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Positron trapping at voids in metals: a generalized model

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Abstract. A general semi-classical model for describing positron trapping at voids in metals is presented. We show how the positron trapping rate increases linearly with temperature from a non-zero value at zero temperature. The model shows that information about the void radius is present in the temperature-dependent part of the trapping rate, and it is found to be consistent with observation in which void size and density have been determined by other techniques.

1. Introduction

The positron annihilation technique (PAT) is a well established method for the study of larger three-dimensional vacancy-type defects (voids) in metals (West 1979). However, the interpretation of experimental results and extraction of physical information is most often based upon comparison with data from earlier measurements (the 'fingerprint method'), rather than upon a well understood theoretical model.

In the existing theoretical descriptions of a positron interacting with a large defect, the process is mainly treated as a geometrical problem (Seeger 1974, Nieminen *et al* 1979), which requires the reservation that the approach is not expected to be valid when the de Broglie wavelength is comparable with the dimensions of the defect. For smaller defects, the interaction rate has, in many instances, been computed on the basis of a Golden Rule approach (Hodges 1970, 1974, McMullen 1977, 1978) which is applicable to purely inelastic processes, and predicts a temperature-independent trapping rate. It appears that neither of the two methods, nor a combination of them, is quite adequate as a general description of the positron–void interaction.

Experimental data for a number of different ensembles of voids in metals indicate that the transition-limited positron trapping rate can generally be described by the empirical formula (e.g. Bentzon *et al* 1985)

$$K_{tr} = K_0(1 + \alpha T) \tag{1}$$

where α is a constant varying from case to case. Recently Bentzon and Evans (1990) have performed positron-trapping measurements on a definite ensemble of voids in molybdenum, the voids being decorated in different ways. They found that the factor α remains virtually constant, despite the decoration, while only the constant K_0 varies with the impurity doping. This observation gives us reason to propose a model expected to be valid for positron trapping at voids of all sizes.

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2. Trapping model and transition-dependent trapping rate

In order to characterize metal voids by means of a positron lifetime spectrum, the first step is to employ the two- or three-state trapping model (Brandt 1974) which permits the calculation of trapping rates K for each trapped state. When a component with a lifetime comparable with the spin-averaged positronium lifetime (500 ps) is present in the spectrum, this can usually be interpreted as representing annihilation in voids. As a function of temperature the positron–void trapping rate $K(T)$ will generally increase from a value K_0 at $T = 0$, and in some cases $K(T)$ is observed to pass through a maximum, decreasing slightly as T is raised further. The curved shape and the reduction in $K(T)$ at high temperature can be ascribed to diffusion limitation of the positron flux near the void.

In order to extract information about the transition-dependent contribution to the trapping rate, K_{tr}, from the observed quantity $K(T)$, it is in principle necessary to take into consideration the time dependence of $K(T)$. This time dependence is caused by the transient depletion of the positron density near the defect (Seeger 1974). The influence of this effect should be evaluated in each case. Bentzon and Evans (1990) find that the transient behaviour contributes by less than 2% to the trapping rate in their studies of positron trapping at small voids (1 nm) in molybdenum. For an experiment on voids in aluminium Nieminen *et al* (1979) conclude that the time needed to reach the time-independent form of the positron flux is less than 5 ps. In such cases a good approximation to the transition-limited trapping rate is given by the interpolation formula (Brandt 1974),

$$K_{tr} = (K^{-1} - K_{diff}^{-1})^{-1} \tag{2}$$

where K_{diff} is the diffusion-limited trapping rate (Seeger 1974, Brandt 1974),

$$K_{diff} = 4\pi r n D_+. \tag{3}$$

Here r and n are the void radius and density, respectively, and D_+ is the positron diffusion coefficient

$$D_+ = D_{300}(300K/T)^{1/2} \tag{4}$$

with D_{300} being temperature independent. The description in terms of (3) and (4) is now experimentally well established (Bentzon and Evans 1990, Huomo *et al* 1987, Soininen *et al* 1990). In cases where the temperature dependence of the transition-limited contribution to the total trapping rate has been observed, it appears to obey the linear behaviour expressed by equation (1).

3. General model for positron trapping at voids

The mechanism of a positron being trapped at the interior of a void will be described as the sum of two independent processes (see figure 1):

(i) the direct inelastic transition of a positron with a wavevector k_+ to the captured volume or surface state at the void, the specific trapping rate being ν_i;

(ii) the elastic transmission of the positron through the potential step at the edge of the void, to a free volume state within the void, with the transmission rate ν_f, followed by a transition with the probability β to a captured state.

