

PHYSICAL REVIEW B

CONDENSED MATTER

THIRD SERIES, VOLUME 27, NUMBER 1

1 JANUARY 1983

Temperature dependence of positron annihilation at dislocations in Pb(Cd)

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(Received 26 April 1982; revised manuscript received 14 June 1982)

We have measured the temperature dependence of positron lifetimes in a $\text{Pb}_{96.1}\text{Cd}_{3.9}$ alloy having a dislocation density large enough to significantly affect the e^+e^- annihilation spectra. These spectra were resolved into two unconstrained lifetimes in addition to a source-term lifetime. The longer of the two lifetimes, which is attributed to positrons localized by traps associated with dislocations, decreases with increasing sample temperature T at about 0.1 psec K^{-1} for $90 \leq T \leq 300 \text{ K}$. These data represent the first reported observation of a temperature-dependent lifetime of positrons localized at dislocation traps. It is proposed that the temperature dependence is principally due to atoms penetrating deeper into the traps as T is raised. We suggest that these atomic incursions also effect a negative contribution to the slope of the line-shape parameter $S(T)$. Analysis of the data in terms of the two-state trapping model reveals (i) that both the trapping rate per unit dislocation density and the probability of annihilation occurring at the dislocation traps for constant dislocation density are weakly temperature dependent, and (ii) that during recovery anneals the total trapping rate decreases markedly, whereas little change is exhibited by the lifetime of the trapped positron.

I. INTRODUCTION

It is well established that positron-annihilation spectroscopy is a sensitive tool for investigating voidlike defects in metals.¹ Quantitative analysis of the e^+e^- annihilation spectra, however, often requires a knowledge of the temperature dependence of several quantities affecting the annihilation process, one being the lifetime of positrons localized at a particular type of defect. It is difficult to deduce this type of information on the basis of a typical analysis of Doppler-broadening or mean-lifetime data, which describes the annihilation behavior by a single quantity. We cite one example which illustrates both the shortcomings of a single-parameter analysis and the motivation for the present work. Doppler-broadened energy profiles of the e^+e^- annihilation photons were recently measured for rolled and unrolled Pb, Pb(Cd), and Pb(Tl),² where the rol-

ling was performed at room temperature. A measured energy profile reflects, by the Doppler effect, the momentum distribution of electrons sampled by positrons. The major contribution to this profile arises from positrons annihilating with conduction electrons; a second contribution, smaller in intensity and broader in energy, results from annihilations with core electrons. To study the temperature dependence of the relative change in these two contributions, the investigators in Ref. 2 used the conventional S parameter,³ which is a measure of the height of a normalized profile curve. Thus, for example, an increase in the fraction of positrons annihilating with high-momentum core electrons would broaden the profile curve and, consequently, decrease S . For sample temperatures T between 77 and 350 K, S was much larger for as-rolled Pb(Cd) than for rolled or unrolled pure Pb, rolled or unrolled Pb(Tl), rolled and recrystallized Pb(Cd), or

as-cast Pb(Cd). These large S values were explained in terms of positrons annihilating at voidlike traps associated with dislocations in the as-rolled Pb(Cd) specimens. It is significant that, for the as-rolled Pb(Cd) samples, S decreased with increasing T .⁴ That is, the intensity in the wings of the Doppler-broadened energy profile increased with T , indicating an increase of positrons annihilating with high-momentum core electrons. This behavior, inexplicable in terms of a recovery process since S varied reversibly with T , suggests that S_D , the line-shape parameter representative of annihilation at the dislocation traps, is decreasing with T and/or that fewer positrons are annihilating at these traps as the sample temperature is raised. As already indicated, it would be difficult to choose between these two possibilities on the basis of the Doppler-profile studies of Ref. 2.

In this paper we describe an investigation of the temperature dependence of positron lifetimes in a Pb(Cd) alloy (one similar to those used in Ref. 2) during which we have resolved the e^+e^- annihilation spectra into their components; using the more complete information obtained from this approach, the first of the two possibilities listed above emerges as the more reasonable interpretation of the origin of the negative slope exhibited by S . Our objective was to obtain this additional insight concerning the Doppler data and, in particular, to determine the *temperature dependence* of three quantities which have been the focus of much attention⁵: (1) the lifetime of positrons localized at traps associated with dislocations, (2) the probability of annihilation occurring at these traps for constant dislocation density, and (3) the trapping rate per unit dislocation density.

