Temperature dependence of positron trapping at voids in metals

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We report positron-lifetime measurements in void-containing aluminum samples, which show strong temperature dependence for the positron trapping probability. A theory is presented for the positron motion and trapping in a three-dimensional array of large voids, which compares favorably with the experimental data. It is shown that at low temperatures the trapping is transition limited and strongly temperature dependent with a crossover to diffusion-limited and weakly-temperature-dependent behavior at high temperatures.

I. INTRODUCTION

Thermalized positrons are trapped by lattice defects in solids and can convey information about defect structures and concentrations.[1] The concentrations are deduced from the fraction of positrons trapped before annihilation. For thermally generated defects one can then determine, e.g., the activation energy by measuring the trapping fraction at different temperatures. However, to relate the trapping probability to the defect concentration it is crucial to know the temperature dependence of the trapping process itself. This has been a subject of lively discussion in recent years.[2–8] A consensus seem to have been reached that for monovacancy trapping there should be practically no temperature dependence. The situation for edge dislocations is not quite clear, but even there the temperature dependence seems to be weak.[9] The key factors seem to be that (i) both vacancies and dislocations are relatively weak traps for positrons in the sense that the trapping rates are not too large and (ii) the thermal wavelength of positrons is usually much larger than the trapped-state radius.

In this paper we report studies on large voids in neutron-irradiated aluminum. Positron-lifetime measurement was chosen as the experimental method, since it assigns unambiguous values for the intensity of trapped positrons. The results show striking temperature dependence: the trapping probability increases nearly by a factor of 2 between liquid-helium and room temperatures. This is in sharp contradiction to the case with less extended defects, but it explains the type of behavior visible in the recent line-shape measurements by Mantl *et al.*[8] and in the earlier lifetime measurements by Petersen *et al.*[10] Furthermore, we present a theory of positron motion and trapping in the presence of spherical voids, which accounts for the observed temperature dependence of the trapping fraction. We show that at low temperatures the trapping process is transition limited and strongly temperature dependent in the case of large voids. At temperatures above 200 K the process becomes diffusion limited and only weakly dependent on temperature.

II. EXPERIMENTAL

The aluminum samples were two pieces of 99.9999% pure crystals spark-cut from the specimen Al-7 of Hendricks *et al.*[11] It was exposed to a fast-neutron fluence of 1.9×10^{21} n/cm^2 in Oak Ridge High-Flux Isotope Reactor and the resulting void density was 2×10^{14} cm^{-3} with an average void diameter of 500 Å.[11] The pieces were electrolytically polished and sandwiched with a 30-μCi ^{22}Na positron source evaporated onto a 1.1-mg/cm^2 thin nickel foil. The temperature of the sample in the sample holder could be adjusted from 3 to 400 K with a stability of ±0.5 K. The positron lifetime measurements were carried out using a conventional fast-slow coincidence system with a resolution of 300 psec [full width at half-maximum (FWHM)]. The counting time per one spectrum was 10 h during which about 8×10^5 pulses were collected.

The lifetime spectra were seen to depend strongly on sample temperature. After source-background subtractions the curves were analyzed as a sum of exponential terms and in all cases two lifetime components were sufficient for a satisfactory fit. The results are shown in Table I. The lifetime values are temperature independent being $\tau_1=210\pm10$ psec and $\tau_2=450\pm10$ psec. The longer component originates from trapped positrons annihilating in the

voids. Taking into account different fitting procedures of various authors its value agrees well with those reported previously for aluminum voids.[10,12] The previously observed temperature dependence of the void lifetime values[10] starts only at higher temperatures ($T > 500$ K), where the voids begin to anneal out. The shorter component includes free positrons and positrons trapped by smaller defects like dislocation loops, impurity-bound vacancies, etc. To reduce the statistical scatter in the intensity I_2 we fixed the longer lifetime to its average value 450 psec and repeated the analysis. The intensity I_2 of the longer lifetime is given in Fig. 1 as a function of sample temperature. In repeated measurements no hysteresis was observed. The maximum temperature used (400 K) was limited by the recovery of the defect structure which starts slowly already around 500 K.[10,12] Figure 1 shows the drastic effect of sample temperature on positron trapping at voids; the trapping component at low temperatures is reduced to about one-half of its room-temperature value. Below 200 K a strikingly linear dependence of I_2 on temperature is seen and above 200 K the intensity I_2 stays nearly constant as long as the defect structure is maintained.