The specific trapping rate is then

$$K_{tr}/n = \nu_{tr} = \nu_i + \nu_e = \nu_i + \nu_f\beta \tag{5}$$

where ν_e represents the part initiated by elastic transmission, and n is the density of voids. In terms of cross sections this becomes

$$\nu_{tr} = v_+(\sigma_i + \sigma_f\beta) = 4\pi r^2\mu + v_+\sigma_f\beta. \tag{6}$$

Here v_+ is the velocity of the free positron in bulk metal, and μ is the notation for area specific inelastic trapping rate.

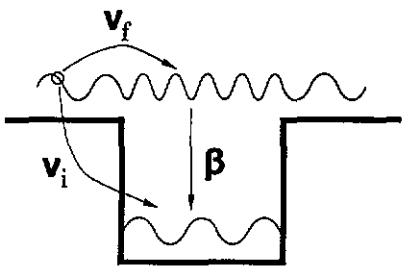

Figure 1. Mechanisms for positron trapping at voids: ν_i, the rate of direct, inelastic transition to a captured state; ν_f, the rate of elastic transmission to a free volume state; β, the probability of transition to a captured state.

3.1. Cross section

The quantum mechanical cross section for the absorption of a particle by a spherical object (a void) can be expressed as an infinite sum (Blatt and Weisskopf 1952)

$$\sigma_f = \pi\lambda^2\Sigma(2l+1)T_l \tag{7}$$

where l is the orbital angular momentum quantum number for the incident particle and $\lambda = k_+^{-1}$ represents its wavelength. The quantity T_l is the l-channel coefficient for the transmission of the particle to the interior of the void, and is a function of the wavenumber k_+, the void radius r, and the wavenumber K_+ of the particle inside the void. The variation of σ_f against k_+r has been computed by Blatt and Weisskopf (1952) for different values of K_+r.

We now consider the case where $K_+ \gg k_+$. This is relevant for positrons interacting with metal voids, since the kinetic energy of an untrapped positron inside a void is of the order of 1 eV, while k_+ represents thermal energies. We now assert that a useful approximation to the quantum mechanical particle–void cross section can be written

$$\sigma_f = \pi(\lambda^2 + r^2)T_0 \tag{8}$$

where T_0 is the penetration factor for positron transmission from bulk metal to the void interior. A more conventionally employed expression for the the cross section of

a particle approaching a spherical object is (Blatt and Weisskopf 1952, Brandt 1974, Nieminen *et al* 1979)

$$\sigma_t = \pi(\lambda + r)^2 T_0. \tag{9}$$

By comparison with the comprehensive calculations performed by Blatt and Weisskopf it is seen that equation (8) underestimates, while equation (9) overestimates σ_t. For $K_+ r = 11$ (corresponding to 1 nm sized voids and a binding energy of 3 eV) and $k_+ r = 0.1$, 0.3 and 0.8 (temperatures from 3–200 K), respectively, we find that the deviations from the correct numbers are 1, 10 and 22% by equation (8), while they are 22, 39 and 53% by equation (9).

We suggest that equation (8) may be useful for the interpretation of experimental data. It should be particularly suitable for small voids and low temperatures.

3.2. Penetration factor

For the penetration factor T_0 Blatt and Weisskopf employed the elementary expression for a square potential well, which is appropriate for the neutron–nucleus interaction. However, in the present case, the edge of a void as seen by a positron is not an infinitely sharp boundary; rather, the potential varies gradually on a scale comparable with a lattice constant, and over a distance corresponding to K_+^{-1}. Such a smooth boundary will reduce reflection and increase transmission.

We consider an incoming wave with amplitude A_i and a wavenumber k_+ approaching a potential step with a continuously diminishing potential $V(x)$. After passage of the step, a transmitted wave with amplitude A_t and wavenumber K_+ is propagating in the same direction as the incoming wave. An approximate expression for the amplitude ratio A_t/A_i can be obtained by considering $V(x)$ to consist of a number of rectangular steps, provided that the passage along any single step gives rise to a change of phase which is a small number compared with 1. The result can be written as

$$A_t/A_i = 2k_+/H(k_+, V(x)) \tag{10}$$

where $H(k_+, V)$ is a functional of the varying potential $V(x)$. In general, if $k_+ \ll K_+$, $H(k_+, V)$ is only weakly dependent upon k_+. If $V(x)$ is a single rectangular step one has $H = k_+ + K_+$.

The penetration factor is the ratio between the transmitted and incoming currents

$$T_0 = K_+|A_t|^2/(k_+|A_i|^2) = 4k_+K_+F/(k_+ + K_+)^2 \approx 4k_+F/K_+ \tag{11}$$

where we have introduced a 'potential step factor' $F \geqslant 1$. For a single rectangular step $F = 1$, while for an extended, smoothly varying potential one obtains $T_0 \to 1$.

