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1992 J. Phys.: Condens. Matter 4 1973

(http://iopscience.iop.org/0953-8984/4/8/012)

J. Phys.: Condens. Matter 4 (1992) 1973–1980. Printed in the UK

Low-temperature positron trapping into voids in metals

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Received 10 September 1991

Abstract. We present a theoretical analysis of positron trapping into large voids in metals. We show that, even if the trapping rate vanishes as the positron energy goes to zero, a significant amount of trapping of non-thermalized positrons ensures a substantial trapping probability even at very low temperatures.

1. Introduction

The construction of low-energy positron beams has made a whole new range of techniques accessible for studying surfaces and defects near surfaces [1]. It has also led to an increased insight into the fascinating and complex interaction of positrons with surfaces. A positron implanted at a low energy into a metal can return to the surface after losing energy to electronic and vibrational excitations. The positron may become reflected by the surface potential, it can be emitted as a free positron or combined with an electron as a positronium (Ps) atom, or it can make the transition into a localized positron surface state [1]. The surface state is the result of the repulsion from the ion cores of the solid and the attractive image correlation potential which lead to a potential well immediately outside the surface in which the positron may become localized [2, 3]. The positron beams have made it posible to study the probabilities for each of these channels experimentally. The low-temperature behaviour is particularly interesting. For many metal surfaces the positron work function is negative, i.e. classically the positron can always be emitted from the surface [1]. However, because of quantum effects the surface potential can reflect the positron and, as the positron kinetic energy E_{kin} goes to zero, the reflection becomes total, i.e. no positrons escape. Thus, as the sample temperature T is lowered towards 0 K, the probability that thermalized positrons, for which $\langle E_{kin} \rangle = \frac{3}{2} k_B T$, escape the surface should vanish. The effect was predicted some time ago [1] but it was only recently confirmed experimentally for Cu and Al when the effects of non-thermalized positron emission were taken duly into account [4]. The experiments showed that both free-positron emission and Ps formation vanish as the positron kinetic energy goes to zero.

Recently, we have given a theoretical analysis of the transition rate into the surface state [5] and we showed that this rate also goes to zero for E_{kin} going to zero when the transition is calculated using the Fermi golden rule, since the overlap between the initial positron state and the final localized state vanishes as the surface becomes totally reflecting. Thus, according to this result, all surface processes vanish at $E_{kin} = 0$ and the positron is always reflected back into the metal. There is litle evidence for or against

0953-8984/92/081973 + 08 $04.50

this hypothesis from the positron beam experiments owing mainly to the difficulty of separating positrons annihilating in the bulk from those annihilating from the surface state. Britton *et al* [4] assumed a temperature-independent surface state transition rate ν_s. They obtained a large uncertainty in the value obtained from the data analysis and it does not seem possible to deduce the temperature dependence of ν_s reliably. However, there are positron lifetime studies of bulk metals, e.g. Al [6–8], Cu [9] and $Fe_{59}Ni_{25}Cr_{16}$ [10], containing large voids which clearly show that positrons annihilate from the surface state in the voids even for sample temperatures approaching 0 K. These results thus seemingly contradict the theoretical predictions. The purpose of the present paper is to examine whether the void results can be reconciled with the picture of the positron–surface interaction given above. We shall in particular focus on the role of trapping of non-thermalized positrons and we show that non-thermal trapping may dominate at low temperatures and thus explain the observed results.

The above discussion refers to positrons trapping into large voids, where reflection at the surface must be taken into account when considering the trapping rates at low temperatures. Positron trapping rates have been measured for small voids in Mo [11–13] i.e. voids whose radius (about 2 nm) is much smaller than the positron de Broglie wavelength at low temperatures. Here, the positron state extends throughout the void [13], and it is hard to see how reflection at the void surface can bring the trapping rate into the void to zero at low temperatures. Accordingly, the work presented here does not contradict the discussion of temperature-dependent trapping rates given by Bentzon and Evans [11] as we show in more detail in section 2 below.

In the next section we give an overview of our picture of positron interactions with metals containing voids and describe the details of our calculations. The results are presented and discussed in section 3, and section 4 summarizes our conclusions.

2. Theoretical model

2.1. Positron interactions with voids

Positrons trapped in large voids normally annihilate from the surface state [14]. This is indicated by angular correlation of annihilation radiation (ACAR) spectra [15] for metals containing voids that are similar to the spectra measured for positrons trapped at external surfaces [16]. It is also suggested by the fact the surface state in most cases is the ground state of the metal–positron system [1]. Only in a few exceptional cases have signatures of Ps-like states been observed in ACAR spectra [17–19].