II. EXPERIMENTAL INFORMATION

A. The sample

The Pb_{96.1}Cd_{3.9} alloy was prepared by melting and casting Cominco 99.9999% pure Pb and Ventron 99.9999% pure Cd in a fused silica tube under $\frac{1}{3}$ atm of a reducing gas (80 mol % He—20 mol % H₂). The solidified rod was homogenized, rolled at room temperature to a thickness of 0.5 mm from a diameter of 13 mm, and cut into circular disks 13 mm in diameter. The positron source, about 10 μ Ci of ²²NaCl, was deposited from an aqueous solution between two pieces of 1- μ m Ni foil, which were then sandwiched between two specimen disks. The source-sample sandwich was sealed in an aluminum capsule under 1 atm of H₂ gas. The sample in this rolled and unannealed condition is referred to as the "as-rolled" specimen. Isothermal measurements of the annihilation spectra were made on the as-rolled

specimen at several sample temperatures between 80 and 300 K during repeated heating and cooling cycles. The results of these measurements are discussed in Sec. IV A.

After the measurements on the as-rolled specimen were made, the encapsulated source-sample sandwich was annealed at 255°C for 1 h and quenched into ice water. The capsule was dried within about 20 sec and stored in liquid nitrogen until it was mounted on a 100-K stage, which resulted in the sample temperature increasing to about 180 K for approximately 100 sec. After this thermal treatment, the sample is referred to as the "quenched" specimen. The results of isothermal measurements made on the quenched specimen are discussed in Sec. IV B.

Two additional sets of measurements were made on the same encapsulated Pb_{96.1}Cd_{3.9} specimen. These were taken at a sample temperature of about 100 K following 1-h anneals at temperatures T_A between 20 and 165°C; about 40 min was required to heat the sample to T_A , and it took about the same time to recool it to 100 K after each anneal. One set was made on the specimen in the as-rolled condition, after the measurements described in Sec. IV A but before those of Sec. IV B, and a second set was made after the measurements described in Sec. IV B. The results of both sets of isochronal-annealing measurements are discussed in Sec. IV C.

Because of the nature of this investigation, it was necessary that the samples be free of significant amounts of voidlike defects other than the dislocations under investigation. Based on the following, we assert that micropores, precipitates, and vacancies in the as-rolled specimen had a negligible effect on the annihilation spectra. The same is most likely true for the quenched sample, since the data obtained for it were similar to those obtained for the as-rolled specimen.

Micropores. The solubility of Cd in solid Pb is much less than that in liquid Pb. Hence when preparing Pb(Cd) it is difficult to avoid constitutional supercooling and, consequently, dendritic growth is likely. Under such conditions, microporosity will develop when liquid occluded among the dendrites shrinks upon solidification. However, it has been shown² that rolling reduces the size and number of micropores such that, even in a recrystallized specimen, their presence cannot be detected with positrons.

Precipitates. Transmission electron microscopy was carried out on several Pb(Cd) alloys thinned in an Ar-ion milling machine. The precipitates observed in this manner had the form of rods 0.4–2 μ m in diameter that spheroidized in time. However, for sample temperatures between 7 and 350 K, the

behavior of positrons in pure, well-annealed Pb is almost identical to that in a rolled and recrystallized Pb(Cd) alloy,² i.e., in a Pb(Cd) alloy free of significant amounts of defects other than precipitates.

Vacancies. Previous work⁸ has shown that the effects of positron trapping at vacancies in Pb(Cd) are below the detection limit for sample temperatures less than 100°C.

B. The measurement

The e^+e^- annihilation spectra were measured with a delayed coincidence system⁹ having a full width at half maximum of 200 psec with 50% side windows (²²Na) and a centroid stability of 3 psec/day. A typical spectrum contains (1.5–4) $\times 10^6$ counts and has a peak to background ratio of 3500 to 1. Essentially, a measured annihilation spectrum is the time derivative of the distribution function describing the decay of some initial positron population. For example, if the two-state trapping model (see Sec. III) is applicable, in essence, one is measuring $\dot{n}(t)$, where the distribution function $n(t)$ is given below by Eq. (4).

III. DATA TREATMENT

The lifetime spectra were analyzed using two computer programs. One is the standard program POSFIT,¹¹ and the other, DBLCON,¹² is a modification of POSFIT using an exponential-sided Gaussian¹³ for the instrumental resolution or prompt. The spectra cutoffs determined by Sharma¹⁰ and Hu¹⁴ were used with POSFIT and DBLCON, respectively. POSFIT analyses utilized data to the right of and less than 80% of each spectrum peak. For DBLCON analyses, all the spectra were used except those portions to the left of and less than 85% of each peak. The results of using POSFIT are similar to those of DBLCON, and only the former are given in this paper.