3.3. Trapping rate

The positron's elastic transmission rate to a volume state in the void can now be expressed as

$$\nu_t = v_+\sigma_t = 4\pi\hbar(1 + (k_+ r)^2)F/m_* K_+ \tag{12}$$

where m_* is the effective positron mass in bulk metal. If k_+ is taken to represent a mean wavenumber corresponding to the thermal energy $3k_BT/2$, equations (5) and (12) can be combined to give

$$\nu_{tr} = \nu_i + \nu_{e0}(1 + \alpha_e T) \tag{13}$$

where $\nu_{e0} = 4\pi\hbar\beta F/m_* K_+$ and $\alpha_e = 3m_* k_B r^2/\hbar^2$.

We can now define $\alpha = \alpha_e \nu_{e0}/(\nu_i + \nu_{e0})$ which gives the trapping rate in the form of equation (1),

$$\nu_{tr} = (\nu_i + \nu_{e0})(1 + \alpha T). \tag{14}$$

The quantity β represents the rate of dissipation of the positron–defect binding energy. This factor was discussed by Brandt (1974) who argued that in a metal vacancy the energy dissipation will proceed rapidly by electron–hole excitations in the electron gas giving $\beta \approx 1$ in that case. Since the dissipation rate will depend on the overlapping of e^+–e^- wavefunctions β is expected to diminish with increasing void size.

4. Application to small voids

The molybdenum sample employed by Bentzon and Evans (1990) was studied by positron annihilation techniques (PAT) and transmission electron microscopy (TEM). The results are therefore very useful for a control of the proposed model. From TEM observations the values $r = 1.33 \pm 0.05$ nm and $n = (5.3 \pm 1.0) \times 10^{21}$ m^{-3} were established for the void content. PAT lifetime spectroscopy measurements were performed as a function of temperature for the same configuration of voids, both in a pure state, C(vac), decorated by hydrogen, C(H), and decorated with nitrogen, C(N). The data obtained by fitting the results with a three-state trapping model are given in table 1.

Table 1. Data for pure and decorated molybdenum voids as obtained by Bentzon and Evans (1990). Calculated values are based on $r = 1.33$ nm, $n = 5.3 \times 10^{21}$ m^{-3}, $m_* = 1.5m_e$, $K_+ = 8.9$ nm^{-1}.

Sample		C(vac)	C(H)	C(N)	
Observed:	K_0	(ns^{-1})	0.85 ± 0.07	1.75 ± 0.04	4.7 ± 0.3
	α	(kK^{-1})	9.3 ± 1.8	7.4 ± 0.6	7.2 ± 0.9
Calculated:	α_e	(kK^{-1})	9.0	9.0	9.0
	ν_i	(10^{-13} m^3 s^{-1})	< 0.3	0.59 ± 0.06	1.8 ± 0.4
	ν_{e0}	(10^{-13} m^3 s^{-1})	1.7 ± 0.5	2.7 ± 0.3	7.1 ± 1.3
	μ	(km s^{-1})	< 1.3	2.7 ± 0.3	8.0 ± 1.6
	$F\beta/K_+$	(nm)	0.17 ± 0.05	0.28 ± 0.03	0.73 ± 0.14
	$F\beta$		1.5 ± 0.5	2.5 ± 0.3	6.5 ± 1.2

The value for K_+ applied in the calculations for all three cases was obtained by using the value for the positron work function, $\Phi_+ = -3.0$ eV, measured by Huomo *et al* (1987).

The magnitude of α_e was calculated from the known value of r. The near equality of α (measured) and α_e (calculated) for pure voids demonstrates that ν_i is of negligible magnitude and that a measurement of α can provide information about the void size.

Conversely, if α and α_e are determined separately, ν_i can be computed. For pure voids, only an upper limit is obtained for ν_i, and also for impurity-containing voids ν_i contributes only a minor part to the total specific trapping rate ν_{tr}. This is seen to be the reason why Bentzon and Evans (1990) find values for α which are only weakly affected by void decoration.

The magnitude of ν_i can also be estimated by reference to a calculation by Nieminen *et al* (1979), who obtain $\nu_i = 0.6 \times 10^{-11}$ m^3 s^{-1} for voids with a radius of 25 nm in aluminium. If μ is of similar magnitude in molybdenum and aluminium, one obtains $\nu_i = 4\pi r^2 \mu \approx 10^{-15}$ m^3 s^{-1} for the present case, which is negligible in comparison with the observed ν_{e0}. It is interesting that the inelastic trapping rate increases to small but significant magnitudes as the voids are decorated with impurity atoms. This is consistent with results from positron beam experiments, showing that impurities at surfaces can be the cause of inelastic positron re-emission (Schultz and Lynn 1988).