TABLE I. Results for positron-lifetime measurements as a function of sample temperature in aluminum voids with mean radius 250 Å and density 2×10^{14} cm^{-3}. The errors are statistical standard deviations from the two-component fit.

T (K)	τ_1 (psec)	τ_2 (psec)	I_2 (%)
3.8	204 ± 5	442 ± 7	29.6 ± 2.0
4.5	197 ± 5	453 ± 7	27.7 ± 2.0
14	200 ± 5	449 ± 8	29.4 ± 1.9
26	220 ± 4	446 ± 10	27.1 ± 2.0
46	215 ± 4	444 ± 9	33.3 ± 2.2
60	209 ± 5	440 ± 8	35.3 ± 2.2
77	208 ± 4	452 ± 7	34.1 ± 1.7
88	210 ± 4	441 ± 7	38.1 ± 2.1
115	216 ± 4	461 ± 8	39.5 ± 2.2
145	204 ± 5	445 ± 7	40.2 ± 2.0
175	198 ± 5	442 ± 6	43.2 ± 1.9
200	204 ± 4	446 ± 6	42.3 ± 1.7
230	198 ± 4	449 ± 6	43.9 ± 1.7
256	196 ± 4	451 ± 5	44.0 ± 1.5
295	204 ± 4	459 ± 7	42.1 ± 2.0
335	197 ± 5	433 ± 6	46.8 ± 2.0
378	205 ± 5	454 ± 6	42.3 ± 2.1

III. THEORY

In the following we present a theory which qualitatively explains the behavior depicted in Fig. 1. Nontrapped positrons in metals are nearly free Bloch particles with a mobility essentially limited by the interaction with lattice vibrations. Positrons have a tendency to seek out regions of lower than average ionic density and get localized there with a trap binding energy of the order of 1 eV.[13] If the positron-trap interaction is weak, the rate of this process is adequately given by the Golden Rule formula.[2] If the trap is spatially small, this approach gives no temperature dependence for the trapping rate, since the positron thermal wavelength $\lambda\!\!\!^{-}$ at relevant temperatures is substantially larger than trap dimensions. McMullen[5] has shown that the temperature independence remains valid for small traps even beyond the zero-order Golden Rule approximation, i.e., for strong trapping.

On the other hand, if the trap is extended, the positron-trap interaction may become large and then the Golden Rule in most cases underestimates the specific trapping rate. Instead of following McMullen's[5] perturbative method in analyzing this situation, we follow the analogy to neutron capture reactions in nuclei.[14] In this semiclassical approach, one has the specific trapping rate $\nu = v_+\sigma$, where $v_+ = (3kT/m^*)^{1/2} = \hbar k_+/m^*$ is the thermal velocity of positrons with the effective mass m^*, and σ is the positron capture cross section of the void. The s-wave part is dominant and may be approximated $\sigma = \pi(r_v + \lambda\!\!\!^{-})^2\xi$, *where* r_v is the void radius, $\lambda\!\!\!^{-}$ is the thermal wavelength of the positron and ξ is the trapping coefficient, which accounts for the absorption of the incident wave at the trap boundary as well as for the efficiency of the energy release mechanisms of the medium, which have to carry away the binding energy of the positron to the trap. In metals, the bulk of the binding energy E_b is taken very rapidly by electron-hole excitations (the Fermi energy $\hbar^2k_F^2/2m > E_b$, $k_F >> k_+$) and thus ξ is limited by the wave absorption:
$\xi \simeq 4k_+K_+/(k_+ + K_+)^2$, where K_+ is the positron wave number in the localized trap state.