The trapping can occur directly from the bulk into the surface state. It can also take place when the positron enters the void either as a free positron or as a Ps atom and makes the transition into the surface state in a subsequent interaction with the void surface (note that the positron may also escape the void and re-enter the bulk of the metal if its energy is less than the work function for the void surface). In all cases the positron must penetrate the surface of the void. Thus, for a perfectly reflecting surface, i.e. for T approaching 0 K, the trapping rate should go to zero, according to the picture outlined in the preceding section.

The finite size of voids means that there is a qualitative difference between trapping in voids and transmission through external surfaces. This difference is illustrated by the analysis of the specific trapping rate by Nieminen *et al* [6] who employed the analogy to

neutron capture by atomic nuclei [20]. They wrote the specific positron trapping rate of a cavity as

$$\nu_T \simeq \pi(\hbar p/m)(R + \lambda\!\!\!^{-})^2\, 4pK/(p + K)^2 \tag{1}$$

where $\hbar p$ is the positron momentum, $\lambda\!\!\!^{-} = 1/p$ its wavelength, R is the cavity radius and K is the wavevector of the positron state inside the cavity. Equation (1) is only a simple approximation but it can be used to examine the qualitative variation in the trapping rate for different cavity sizes. When R is much larger than $\lambda\!\!\!^{-}$, $R + \lambda\!\!\!^{-} \simeq R$. In this limit, ν_T is proportional to the transmission coefficient $4pK/(p + K)^2$ and vanishes as p goes to zero. For small cavities, on the other hand,

$$\nu_T \simeq \hbar/\pi mK \qquad R \ll \lambda \quad p \ll K \tag{2}$$

which means that ν_T is independent of p. Surface reflection thus is important only when $\lambda\!\!\!^{-}$ is comparable with or smaller than the void size. For a thermalized positron,

$$\lambda\!\!\!^{-} = \hbar/(3mk_BT)^{1/2} = 17\ \text{nm}/[T(K)]^{1/2}. \tag{3}$$

This means that surface reflection is important at all temperatures above 1 K for 25 nm radius voids such as those studied in Al [6–8] and $Fe_{59}Ni_{25}Cr_{16}$ [10], while for the smaller voids studied in Mo (radii, about 2 nm) [11–13] the influence of surface reflection is reduced owing to the small void sizes below about 100 K. The cavities in the Cu samples in [9] are of micrometre size which means that the cavity surfaces are equivalent to planar surfaces at all experimentally attainable temperatures. Thus, the small void sizes in Mo may explain why the trapping rate does not vanish at low T but, for the other systems, alternative explanations must be found.

In bulk positron studies the positrons are injected into the sample at a high energy (in the case of lifetime experiments from a radioactive source) and lose energy by exciting the solid. As the positrons approach electronvolt energies, which takes less than 1 ps [1], the spatial distribution is slowly varying on length scales much shorter than the mean implantation depth which is of the order of 0.1 mm. As time goes on and the positrons approach thermal energies, trapping into the voids starts to become important and the positron distribution becomes increasingly depleted near the void surfaces. Eventually, a spatial distribution of thermalized positrons develops with a depletion near the void surfaces determined by the loss of diffusing positrons due to trapping [6]. The transient time before this spatial distribution is obtained is normally shorter than positron lifetimes but longer than thermalization times [6].

The trapping rate is governed both by the diffusion of the positrons to the void surfaces and by the quantum mechanical transition from the bulk state to a trapped void state. The slower of these processes is the rate-limiting process. After the steady state distribution is established the specific trapping rate can by expressed phenomenologically as [8, 21]

$$\nu = (\nu_D^{-1} + \nu_T^{-1})^{-1} \tag{4}$$

where ν_D is the specific trapping rate for purely diffusion-limited trapping and ν_T is the purely transition-limited specific trapping rate. The diffusion-limited rate is given by $\nu_D = 4\pi D_+ R$ where D_+ is the positron diffusion constant and ν_T is a product of the void surface area and the transition rate μ per unit area: $\nu_1 = 4\pi R^2\mu$. Note that μ here, in the limit of a large void radius, is equal to the transition rate calculated in [5].

