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Positron trapping rates in metal single crystals at low temperatures

G Trumpy† and K Petersen‡

† Department of Electrophysics, Technical University of Denmark, DK-2800 Lyngby, Denmark

‡ Department of Physics, Technical University of Denmark, DK-2800 Lyngby, Denmark

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Abstract. For a number of single-crystal metal samples with small defect density, we have observed 'anomalous' low-temperature behaviour of positron trapping, previously seen in certain cases of metals prepared with various contents of defects. In general, the trapping rate is found to be a linear function of $T^{-1/2}$. It is suggested that an explanation for this effect can be given on the basis of the quantum mechanical cross section between positrons at low velocity, and point defects.

1. Introduction

Lifetimes for positrons in metals at low temperatures have previously been observed in a few cases (Herlach *et al* 1977, Smedskjaer *et al* 1980a, Kupca *et al* 1980, Shirai *et al* 1986, Hidalgo *et al* 1987). The results scarcely reflect uniform behaviour, although it has generally been found that one lifetime of medium length (typical for vacancies) appears with increasing intensity as the temperature is lowered below about 100 K. Parallel behaviour has been observed in the temperature dependence of annihilation lineshapes (Rice-Evans *et al* 1978, Herlach *et al* 1977). In most of the cases reported, the metal samples had a large, and sometimes complicated, defect content.

In order to carry out a further study of this particular low-temperature effect, we have obtained positron lifetime spectra at temperatures between 10 and 300 K for a number of pure, single-crystal metal samples, presumably containing a relatively small number of defects. Qualitatively, it appears that the low-temperature effect prevails when the defect concentration is small. A large number of theoretical models have been published on the calculation of positron trapping rates at defects in solids: however, none of them seem to give an adequate description of the observed low-T dependence. Without making detailed calculations we shall propose that a combination of simple assumptions and available models could provide a satisfactory description.

2. Experimental method

Positron lifetime spectra have been obtained at temperatures between 10 and 300 K for single-crystal samples of aluminium, copper, nickel, zinc and molybdenum. The purities of the metals were: Al(6N = 99.9999%), Cu(5N), Ni(5N?), Zn(4N), Mo(4N). The samples were circular discs, about 0.5 mm thick, spark cut from single-crystal metal rods of about

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8 mm diameter. The discs were electrolytically etched and polished. The defect content was not known. On the basis of the observed positron trapping rates, the concentration of vacancy-like defects can be estimated to be of the order of 10^{-6}. At this density there is no other method available for determination of the defect content. In some cases, the samples were annealed for 3–4 hours in order to reduce the defect content. The source–sample unit was a sandwich consisting of two discs enclosing a ^{22}Na positron source, and was mounted in a helium-flow vacuum cryostat.

Positron lifetime spectra were recorded by a conventional fast–fast lifetime spectrometer with a time resolution corresponding to 250 ps FWHM. The spectra obtained were analyzed by use of the program PATFIT-88 (Kirkegaard *et al* 1989). In two-component fits, the longer lifetime, τ_2, was generally of the same magnitude as the typical value of positron annihilation in vacancies of the respective metals. Therefore, in the final analysis this component was constrained to a constant value.

3. Experimental results

In figure 1 we show the results for an aluminium single-crystal sample before heat treatment, of determinations of positron lifetime spectra analysed with the longer lifetime τ_2 constrained to be 240 ps. The decrease in intensity I_2 with increasing temperature is typical of all the samples studied. It appears that at low temperatures, positron trapping to defects provides an essential contribution to the lifetime spectra, even in cases of very pure and well annealed metals.