The source term is a spurious contribution due to annihilation at or near the surfaces about the positron source and, to a lesser extent, in the source material itself. This contribution was approximated by a single exponential decay function with lifetime τ_s and intensity I_s . These parameters were found to be temperature independent ($\tau_s = 451 \pm 5$ psec, $I_s = 6.0 \pm 0.2\%$) by unconstrained analysis of the spectra for sample temperatures $T < 100^\circ\text{C}$. The analysis was made after the specimen was annealed above 165°C for a minimum of 1 h, an anneal which apparently reduced the dislocation density enough so that only one lifetime in addition to τ_s could be resolved. Further evidence for a temperature-independent source term was obtained by fitting the spectra for the unannealed sample, in both as-rolled and quenched conditions, to three exponential decay

functions, holding only τ_s constant; within experimental uncertainty, this analysis gave an I_s independent of temperature. Consequently, all data were reanalyzed for two unconstrained lifetimes ($\tau_2 > \tau_1$) and intensities (normalized such that $I_1 + I_2 = 1$) after subtracting the constant source term from each spectrum.

The source term incorporates the effect of annihilations of positrons having diffused back to the Ni-foil surfaces. It does not include the contribution due to annihilations occurring within the Ni, which involves as many as 5% of the positrons. Although no correction has been made for these annihilations, their contribution is sufficiently small that the omission in our analysis should not significantly affect our conclusions.

Some interpretation of the data is made in terms of the simple two-state trapping model.¹ The basic idea of this model in the present context is as follows: A positron injected into a rolled or quenched Pb(Cd) sample diffuses through that sample until it either annihilates from the bulk or becomes localized at a trap associated with a dislocation and then annihilates. In terms of this model, the net disappearance rate of delocalized positrons λ_1 equals the sum of their annihilation rate λ_B plus their trapping rate $\mu_D C_D$:

$$\lambda_1 = \lambda_B + \mu_D C_D . \quad (1)$$

Here μ_D is the trapping rate per unit dislocation density and C_D is the dislocation density. The rate equations describing the annihilation behavior are given by

$$\dot{n}_B = -\lambda_1 n_B \quad (2)$$

and

$$\dot{n}_D = -\lambda_2 n_D + \mu_D C_D n_B , \quad (3)$$

where the distribution functions n_B and n_D give, respectively, the fraction of delocalized and localized positrons remaining in the sample at time t , and λ_2 is the annihilation rate for localized positrons. Solving Eqs. (2) and (3) with the initial condition that $n_B(t=0) = 1$, one finds that $n \equiv n_B + n_D$ may be expressed as

$$n(t) = (1 - I_2) \exp(-t/\tau_1) + I_2 \exp(-t/\tau_2) , \quad (4)$$

where

$$I_2 = \frac{\mu_D C_D}{(\lambda_1 - \lambda_2)} , \quad (5)$$

$$\tau_1 \equiv \lambda_1^{-1} , \quad (6)$$

and

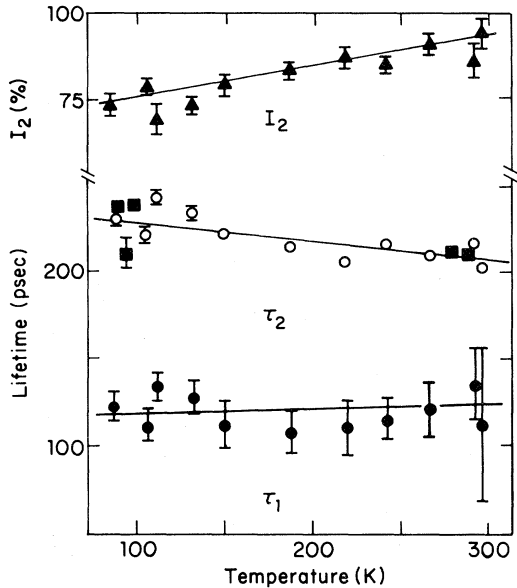


FIG. 1. Results of POSFIT analysis for $\text{Pb}_{96.1}\text{Cd}_{3.9}$ rolled at room temperature and stored at or below room temperature. Lines I_2 and τ_2 are weighted least-squares fits; line τ_1 was computed using Eq. (9) (see text). The slope of τ_2 (open circles) is 0.11 ± 0.04 psec/ $^\circ\text{C}$. Also shown are five points τ_2 (solid squares) which correspond to the five points shown in Fig. 3; the square with error bars corresponds to point 1.