If $r_v << \lambda\!\!\!^{-}$, as is the case for monovacancies, one sees that ν is independent of temperature (since $K_+ >> k_+$). On the other hand, for large voids (except at very low temperatures) $r_v >> \lambda\!\!\!^{-}$ and thus

$$\nu \simeq 4\pi r_v^2 v_+ k_+/K_+ \propto T^{+1} \quad . \tag{1}$$

Strictly speaking, Eq. (1) cannot hold as $T \to 0$; a lower limit to ν is provided by the Golden Rule formula. Consequently, we can adopt the following temperature dependence for the specific trapping rate to large voids:

$$\nu = \nu_0 + \gamma T \quad , \tag{2}$$

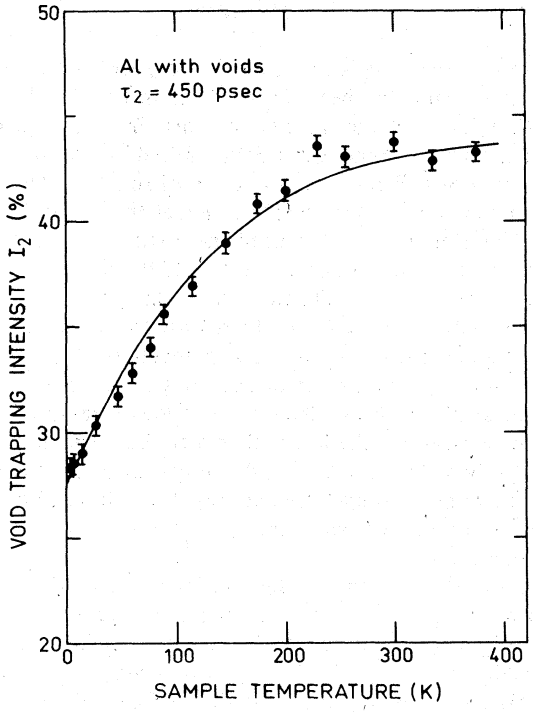

FIG. 1. Lifetime intensity I_2 as a function of temperature for positrons annihilating in aluminum voids with mean radius 250 Å and density 2×10^{14} cm^{-3}. The error bars are statistical standard deviations with the void lifetime fixed to its average value $\tau_2=450$ psec. The solid curve is the result of theoretical calculations.

where the constants ν_0 and γ will be specified later.

So far we have not considered the effect of positron motion on the trapping process. If the voids act as very efficient absorbers, i.e., the ratio of the trapping rate to the positron mobility is large, it may be that the positron current is not sufficient to maintain the full rate, which would follow from Eq. (2). Near a strong trap the density of positrons can be depleted with ensuing fall in the net trapping probability. The simplest approach to the positron transport problem is via the diffusion equation,[15] which is valid over distances large compared to a positron mean free path. The positron mobility μ_+ in metals is limited by acoustical-phonon scattering, and consequently the temperature dependence of the diffusion coefficient, defined via the Einstein relation $D_+=kT\mu_+/e$ is given by

$$D_+=D_0(300/T)^{1/2} \ , \tag{3}$$

where T is given in degrees K. Following the calculation of Bergersen *et al.*[16] we find $D_0=0.6$ cm^{2}sec^{-1}, corresponding to a positron effective mass in Al $m^*=1.5$. Given a density N_v of voids with radius r_v we define the intervoid Wigner-Seitz radius $R_v=(3/4\pi N_v)^{1/3}$. The positron density distributions $n_f(r,t)$ and $n_t(t)$ (for free and trapped positrons, respectively) are obtained from the solution to the diffusion-annihilation equations

$$\frac{\partial n_f(r,t)}{\partial t}=D_+\nabla^2 n_f(r,t)-\lambda_f n_f(r,t) \ , \tag{4}$$

$$\frac{\partial n_t(t)}{\partial t}=-\lambda_t n_t(t)+\nu n_f(r_v,t) \ , \tag{5}$$

subject to the boundary and initial conditions

$$D_+4\pi r_v^2\frac{\partial n_f(r,t)}{\partial r}\bigg|_{r=r_v}=\nu n_f(r_v,t) \ , \tag{6}$$