In the transient time region, ν is higher than expressed by equation (4) since the positron density is less depleted near the void surfaces. At very short times before any

significant depletion has occurred, $\nu \simeq \nu_T$. This means that the trapping rate during thermalization is higher than for thermalized positrons, not only because ν_T normally increases with increasing positron momentum [5] but also because the trapping is less limited by the motion of the positron to the trap.

In the section 2.2 we describe a theory that allows us to evaluate the trapping probability including the contribution from both non-thermalized and thermalized positrons.

2.2. Calculations; the Boltzmann equation

We obtain the momentum distribution of positrons in the sample by solving the Boltzmann equation for the positron slowing down in a homogeneous medium for a steady flux of incoming positrons [22]:

$$\int \mathrm{d}q\, K(q,p)f(q) - [H_{sc}(p) + \lambda + \kappa(p)]f(p) + f_i(p) = 0 \tag{5}$$

where $\hbar p$ is the positron momentum, $f(p)$ the steady state momentum distribution, $f_i(p)$ is the momentum-dependent rate of incoming positrons, λ is the bulk positron annihilation rate, $\kappa(p)$ is the momentum-dependent trapping rate (we assume no detrapping), and the functions K and H_{sc} are defined as

$$K(q,p) = p^2 \int \mathrm{d}\theta_q \sin\theta_q \int \mathrm{d}\varphi_q\, R(q,p) \tag{6}$$

(θ_q and φ_q are the directional angles of q) are

$$H_{sc}(p) = \int \mathrm{d}^3q\, R(p,q) \tag{7}$$

where $R(p,q)\,\mathrm{d}^3q$ is the scattering rate from momentum state $\hbar p$ to states in the volume $\hbar^3\,\mathrm{d}^3q$ around $\hbar q$. Both positron–electron and positron–phonon scattering are included in $R(p,q)$ and we use parameter values corresponding to Al described in [22]. Equation (5) is solved using an iterative technique [22]. The probability that the positron is trapped into a void and consequently annihilating from a trapped state is given by

$$N = \bar{\kappa}/(\lambda + \bar{\kappa}) \tag{8}$$

where the average trapping rate $\bar{\kappa}$ is given by

$$\bar{\kappa} = \int \mathrm{d}p\, \kappa(p)f(p) \Big/ \int \mathrm{d}p\, f(p). \tag{9}$$

For $\kappa(p)$ we use (cf equation (4))

$$\kappa(p) = C[\nu_D(T)^{-1} + \nu_T(p)^{-1}]^{-1} \tag{10}$$

where C is the defect concentration, and ν_D and ν_T are given by

$$\nu_D(T) = 4\pi R D_0 (T/300\ \mathrm{K})^{-a} \tag{11}$$

and

$$\nu_T(p) = 4\pi R^2 \gamma(\hbar^2 p^2/2m). \tag{12}$$

The expression for ν_D is obtained by assuming that the temperature dependence of the

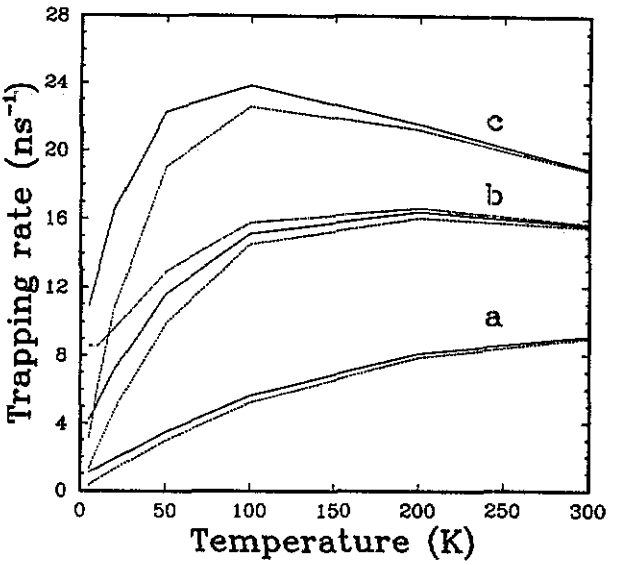

Figure 1. Calculated average trapping rate in voids as a function of temperature. The broken lines correspond to thermal trapping only and the full curves to the result from the full solution of the Boltzmann equation. The annihilation rate is in both cases taken to be the bulk Al value, $\lambda_{Al} = 6.135\ ns^{-1}$. The sets of curves a, b and c correspond to different value of γ: curves a, $\gamma = 7.7 \times 10^4\ m\ eV^{-1}\ s^{-1}$; curves b, $\gamma = 3.1 \times 10^5\ m\ eV^{-1}\ s^{-1}$; curves c, $\gamma = 7.7 \times 10^5\ m\ eV^{-1}\ s^{-1}$. The chain curve was obtained from the Boltzmann equation assuming an effective annihilation rate $\lambda_{eff} = 3\lambda_{Al}$ to take into account trapping into defects other than voids. For all curves $D_0 = 1.60 \times 10^{-4}\ m^2\ s^{-1}$, $\alpha = 0.6$, $R = 45$ nm and $C = 2.5 \times 10^{20}\ m^{-3}$.