$$\tau_2 \equiv \lambda_2^{-1}. \quad (7)$$

It is assumed that positron trapping is limited by the transition to the trapped state, as opposed to being diffusion limited, and that detrapping is negligible. If so, the probability of annihilation occurring at defect traps may be written as the ratio of the trapping rate to the net disappearance rate of delocalized positrons:

$$P_D = \frac{\mu_D C_D}{\lambda_1} = I_2 (\lambda_1 - \lambda_2) / \lambda_1. \quad (8)$$

A more complete description of the data treatment can be found in Ref. 14.

IV. RESULTS AND INTERPRETATION

A. The as-rolled specimen

The resolved lifetimes for the sample in the as-rolled condition are shown in Fig. 1. As seen, τ_2 decreases with increasing T . One might consider explaining this behavior by allowing positrons to detrapp and return to the bulk; however, the detrapping rates required would be relatively large and would effect a substantial drop in τ_1 with T , which is not observed. Therefore, τ_2 is interpreted as the lifetime

of positrons localized by traps associated with dislocations. If so, the negative slope of τ_2 (about 0.1 psec K^{-1}) indicates that, on the average, the electron density sampled by the trapped positrons increases with temperature. This suggests several possibilities. If more than one bound state exists for a trapped positron, then thermal excitations into a higher-energy state would result in the positron becoming more spread out.¹⁵ If more than one type of trap is associated with the dislocations, then interplay between traps similar to that suggested in Ref. 6 is possible. These two ideas are not exclusive and they may be related if the different types of traps are physically connected, e.g., if they correspond to different positions along a single dislocation.¹⁵ A third possibility is that the electron density sampled by the trapped positrons increases with temperature because, as T is raised, atoms are penetrating the trap more deeply. We favor this last idea since one may apply the same reasoning to also explain why the lifetime of positrons trapped by vacancies in either Pb or Pb(Cd) decreases with increasing T .⁸ Moreover, if this reasoning is valid, then the lack of observations of similar behavior in materials other than Pb and some Pb alloys may be attributable, in part, to the relatively large magnitude and temperature dependence of the atomic mean-square displacement in Pb. We also suggest that the increase of atomic incursions into the trap as T is raised effects a negative contribution to the slope of the line-shape parameter $S(T)$. If the dislocation density is large enough, i.e., if $P_D > \frac{1}{3}$ or so, this contribution will cause $S(T)$ to drop with T , as discussed in Sec. I. Finally, we note the possibly related finding¹⁶ that the peak counting rate of the angular correlation curve for positrons in liquid Pb also decreases with increasing sample temperature.

The temperature dependence of the other quantities in Fig. 1, I_2 and τ_1 , can be understood in terms of the simple two-state trapping model.¹ According to Eq. (5), $I_2(T)$ is determined by $\mu_D C_D$ and $(\lambda_1 - \lambda_2) = (\lambda_B - \lambda_2 + \mu_D C_D)$. Later in this section it is shown that $\mu_D C_D$ varies weakly with temperature; hence, the increase of I_2 with T is due primarily to $(\lambda_1 - \lambda_2)$ decreasing with T , which is evidenced in Fig. 1. Furthermore, I_2 extrapolates to unity at about 90°C , the same temperature, within experimental uncertainty, at which the positive quantity $(\lambda_B - \lambda_2)$ extrapolates to zero. Therefore, the temperature dependence of I_2 primarily reflects that the system is approaching the limit where the lifetime of positrons localized at dislocation traps (λ_2^{-1}) equals the lifetime of delocalized positrons ($\tau_B \equiv \lambda_B^{-1}$). On the other hand, the small positive slope exhibited by τ_1 primarily reflects the temperature dependence of λ_B ; this point is discussed further below.