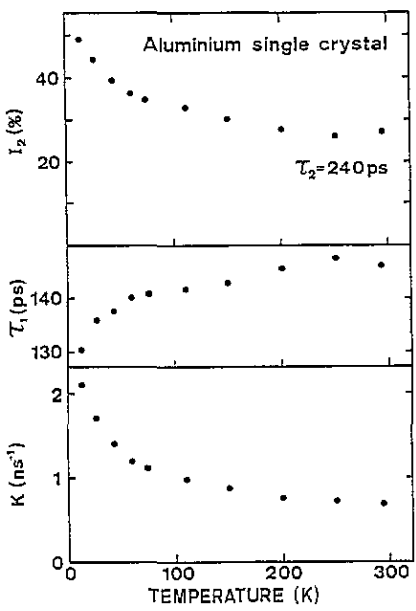

Figure 1. Observed data for non-annealed single-crystal aluminium. Calculated values for trapping rate K.

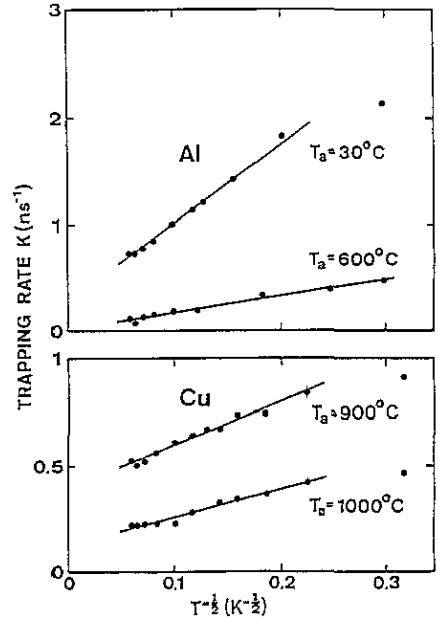

Figure 2. Trapping rates in single crystals of aluminium and copper, annealed at different temperatures, plotted as a function of $T^{-1/2}$.

The simple two-state trapping model (Brandt 1967, West 1979) was employed to compute the transition rates K to the positron traps, having the properties of vacancy-like defects. For these calculations it was assumed that the positron lifetime in bulk metals, τ_b, was a slowly varying, linear function of T (Stott and West 1987, McKee and McMullen 1978). In figure 1 the values obtained for K are also presented.

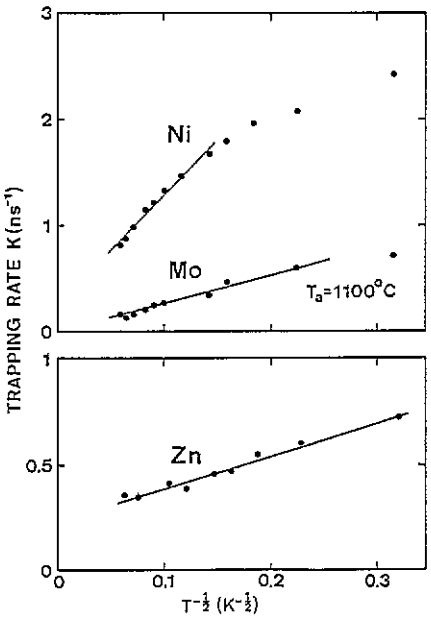

Figure 3. Trapping rates in single crystals of nickel, molybdenum and zinc, plotted as a function of $T^{-1/2}$.

Upon inspection of the trapping rate data, it appears that $K(T)$ is described quite well by a linear function of $T^{-1/2}$ within a wide temperature range, for all the samples studied. In order to visualize this behaviour, the rates obtained, $K(T)$, have been plotted as a function of $T^{-1/2}$ in figures 2 and 3. The straight lines drawn represent the function

$$K(T) = a + bT^{-1/2} \tag{1}$$

where a and b are constants. Numerical values for a and b have been listed in table 1. If n is the density of defects, the 'specific trapping rate' is

$$\nu(T) = K(T)/n = \nu_0 + \nu_1(T) \tag{2}$$

where ν_0 is a constant and $\nu_1(T)$ represents the $T^{-1/2}$ dependence. For the samples of aluminium and copper, a change in the concentration of defects was produced by thermal annealing. As seen in figure 2, the linear dependence of equation (1) is also valid after a reduction of the defect density.