$$\frac{\partial n_f(r,t)}{\partial r}\bigg|_{r=R_v}=0 \ , \tag{7}$$

$$n_f(r,0)=n_0 \ , \tag{8}$$

$$n_t(0)=0 \ . \tag{9}$$

Above, λ_f is the annihilation rate of positrons in bulk metal (for Al, $\lambda_f=5.9$ nsec^{-1})[17] and λ_t the annihilation rate in the trap state. Equation (6) describes the capture of positrons at the void surface, while Eq. (7) is the usual continuity condition. We assume that no positrons thermalize inside the voids (no prethermalization trapping) and that there is no escape from the voids. The solution to Eqs. (4) and (6)–(8) is[18]

$$n_f(r,t)=\sum_{n=1}^{\infty}e^{-(D_+\alpha_n^2+\lambda_f)t}f_n(r) \ , \tag{10}$$

where the eigenfunctions $f_n(r)$ can be constructed in terms of sine and cosine functions[18]

$$f_n(r)=\frac{2n_0}{r}R_n(r)\int_{r_v}^{R_v}dr'\,r'R_n(r') \ , \tag{11}$$

where

$$R_n(r)=\frac{(1+R_v^2\alpha_n^2)^{1/2}\{(r_v\nu+4\pi r_v^2D_+)\sin[\alpha_n(r-r_v)]+4\pi r_v^3D_+\alpha_n\cos[(r-r_v)\alpha_n]\}}{(A+B)^{1/2}} \ , \tag{12}$$

with

$$A=(R_v-r_v)r_v^2[(4\pi r_vD_+)^2(r_v^2\alpha_n^2+1)2\nu^2+8\pi r_vD_+](R_v^2\alpha_n^2+1) \ , \tag{13}$$

$$B=r_v^2[4\pi r_vD_+(R_v-r_v)+R_v][4\pi r_vD_+(R_vr_v\alpha_n^2-1)-\nu] \ . \tag{14}$$

Above $\alpha_n \neq 0$ is a solution to

$$\tan[(R_v - r_v)\alpha_n] = \frac{\alpha_n[R_v\nu + 4\pi r_v D_+(R_v - r_v)]}{\nu + 4\pi r_v D_+(1 + R_v r_v \alpha_n^2)} \quad . \tag{15}$$

Figure 2 shows typical instantaneous positron density distributions at different temperatures. The transient time needed to reach the quasiequilibrium, time-independent form for the flux is very short (<5 psec) compared to positron lifetimes. In the inset, the quasiequilibrium positron density at the void boundary $n(r_v)$ is shown as a function of temperature. At low T, the positron diffusion constant is large and the flux nearly uniform. This is the transition-limited regime, where the temperature dependence of the trapping is governed by that of ν. At higher temperatures D_+ diminishes, while ν increases and the positron flux is depleted near the void. The temperature dependence in this diffusion-limited regime is due to that of D_+.

The fraction of positrons that are trapped is

$$F = \frac{R_v^3}{R_v^3 - r_v^3}\frac{\nu N_v}{n_0}\int_0^\infty dt\, n_f(r_v,t) = \frac{R_v^3}{R_v^3 - r_v^3}\frac{\nu N_v}{n_0}\sum_{n=1}^{\infty}\frac{f_n(r_v)}{\lambda_f + D_+\alpha_n^2} \quad . \tag{16}$$

The geometrical factor in Eq. (16) enters because the diffusing positrons are initially distributed in the annular regions between r_v and R_v. The conventional parameter in the lifetime spectra analysis is I_2, the intensity of the void component, which is from Eq. (5),

$$I_2 = \frac{R_v^3}{R_v^3 - r_v^3}\frac{\nu N_v}{n_0}\sum_{n=1}^{\infty}\frac{f_n(r_v)}{\lambda_f - \lambda_t + D_+\alpha_n^2} \quad . \tag{17}$$

At low temperatures, the positron mobility is high ($4\pi r_v D_+ >> \nu$) and only the lowest root of Eq. (15) contributes

$$\alpha^2 \cong 3\nu/4\pi D_+(R_v^3 - r_v^3) \quad . \tag{18}$$

This leads to the familiar result for the trapping fraction

$$F \cong \nu N_v'/(\nu N_v' + \lambda_f) \quad , \tag{19}$$

where $N_v' = 3/4\pi(R_v^3 - r_v^3)$. In the opposite limit with $4\pi r_v D_+ << \nu$ the lowest root of Eq. (15) is

$$\alpha^2 \cong 3r_v/(R_v^3 - r_v^3) \quad . \tag{20}$$

If $R_v >> r_v$, this again dominates and one obtains the result valid for extreme diffusion limited trapping

$$F \cong 4\pi N_v' D_+/(4\pi N_v' D_+ + \lambda_f) \quad . \tag{21}$$

The solid line in Fig. 1 shows the calculated intensity I_2 from Eq. (17) as a function of temperature in Al containing a density of $N_v = 2 \times 10^{14}$ cm^{-3} of voids with a radius $r_v = 250$ Å.

