contribution. This is expected to be the case for Na and Mg solutes in Al.

3.2 LIFETIME IN COMPLEXES. — Positron lifetimes in complexes (LY_m) are given in table II for solute atoms Na, Mg, and Zn in Al. As indicated above these values are calculated with only the valence electron contribution. In table I the annihilation rate λ_{nl}^i from different valence electron shells (nl) of an atom at the nearest neighbour site of a vacancy is given.

Table I. — IPM annihilation rate λ_{nl}^i (in ns^{-1}) due to the (n, l) shell of an atom at the first neighbour site of a vacancy.

	3S	3S ²	3p ¹	4S ²
Al	—	—	—	—
Na	0.022 70	0.031 06	0.020 17	
Mg		0.039 08		
Zn				0.039 66

Table II. — Positron lifetime (in ps) in the vacancy-multiple solute complexes LY_m . (Lifetime in monovacancy $\tau_v = 258$ ps.)

<div>Y \ m</div>	1	2	3	4
Na	263	268	274	280
Mg	260	262	265	267
Zn	261	264	267	271

It is clear that in this simple model for Al^i the spatial structure of the complex does not appear as long as all the solutes are in the nearest neighbour configuration to a vacancy. All possible different geometries yield the same value. In a complex the positron wavefunction does not have the cubic symmetry since the repulsion of the positron from different ions is not the same. The anisotropy of the positron wavefunction depends upon the geometry of the complex. However, for the solutes under consideration the anisotropy is expected to be small and the square potential well at the vacancy site to be very little affected. We have for this reason, not changed the depth of the potential well with the addition of the solute.

It is interesting to compare the values of the positron lifetime in the complexes given in table II and those found in the case of positron trapping by defects formed only with vacancies. Divacancy is the simplest defect but our model does not allow the calculation of the positron lifetime in a divacancy since the positron wavefunction needs to be calculated first. However, if we make an estimate simply by removing the first nearest neighbour Al atom and thus suppressing its contribution, an increase of 10 ps over the monovacancy value is obtained. This is to be compared with an increase of 40 ps obtained by Hautojärvi *et al.* [5] and 14 ± 8 ps by Welch *et al.* [14]. It should be noted that in the calculation of Hautojärvi *et al.* the divacancy is represented by a sphere nearly twice the volume of a Wigner-Seitz sphere, and an increase of 40 ps is therefore quite likely an overestimate. From table II one finds that the lifetime increases by 10 ps (over the monovacancy value) with the formation of a $LiNa_2$ complex. It is thus clear that vacancy-multiple solute atom complexes can result in positron lifetimes comparable to those found in vacancy complexes.

As yet there are very few experimental data on the positron lifetime in vacancy-solute complexes, so a comparison with our calculations is not possible. In Al-1.7 % Zn, measurement of the lifetime as a function of temperature was performed by Hall *et al.* [12]. The fit of the mean lifetime $\bar{\tau}(T)$ gives a lifetime in the complex of the same order as in the monovacancy. This is in agreement with our calculations since we find the lifetimes in the vacancy and vacancy-solute complex to be respectively 258 and 261 ps respectively. Note that a slightly larger core contribution from Zn (not included in our calculation) relative to the one in Al will decrease the lifetime in the complex.

4. Conclusion. — The simple model presented in this work yields essentially the same value for the positron lifetime in a monovacancy in Al as the more elaborate previous calculations [13]. However, since the behaviour of the positron wavefunction in the ion-core region is not correctly represented in our

model, it is not possible to include annihilations from the core electrons. This is not a new problem and is faced by nearly all the simple models. This model has the advantage however that in a very simple manner it has been possible to calculate the modifications in the positron annihilation characteristics in vacancy-solute complexes. In Al with Na,

Mg, Zn solutes, the lifetimes in the monovacancy and the monovacancy-monosolute complex are too close to be distinguishable easily by lifetime measurements. Thus to identify a complex lifetime measurements alone may not be sufficient. Perhaps, experimental coupling between lifetime and angular correlation measurement may yield more insight.

References

- [1] WEST, R. N., *Adv. Phys.* **22** (1973) 264.
 - [2] BRANDT, W., in *Positron Annihilation*, edited by L. O. Roellig (Academic Press, New York) 1967, p. 155.
 - [3] WEST, R. N., in *Positrons in Solids*, edited by P. Hautojärvi (Springer, Heidelberg) 1979, p. 89.
 - [4] HAUTOJÄRVI, P., JOHANSSON, J., VEHANEN, A., YLI-KAUPILA, J. and MOSER, P., *Phys. Rev. Lett.* **40** (1980) 1326.
 - [5] HAUTOJÄRVI, P., HEINIO, J., MANNINEN, M., NIEMINEN, R., *Philos. Mag.* **35** (1977) 973.
 - [6] MANNINEN, M., NIEMINEN, R., HAUTOJÄRVI, P., ARPONEN, J., *Phys. Rev. B* **12** (1975) 4012.
 - [7] GUPTA, Raju P., SIEGEL, R. W., *Phys. Rev. B* **22** (1980) 4572.
 - [8] BRANDT, W., REINHEIMER, J., *Phys. Lett. A* **35** (1971) 109.
 - [9] HERMANN, F., SKILLMAN, S., *Atomic Structure Calculations* (Prentice-Hall, Englewood Cliffs) 1963.
 - [10] HODGES, C. H., *Phys. Rev. Lett.* **25** (1970) 284.
 - [11] LÖWDIN, P. O., *Adv. Phys.* **5** (1956) 1.
 - [12] HALL, T. M., GOLAND, A. N., SNEAD Jr., C. L., *Phys. Rev. B* **10** (1974) 3062.
 - [13] ARPONEN, J., HAUTOJÄRVI, P., NIEMINEN, R., PAJANNE, E., *Solid State Commun.* **12** (1973) 143.
 - [14] WELCH, D. O., LYNN, K. G., *J. Nucl. Mat.* **69-70** (1978) 611.
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