positron diffusion constant is $D_+(T) = D_0(T/300\ K)^{-\alpha}$ where D_0 is a constant [23, 24]. The form used for ν_T assumes that the trapping rate per unit surface area is proportional to the energy with a proportionality constant γ. This behaviour is predicted by equation (1) above for large R and small p. It also agrees with the energy dependence of the surface state trapping rates calculated in [5] and with the experimentally deduced surface transmission rates [4]. The reasoning behind equation (10) is that the transition-limited trapping rate is determined by the momentum of the positron as it strikes the surfaces, while the diffusion-limited rate is determined by the diffusion constant for thermal positrons. Hence, we assume that ν_T depends on p but not T and ν_D on T but not p. Equation (10) thus gives the correct temperature dependence for the trapping rate for thermalized positrons, since the average positron momentum in this case is determined by the temperature but also takes into account, in an approximate way, the fact that the trapping rate is higher for non-thermalized than for thermalized positrons. We only expect our model to be applicable when there is a well defined trapping rate for thermalized positrons, which means that the transient time referred to in section 2.1 must be significantly smaller than the positron lifetime. This restriction would not hold if we solved the steady state Boltzmann equation that allowed for spatial variation in the positron distribution.

We use the following parameter values: $D_0 = 1.60 \times 10^{-4}\ m^2\ s^{-1}$, $\alpha = 0.6$ [24], $R = 45$ nm and $C = 2.5 \times 10^{20}\ m^{-3}$. The values of R and C are typical for neutron-irradiation-induced voids in Al [8]. The correct value of the parameter γ is somewhat uncertain and

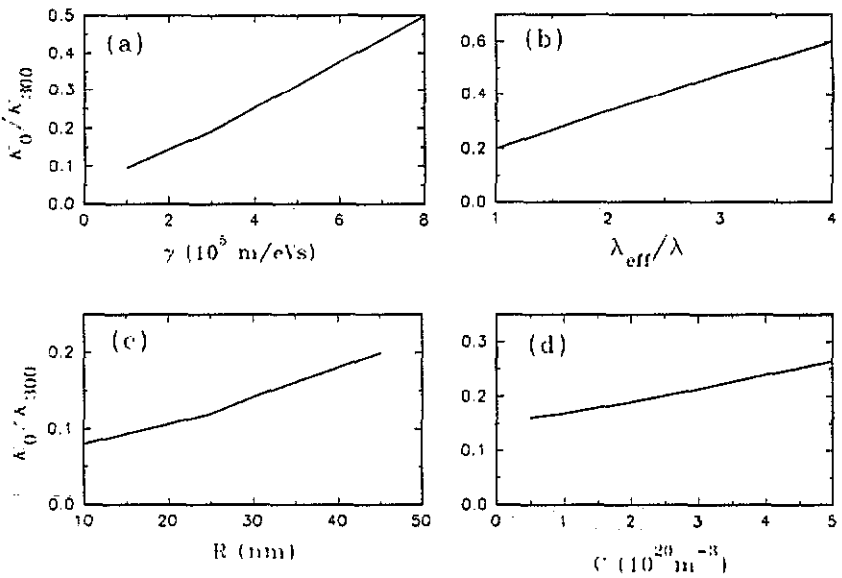

Figure 2. The dependence of $\bar{\kappa}_0/\bar{\kappa}_{300}$ on (*a*) the surface state transition rate γ, (*b*) the effective annihilation rate λ_{eff} and (*c*) the radius R and (*d*) the concentration C of cavities. Only one parameter is varied at a time. The three fixed parameters were taken from the following set of values: $\gamma = 3.1 \times 10^5$ m eV^{-1} s^{-1}, $\lambda_{eff}/\lambda = 1$, $R = 45$ nm and $C = 2.5 \times 10^{20}$ m^{-3}.

is likely to vary considerably depending on the properties of the cavity surfaces (e.g. impurity coverage). Typical values deduced from the temperature dependence of trapping rates into voids are of the order of (1–5) $\times 10^5$ m eV s^{-1} [6, 8, 10]. A value of 3×10^5 m eV s^{-1} is given for γ in [3].