We have estimated ν_0 in Eq. (2) by doing a Monte Carlo integration of the Golden Rule formula[2] for positron trapping into surface states and multiplied by the void surface area. In metals, electron-hole mediated trapping dominates and one has

$$\nu_0 = 4\pi R_v^2 \frac{4\pi}{\hbar}\sum_{\bar{k}\bar{q}}|M_{\bar{k}\bar{q}\bar{l}}|^2\,\delta(\epsilon_{\bar{k}} - \epsilon_{\bar{k}+\bar{q}} - \epsilon_t) \quad , \tag{22}$$

where $\bar{k}$ labels the initial electron state (assumed to be a plane wave), $\bar{q}$ is the momentum transfer, and ϵ_t is the positron binding energy to the trap which is carried away by single electron-hole pair excitations. Above, the matrix element is

$$M_{\bar{k}\bar{q}\bar{l}} = [v_s(q)/\sqrt{\Omega}]\,\langle\phi|\bar{q}\rangle f_{\bar{k}}(1 - f_{\bar{k}+\bar{q}}) \quad , \tag{23}$$

where $v_s(\bar{q})$ is the (Thomas-Fermi) screened-Coulomb interaction, Ω is the volume, ϕ is the trapped state, and $f_{\bar{k}}$ is the Fermi function. Using the theoretically estimated trapped-state wave function and binding energy[19] for surface states in Al we find $\nu_0 \simeq 0.6 \times 10^{-5}$ cm^3/sec. The value is sensitive to surface state parameters and in view of their uncertainty we have adjusted ν_0 to 0.75×10^{-5} cm^3/sec, which correctly reproduces the low-temperature limit in Fig. 1. The coefficient γ in Eq. (2) has been chosen so ($\gamma = 6.8 \times 10^{-8}$ cm^{3}sec^{-1} K^{-1}) that the calculated curve coincides with the experimental one around 100 K. Using Eq. (1), this value of γ corresponds to a positron final-state localization distance $K_+^{-1} \simeq 2$ Å, which is consistent with the surface-state model.[19] It is also essential that the value of the diffusion constant D_+ is of the magnitude calculated above, otherwise there is no value of γ which would fit the data. From Fig. 1 we see a good overall agreement between theory and experiment. At low T, the temperature dependence of I_2 is nearly linear. At around 200 K, both the experimental and theoretical curves flatten, and at high temperatures the latter eventually starts to fall off slowly as $T^{-1/2}$.

Figure 3 shows a set of curves for the trapping fraction F calculated from Eq. (16) for different void sizes and densities, the values of which correspond to those reported by Mantl *et al.*[8] in their Doppler-broadening study. We have used the above values for the coefficients ν_0 and γ, scaled for voids of different radii with the square of r_v. This procedure brings the saturation level of the trapping fraction for the sample with the highest density of voids to about 35% at room temperature. The right-hand panel of

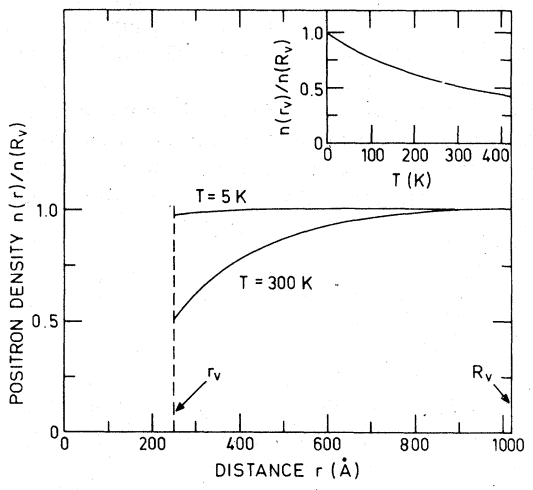

FIG. 2. Quasiequilibrium positron density distributions in the presence of voids calculated at two different temperatures. The void radius is denoted by r_v and the intervoid Wigner-Seitz radius by R_v. The inset shows the positron density at the void boundary as a function of temperature.

