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AN APPROXIMATE METHOD FOR CALCULATING THE LIFETIME OF
POSITRONS TRAPPED BY LATTICE DEFECTS*

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ABSTRACT

A method which takes account of core as well as valence electrons is presented for theoretically estimating the lifetime of positrons trapped in lattice defects. The method is illustrated by calculations for vacancies and divacancies in aluminum.

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It is desirable to have a method for estimating the lifetime of positrons trapped at lattice defects which is flexible enough to use for defects more complex than single vacancies and which is not restricted to simple metals in which the pseudopotential technique is applicable. The method described here is motivated by the fact that positron lifetimes are essentially determined by the electron density and its spatial variation and that the electron density in solids, as determined by x-ray scattering, is described quite accurately by a superposition of free atom electron densities. Thus we shall approximate the electron density in a solid containing defects by superimposing electron densities appropriate to free atoms centered on each of the atomic sites of the configuration of interest. This zeroth order approximation to the electron density has the virtue that it can be applied to complex as well as simple defects. We shall approximate the positron wave function by choosing a simple variational function suitable to the defect of interest and minimizing the positron ground state energy in the Hartree field of the nuclei and the electron density distribution. (The spatial dependence of the electron-positron correlation energy is ignored in finding the positron wave function.) The lifetime is then obtained from the electron density averaged by the square of the positron wave function together with an expression for the density-dependent enhancement of the annihilation rate.

Clearly the utility of the method depends upon suitably choosing the variational wave function. The general form should be chosen so that the wave function has the proper symmetry and keeps the positron away from the positively charged nuclei. For a defect-free crystal one such form for the wave function is:

$$\psi_{+}^{\text{perfect}}(\vec{r}) = C_{+} \left[1 - \sum_j \phi_j(\vec{r} - \vec{R}_j) \right] \quad (1)$$

where ϕ_j is a positive function (to be chosen) centered on the atom at R_j which decreases with distance from the site and keeps the positron away from the nucleus. C_N is a normalization constant. A similar form can be written for a crystal with n vacant sites:

$$\psi_{+}^{n \text{ vacancies}} = C \left[\sum_{i=1}^n \phi_i^v(\vec{r}-\vec{R}_i) - \sum_{j \text{ occupied}} c_j \phi_j(\vec{r}-\vec{R}_j) \right] \quad (2)$$

The more strongly bound the positron is to the vacant sites, the more localized are the functions ϕ_i^v and the more rapidly the coefficients c_j decay with distance from the vacant sites. Variational functions, such as Equations (1) and (2), which are linear combinations of atom-centered functions are particularly useful if the potential consists of superimposed rapidly decreasing atomic potentials, as is the case with our approximation for the electron density.

A particularly simple one-parameter form for the perfect crystal wave function, Equation 1, is obtained by choosing ϕ_j to be a gaussian of the form:

$$\phi_j(\vec{r}) = \frac{e^{-|\vec{r}-\vec{R}_j|^2 \Lambda^2}}{1 + \sum_{k \neq j} e^{-|\vec{R}_j-\vec{R}_k|^2 \Lambda^2}} \quad (3)$$

This choice of ϕ_j makes the positron wave function vanish at each nucleus, and furthermore greatly facilitates the evaluation of multicenter integrals. The gaussian decay parameter Λ is chosen so as to minimize the energy.

An analagous one-parameter wave function for the n -vacancy case, Equation (2), is obtained by using the repulsive

functions ϕ_j of Equation 3 with the same decay parameter as the perfect crystal using a gaussian for the vacancy functions ϕ_i^v :

$$\phi_i^v = e^{-|\vec{r} - \vec{R}_i|^2 \Lambda_v^2} \quad (4)$$

and choosing the coefficients, c_j , of the repulsive functions to be of the form:

$$c_j = \sum_i e^{-|\vec{R}_j - \vec{R}_i|^2 \Lambda_v^2} \quad (5)$$

The decay parameter, Λ_v , for the vacancy functions and the repulsive coefficients is obtained by energy minimization.

To illustrate the accuracy of the results obtained with the use of such one-parameter functions, calculations were made of the binding energy of positrons to single and divacancies, and of the lifetime of positrons in perfect crystals and trapped in single and divacancies, all in aluminum. Although the method was developed in order to be able to include core electron effects, aluminum was chosen for the example, even though core annihilations are not dominant in this material, because of the existence of experimental data and of the results of prior calculations with which to compare the present results.

The electron density and electrostatic potential were constructed from the wave functions of Clementi (1). The decay parameters, Λ_i , for the gaussians of Equations (3-5) which minimized the energy were found to be $.26R_1$ for the repulsive functions and $.73R_1$ for the vacancy functions in the case of both the single and the divacancy.[†] The resulting binding energy of the positron to the single vacancy is 3.28 eV. This should be compared with the value of 3.84 eV obtained by Hodges (2) using a pseudo-potential technique and the values of 1.75 eV and 2.6 eV

[†] In these calculations, only one- and two-center integrals were retained. Furthermore all two-center integrals at distances greater than first neighbor were dropped.

obtained for Thomas-Fermi jellium and Kohn-Sham jellium (with the electron density of aluminum) respectively by Manninen et. al. (3). The present calculation also yields a binding energy for a positron to a divacancy in aluminum of 3.86 eV.

The lifetime was obtained using two different approximations for the annihilation rate. First, an approximate form for the annihilation rate has been proposed by Brandt and Reinheimer (4)

$$\Gamma(\rho) = (2 + 134\rho) 10^9 \text{ sec}^{-1} \quad (6)$$

where ρ is the electron density in atomic units. Thus in this approximation the annihilation rate varies linearly with the electron density averaged by the square of the positron wavefunction. In this approximation, the present calculations yield 150, 237, and 252 psec for positrons in the perfect crystal, the single vacancy, and the divacancy respectively.

Use of the Brandt-Reinheimer expression even in the core region implies enhancement factors for core electrons of about 2. Recent experiments (5) suggest that core enhancement factors are near unity. If it is assumed that annihilations in the neighborhood of a nucleus closer than 1 a.u. have a unit enhancement factor, the calculated lifetimes are 159, 249, and 264 psec for the perfect crystal, single vacancy, and divacancy cases respectively.

Our calculated perfect crystal lifetime of 150-159 psec should be compared to the experimental value(6) of 161 psec. The lifetime expected on the basis of uniform valence electron density alone is 178 psec. Thus our method accounts for the core effects reasonably well.

Our calculated lifetime for the single vacancy, 237-249 psec, should be compared with the experimental value (6) of 243 psec, and with the value of 237 psec calculated by Manninen et. al. (3) for vacancies in Kohn-Sham jellium using the West (7) renormalized density to include core effects.

Finally, we calculated the change in lifetime of a positron trapped in a single or divacancy when the nearest neighbors to the vacancy relaxed inward by 10%. In the case of the single vacancy, the lifetime decreased from 237-249 psec to 168-180 psec, or about 25%. For the divacancy, the decrease was from 252-264 psec to 202-216 psec, or by about 20%.

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