4. Discussion

Since the obtained positron spectra do not contain any components with lifetimes exceeding the single-vacancy lifetime, τ_v, the presence of divacancies or larger vacancy clusters,

Table 1. Lifetimes from analysis, and parameters obtained for equation (1).

Sample	τ_b (300 K) (ps)	τ_2 (ps)	Ann. temp. T_a (°C)	a (±0.05) (ns^{-1})	b (±0.5) (ns^{-1})
Al	164.5	240	30	0.27	7.3
			600	0	1.5
Cu	110.7	180	900	0.39	2.0
			1000	0.13	1.2
Ni	109.8	190	30	0.25	10.4
Mo	109.5	190	1100	−0.04	2.7
Zn	145	240	30	0.23	1.5

including voids, can be excluded. Also, the density of grain boundaries must be negligible, since the samples have been cut from single crystals. However, single vacancies are not expected to exist as free defects at room temperature in the metals considered here (except possibly in molybdenum). It is more likely that the vacancy-like defects observed are bound to dislocation loops. The defects observed in our samples have been created during growth of the single crystals, or during cutting and handling at 300 K. At this temperature, free vacancies and interstitials have been free to migrate and form agglomerations, which are usually known to result in loops consisting of closed dislocation lines. In a loop, any interruption of a rectilinear dislocation constitutes a point defect, such as a jog, which can have a vacancy-like free volume, and can give rise to a positron decay rate close to τ_v^{-1}. The same conclusion has been drawn earlier from positron lifetime studies on aluminium and molybdenum samples that had been neutron-irradiated and annealed (Pagh *et al* 1984, Bentzon *et al* 1985, Linderoth *et al* 1985, Eldrup and Jensen 1987, Bentzon and Evans 1990). In those cases, long- and intermediate-lifetime components were identified as the results of positron trapping at voids and at dislocation loops, respectively. The trapping rate at loops has a negative temperature dependence which is particularly pronounced below 150 K.

Shirai *et al* (1986) observed positron lifetime spectra from monocrystalline cadmium which had been quenched to a low temperature, so the sample would presumably contain monovacancies only. Also in that case, a trapping rate with negative temperature coefficient was observed.

In the case of nickel, the straight line is not followed below $T \simeq 100$ K in the plot of figure 3. This effect may be caused by processes competing with positron trapping at vacancy-type defect. For instance, a temperature-dependent transition of positrons to 'shallow traps', in which the positron's lifetime is close to τ_b, could provide an explanation for non-linearity of the nickel data. Dislocation lines are probable candidates for shallow traps.

More often, a deviation from the linearity is seen only at low (<20 K) temperatures. Shallow traps could also be responsible here, but in these cases an alternative explanation could be that the positrons are not thermalized before annihilation. Calculations of thermalization times for aluminium by Nieminen and Oliva (1980) give 17 ps for a sample at 100 K and 64 ps at 15 K. This indicates that the average positron temperature at annihilation can be considerably higher than the ambient temperature when T is low, explaining why the trapping rate at 10 K in some cases is lower than that expected from the linear relationship (1). In this case, the effect cannot be avoided in the standard experimental procedure. Experiments with low-energy positron beams may give some useful information concerning effects at the lowest temperatures.

5. Theoretical models

Among the numerous calculations concerning trapping of positrons at defects, none seem to be directly applicable for an interpretation of the empirical relation (1). We will comment briefly on the different approaches:

(A) *The Golden Rule calculation* (Hodges 1970, Bergersen and Taylor 1974, McMullen 1977, 1978, Bergersen and McMullen 1977, Nieminen and Laakonen 1979, Puska and Manninen 1987). The initial state of the positron is considered to be, either a delocalized wave in the perfect lattice of the solid, or an 'untrapped state' (or 'scattering state') in the region of a defect. Transition to the final bound positron state in the potential well of the defect takes place by creation of an electron–hole pair, or, less likely, by phonon creation. Calculations have generally been performed by application of the Golden Rule. For trapping of thermal positrons into vacancy-like defects the consensus if that the process is not, or only weakly, temperature dependent.