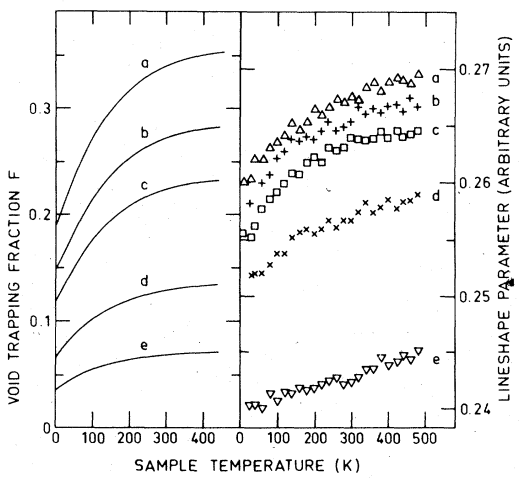

FIG. 3. Calculated positron trapping fractions F as a function of temperature in Al (left hand column). The void radii and densities correspond to average values in the Doppler measurements of Mantl *et al.* (Ref. 8), whose results are plotted in the right-hand column. The void parameters were *a*, $r_v = 195$ Å, $N_v = 3.1 \times 10^{14}$ cm^{-3}; *b*, $r_v = 195$ Å, $N_v = 2.3 \times 10^{14}$ cm^{-3}; *c*, $r_v = 195$ Å, $N_v = 1.8 \times 10^{14}$ cm^{-3}; *d*, $r_v = 210$ Å, $N_v = 8.4 \times 10^{13}$ cm^{-3}; *e*, $r_v = 245$ Å $N_v = 3.3 \times 10^{13}$ cm^{-3}.

Fig. 3 shows the results of the line-shape measurement of Mantl *et al.*[8] The qualitative agreement between the two sets of curves is good. The dependence of the trapping fraction on the void density is correctly reproduced, as well as the tendency of the curves to flatten out when the void density decreases.

IV. DISCUSSION

In conclusion, positron lifetime measurements show strong temperature dependence for the positron trapping probability at large voids in aluminum. A semiclassical theory has been presented which explains the observed phenomena. At low temperatures, the positron mobility is large and the capture rate is small; the trapping process is transition limited and strongly dependent on temperature. At around 200 K, a crossover to diffusion-limited region occurs, as the mobility decreases and the capture rate increases. This is a manifestation of the self-shielding of the voids from the positron flux.

The model proposed is in accord with the idea of positron being trapped at localized surface states in metal voids. However, the existence of possible spin correlation ("positroniumlike states") in these is irrelevant to the present analysis. All efforts so far to find spin correlation have failed, though, and the magnetic quenching experiments[10,20] have clearly excluded free positronium formation in metallic voids.

We should also like to point out that the success of the theory may be regarded as a strong support for the correct magnitude of the positron diffusion constants.[16] Whereas there is no direct measurement of the positron mobility in metals, the experiments by Mills and Pfeiffer[21] give results for the mobility in semiconductors, which are in accord with Eq. (3) and of the same order of magnitude as the values theoretically estimated for metals.

ACKNOWLEDGMENTS

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$$\phi(r) = Ae^{0.532z}e^{i\vec{k}_{\parallel}\cdot\vec{r}_{\parallel}} \quad (z < 0 \text{ in metal})$$

$$= Be^{-0.54z}(z + 0.93)e^{i\vec{k}_{\parallel}\cdot\vec{r}_{\parallel}} \quad (z > 0 \text{ in vacuum}) \ ,$$

with A,B chosen to ensure normalization and continuity. This corresponds to a positron binding energy of 1.4 eV. For details, see R. M. Nieminen and C. H. Hodges, Phys. Rev. B 18, 2568 (1978).

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