3. Results and discussion

The average trapping rates $\bar{\kappa}$ for three different values of γ are shown as functions of temperature in figure 1. The results were obtained from the Boltzmann equation (5) using the momentum-dependent trapping rates given by equation (10). The figure also shows the average trapping rates for thermalized positrons, i.e. positrons with a thermal Maxwell–Boltzmann distribution. The behaviour of $\bar{\kappa}$ at low temperatures is markedly different for the two sets of curves. For thermal positrons, $\bar{\kappa}$ goes to 0 linearly as T goes to 0 K. However, when non-thermal effects are included, $\bar{\kappa}$ approaches a finite value $\bar{\kappa}_0$ at 0 K, in accordance with the experimental observations [6, 7, 10].

Figure 1 shows that the average trapping rate increases at low T (transition-limited regime) and decreases at high T (diffusion-limited regime) for all values of γ. However, the detailed shapes of the $\bar{\kappa}(T)$ curves depend on the value of γ. For low γ the transition-limited regime extends to higher temperatures and the ratio $\bar{\kappa}_0/\bar{\kappa}_{300}$ ($\bar{\kappa}_{300}$ is the value of $\bar{\kappa}$ at 300 K) is smaller than for high values of γ. The dependence of $\bar{\kappa}_0/\bar{\kappa}_{300}$ on γ is illustrated in figure 2(*a*).

Other species of defect, e.g. dislocations [7, 19], are often created when voids are introduced in metals by irradiation. Trapping into these defects enhances non-thermal

trapping into the voids since fewer positrons will thermalize before annihilating or being trapped. We examined this effect by replacing the bulk annihilation rate λ by a higher effective removal rate λ_{eff} in the calculations. The chain curve in figure 1 shows a result for $\lambda_{\text{eff}} = 3\lambda$, corresponding to a trapping rate into defects other than voids of 2λ which is a typical value at low temperatures for neutron-irradiated Al [7]. It is seen that the higher λ_{eff} produces a considerable enhancement in $\bar{\kappa}$ at low temperatures. This is also demonstrated by figure 2(*b*), showing the dependence of low-temperature trapping on the value of λ_{eff}.

Figures 2(*c*) and 2(*d*) show the variation in the low-temperature trapping rates with void size and concentration, respectively, which is seen to be smaller than the variation with γ and λ_{eff}.

Experimentally $\bar{\kappa}_0/\bar{\kappa}_{300}$ is found to range from about 0.1 for $Fe_{59}Ni_{25}Cr_{16}$ [10] to about 0.7 for Al [7, 19]. Figure 2 shows that values in this range can be obtained from the present model with suitable choices of parameters. The model can thus explain the qualitative temperature behaviour of trapping rates into voids. We do not attempt to make a detailed quantitative comparison between model and experiment owing to the approximate nature of the model. Our main concern is to demonstrate the importance of non-thermal trapping into voids at low temperatures and to show that, even if the trapping rate for thermal positrons goes to zero at 0 K, a substantial fraction of the positrons may still become trapped.

The experimental results for micrometre-sized cavities in Cu [9] are qualitatively different from those for 10–50 nm voids in other systems since the intensity of the trapped positron component shows very little temperature variation with a slight increase with decreasing temperature, in contrast with the decrease observed in other systems. However, a number of features distinguish this system from the others. Firstly, the low concentrations of the cavities mean that the transient time before a constant trapping rate is obtained (see section 2.1) is much longer than the positron lifetime. The trapping cannot therefore be described by the present model and requires a more detailed description of the coupled positron transport in real space and momentum space. Secondly, the cavities contain Kr condensed on the surfaces at low temperatures which may change the positron–surface interaction.

4. Conclusion

We have demonstrated that non-thermal trapping leads to a substantial enhancement of positron trapping into voids at low temperatures compared with thermal trapping rates. It means that, even if the trapping rate for thermal positrons vanishes at 0 K, as has been predicted theoretically [5], non-thermal trapping ensures a finite trapping probability and may therefore explain the non-vanishing trapping rates observed experimentally.

Acknowledgments

KOJ would like to thank the Commission of the European Communities for a Research Grant under the Stimulation Action scheme contract SCI-0163.

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