(B) *The cross section model.* In analogy with the theory for scattering and trapping of thermal neutrons at nuclei (Blatt and Weisskopf 1952), the thermal positron cross section with respect to a small attractive defect of radius R can be written as

$$\sigma_+ \simeq \pi(\lambda\!\!\!^{-} + R)^2 T\beta \tag{3}$$

for s waves. Here $\lambda\!\!\!^{-} = k^{-1}$ is the positron's reduced wavelength and T is a penetration coefficient. The factor β is the probability that the positron will be transferred to a bound state while it is inside the potential well. In a metal, the energy is dissipated to electron–hole pairs, which is a fast process, and one may set $\beta \simeq 1$ (Brandt 1974). If the positron's potential energy changes abruptly at the edge of the defect, we have $T = 4kK/(K + k^2)$, where K is the wavenumber of the positron in the potential well. For low and intermediate temperatures ($T \leqslant 300$ K) the radius of a vacancy-type defect is negligible in comparison with λ. Also, for a potential well depth of order 1 eV, one can set $k \ll K$. With these conditions, the specific trapping rate for positrons with velocity v_+ becomes

$$\nu_+ = v_+\sigma_+ \simeq 4\pi\hbar/mK \tag{4}$$

which is independent of temperature.

The cross section model has also been used to describe positron trapping at large voids, for which the empirical positron trapping rate has been obtained as $\nu = \nu_0 + \nu_1 = \nu_0 + \gamma T$, where γ is a constant (Nieminen *et al* 1979, Jensen and Walker 1992, Trumpy and Bentzon 1992). The second term can be interpreted by assuming $\lambda\!\!\!^{-} \ll R$, so that $\sigma \simeq \pi R^2$, and $\nu_1 \propto k^2 \propto T$. The first term, ν_0, could represent a more direct process (Nieminen *et al* 1979, Trumpy and Bentzon 1992), possibly due to the overlap between the incoming positron wave and the exponentially decreasing tail of the bound positron state outside the potential well. Jensen and Walker (1992) provide an alternative explanation for ν_0, based upon the thermal distribution of the positron velocities.

(C) *The 'cloudy crystal ball' model.* Also in analogy with nuclear reactions, Shirai and Takamura (1989) considered the positron–defect interaction in terms of a complex potential, where the real and imaginary parts give rise to the untrapped state and the absorption process, respectively. The results of calculations show a strong negative temperature coefficient for the trapping rate, provided that the cross section is dependent upon resonances in the positron–defect interaction. The results are, qualitatively, in agreement with observations. However, it is rather uncertain whether resonance trapping could be a possible general basis for the magnitude of the positron trapping rates (Hodges 1974).

(D) *Primary trapping into dislocations.* Smedskjaer *et al* (1980b) discussed the trapping of positrons at dislocations by creation of phonons. The trapped positrons can be channelled along the dislocation line to a vacancy-like defect, where it annihilates with the decay rate τ_v^{-1}. Trapping rates were found to have a negative temperature coefficient, but an analytic form has not been given. Calculated curves show a much weaker temperature dependence than $T^{-1/2}$. The processes discussed in that paper are most likely to occur in samples containing long, straight dislocation lines, rather than small dislocation loops.