The elastic trapping rate ν_{e0} also increases significantly when the voids are decorated. This is ascribed to changes in the factor $F\beta/K_+$, where K_+ is given by the positron work function. In decorated voids it is conceivable that all of the three factors F, β and K_+ can differ from the pure case. Assuming that variations in K_+ are small, values for $F\beta$ are given in table 1. Since the probability $\beta \leqslant 1$ the 'potential step factor' F is apparently of the order of 10. When the voids are decorated the shape of the metallic surface remains the same, but it is not known to what extent adsorption of impurity atoms can influence the penetration factor. If F is nearly constant, the data in table 1 indicate that the energy dissipation, as expressed by variations in β, is favoured by impurities.

A molybdenum sample prepared by neutron irradiation followed by annealing at a moderate temperature (150 °C) shows the longest positron lifetime $\tau_3 = 370$ ps, and $\alpha = 1.0(\pm 0.2)\ kK^{-1}$ (Bentzon 1987, Bentzon *et al* 1985). A calculation using the present model, assuming $\nu_i \ll \nu_{e0}$, gives $r = 0.44(\pm 0.07)$ nm. According to Puska and Nieminen (1983) the lifetime 370 ps should correspond to a vacancy cluster containing about 12 vacancies, i.e. $r \approx 0.36$ nm.

5. Application to large voids

In the case of large voids ($r \geqslant 10$ nm) we have $\nu_i = 4\pi r^2 \mu \approx 10^{-12}$ m^3 s^{-1}, if μ is of the order of 1 km s^{-1}. Further, $\nu_{e0} = 4\pi\hbar\beta F/m_* K_+ \approx 10^{-13}\beta F$ m^3 s^{-1}, with $F \geqslant 1$ and $\beta \leqslant 1$. The transition requires dissipation of the positron–void binding energy, and in an empty void this can only take place at the surface. Thus for large voids one expects $\beta \ll 1$, and accordingly $\nu_i \gg \nu_{e0}$. At room temperature $\lambda \approx 1$ nm, and the geometrical part of the cross section will dominate. Good approximations can be expected by use of equation (8) while it should be kept in mind that accurate data might require a more precise cross section formula. We thereby get

$$\nu_{tr} \approx \nu_i + \nu_{e0}\alpha_e T \tag{15}$$

which is the equation employed by Nieminen *et al* (1979) for the discussion of large voids in aluminium. In the large void approximation one can also write

$$K_{tr} = n\nu_{tr} = K_0(1 + \alpha T) \approx K_0[1 + (\nu_{e0}\alpha_e/\nu_i)T] \tag{16}$$

and we notice that in this case the measured parameter α can be expressed as $\alpha = 3k_B\beta F/K_+\hbar\mu$, which is not explicitly dependent on r. We may therefore expect α to provide a direct measure of $\beta F/K_+$.

A calculation of the quantities r and n can be performed on the basis of separate determinations of K_0 and K_{diff}, and a reasonably accurate knowledge of $D_+(T)$ and μ. The idea of extracting information from the K_{diff} term was probably first presented by Linderoth (1984), who used the results of Linderoth *et al* (1985) to extract K_{diff}, and, by applying values for r and n determined by TEM, Linderoth estimated D_+.

There exist a number of experimental studies of positron trapping in the presence of large voids in aluminium (Petersen *et al* 1976, Nieminen *et al* 1979, Linderoth *et al* 1985, Eldrup and Jensen 1987). In all of these cases the dependence of trapping rate upon temperature appears to be consistent with equations (1) and (4). However, we shall not here attempt a detailed analysis of these aluminium data in terms of our model, since it seems that the combination of experimental uncertainties and insufficient knowledge of the physical parameters makes a consistent analysis ambiguous.

6. Conclusion

The model presented here is general in the sense that the expression for the transition-limited positron trapping rate, as expressed by equations (5), (6) and (7) is valid for all void sizes and at all temperatures. The basic idea is that the trapping is assumed to follow two independent routes: (i) a direct inelastic transition into the captured state; or (ii) an elastic transmission from a bulk state into a free volume state, eventually followed by trapping with a probability β. For applications not requiring a high degree of accuracy we suggest that the approximate cross section formula (8) is applied. This approximation is consistent with the linear temperature dependence $K_{tr} = K_0(1+\alpha T)$, observed in several experimental surveys.

It is shown that, to first order, the coefficient α is simply related to physical properties of the void ensemble. In particular, the expression for α_e contains only natural constants and the void radius. The model was discussed by application to different sets of experimental data. It should be noted that for the extraction of experimental values for K_{tr} it may be necessary to consider whether equation (2) should be replaced by a more accurate relationship, comprising the time dependence of the positron distribution.

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