(E) *Diffusion-limited trapping.* It is well known that the positron trapping rate will be proportional to $T^{-1/2}$ it if is limited by the transient depletion of the positron density near the defect (Seeger 1973, Soininen *et al* 1990). It is therefore a natural first thought to consider this effect as the reason for the observed dependence in equation (1). However, the trapping rate cannot be diffusion limited if the transition rate from the free to the bound positron state, ν_{trans}, is much smaller than the diffusion rate $\nu_{\text{diff}} = 4\pi R D_+$, with D_+ being the positron diffusion coefficient. Numerically, for metal vacancies we have $\nu_{\text{trans}} \sim 10^{-8}$–$10^{-7}$ cm^3 s^{-1}, while $\nu_{\text{diff}} \propto 3$–$4 \times 10^{-7}$ cm^3 s^{-1} at 300 K (Soininen *et al* 1990), clearly leaving us in the transition-limited region below room temperature. This is also the conclusion obtained in the considerations of Hodges (1974) and McMullen (1978).

In order to suggest a way towards a model which can describe the observed temperature dependence of equation (1), we shall regard the positron trapping process as if it takes place in two consecutive steps: an adiabatic part, as expressed by the above model (B) (without the factor β), followed by trapping with energy exchange according to model (A).

A low-energy positron in a perfect lattice can be described by a delocalized Bloch function, $u_k(r)\exp(-ik \cdot r)$. If there is a vacancy in the lattice, the positron can be transferred, in an elastic process, to a localized 'untrapped state', determined by the positron–vacancy potential. For $\lambda \gg R$, the cross section for this first step is $\sigma_c \simeq \pi k^{-2}\mathcal{T}$. In the free positron wavefunction, the periodically repeated factor $u_k(r)$ is a sum over wavevectors q, where the qs of the dominant terms are of the order of the inverse lattice constant. Within the vacancy potential well the wavenumber K of the untrapped state is somewhat larger than the average q, but still of the same order of magnitude. Also, the edge of the vacancy's potential well will vary continuously over a distance comparable with the lattice constant. Under these circumstances the reflection of the incoming positron wave is small, and the penetration coefficient $\mathcal{T}$ will be close to unity. The elastic (adiabatic) part of the transition rate is then

$$\nu_e \simeq \hbar\pi/m_* k = \pi\hbar^2 m_*^{-3/2}(3k_B)^{-1/2}T^{-1/2} \tag{5}$$

where $m*$ is the positron's effective mass, and the wavenumber k corresponds to the average kinetic energy at temperature T.

For the next step in the trapping process, the rate ν_i is determined by the inelastic transition from the untrapped to the bound state. This is the process described by model (A) above, for which it has been found that the rate is fast, and only weakly, or not at all, temperature dependent. Taken together, the two steps give, for the temperature-dependent term in equation (1),

$$\nu_1(T) = \left(\nu_e^{-1} + \nu_i^{-1}\right)^{-1} \simeq \nu_e \propto T^{-1/2} \tag{6}$$

when $\nu_i \gg \nu_e$.

It should be noted that these considerations also are related to model (C) above, in the sense that the two steps expressed by ν_e and ν_i can be seen as representing the real and imaginary parts of the trapping potential.

As in the case of positron trapping at voids, different interpretations can be imagined for the temperature independent term ν_0. One possibility is that some of the point defects in the sample have much less symmetry than the elementary vacancy, resulting in a potential well with a deep or sharp edge. That would result in a k-dependent penetration coefficient, $\mathcal{T}$, and, as in the case of neutrons, a trapping rate independent of velocity. Another possibility is the previously-mentioned direct trapping process, which could result from the overlap of the positron Bloch function with the trapped state's exponentially decreasing tail outside the defect. Being a particular example of model (B), that process would give a temperature-independent rate.

6. Conclusion

We have studied positron trapping in normally pure, single-crystal metal samples. Small concentrations of defects are observed, having positron lifetimes typical for trapping at vacancies. The defects are probably caused in production of the crystals or during spark cutting and handling of the samples. A characteristic $T^{-1/2}$-dependent trapping rate is generally observed. The main trend of this effect can be described by a simple model based upon a quantum mechanical expression for positron–defect cross sections at long wavelengths, combined with fast trapping to a bound state in the potential well of a vacancy-like defect. It is probable that these positron traps are intrinsic parts of small dislocation loops, constituting the main content of defects in the samples.

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