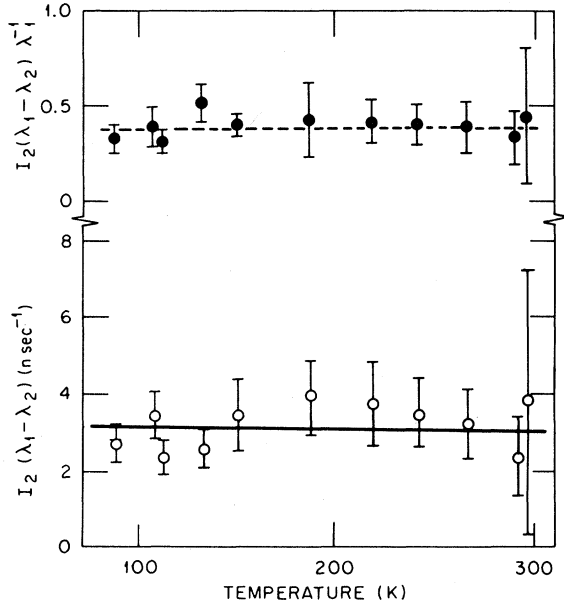


FIG. 2. Variation of $P_D = I_2(\lambda_1 - \lambda_2)\lambda_1^{-1}$ and $\mu_D C_D = I_2(\lambda_1 - \lambda_2)$ with temperature, computed using the data in Fig. 1 for the as-rolled specimen. The dashed line shows a weighted average of 0.37 ± 0.03 ; the solid line was computed using Eq. (10) (see text).

The probability P_D , computed using Eq. (8) and the data in Fig. 1 for the as-rolled sample, is shown in the upper portion of Fig. 2. Since the dislocation density C_D did not change in the course of these measurements (the specimen was rolled at room temperature and stored at room temperature for almost one year prior to this experiment, and the isothermal measurements were unaffected by cycling the sample temperature over the range shown), P_D is the annihilation probability for constant dislocation density. Notice that, within experimental uncertainty, P_D exhibits no temperature dependence. This result is in agreement with curve B or curve C in Fig. 2 of Bergensen and McMullen.⁷

Using Eqs. (1) and (8), one can show that

$$\tau_1 = (1 - P_D)\tau_B \quad (9)$$

or, equivalently

$$\mu_D C_D = \frac{P_D}{1 - P_D} \tau_B^{-1}. \quad (10)$$

Hence if P_D is constant and the assumptions already presented are appropriate, then the temperature dependence of both τ_1 and $\mu_D C_D$ should be governed by that of τ_B .

Before showing that Eqs. (9) and (10) are applica-

ble to the present data, we indicate how τ_B is obtained. If there is no direct prethermalization trapping, one expects¹⁷ that the annihilation rate of delocalized positrons λ_B should equal the initial positron annihilation rate ($I_1\tau_1^{-1} + I_2\tau_2^{-1}$). We find that this relation, which follows from Eqs. (1) and (5), holds in the present system over the entire temperature range studied. In particular, the initial annihilation rate of positrons in the *as-rolled* sample ($I_1\tau_1^{-1} + I_2\tau_2^{-1}$) obtained from the data of Fig. 1 agrees well with the measured⁸ annihilation rate of (delocalized) positrons in the *well-annealed* sample. That is,

$$\tau_{WA}^{-1} = (I_1\tau_1^{-1} + I_2\tau_2^{-1})_{AR}, \quad (11)$$

where the subscripts WA and AR denote well-annealed and as-rolled, respectively; since only one lifetime τ_{WA} (in addition to the source-term lifetime) could be resolved in the well-annealed specimen for $T < 350$ K, the identification $\lambda_B = \tau_{WA}^{-1}$ is appropriate. Hence either the left-hand or right-hand side of Eq. (11) can be used to obtain τ_B .

The applicability of Eqs. (9) and (10) to the present data is demonstrated by the solid lines τ_1 and $\mu_D C_D = I_2(\lambda_1 - \lambda_2)$ shown in the lower portions of Figs. 1 and 2, respectively. These lines are not direct fits to the data but were computed using these equations with $P_D = 0.37$, the weighted average of $I_2(\lambda_1 - \lambda_2)\lambda_1^{-1}$ (see Fig. 2), and τ_B taken as the weighted least-squares fit to τ_{WA} , given in picoseconds by $(196 + 4.71 \times 10^{-2}T)$, where T is in $^{\circ}\text{C}$. τ_B^{-1} , in inverse nanoseconds, was taken as $(5.1 - 1.22 \times 10^{-3}T)$. The good agreement between the computed lines and data lends support to our use of the two-state trapping model, with the assumptions that trapping is limited by the transition to the trapped state and that both prethermalization trapping and detrapping are negligible.

As already mentioned, the quantity $I_2(\lambda_1 - \lambda_2)$ shown in the lower portion of Fig. 2 is interpreted as the trapping rate $\mu_D C_D$ for positrons in the as-rolled $\text{Pb}_{96.1}\text{Cd}_{3.9}$ alloy. Since C_D is constant (see above), the weak temperature dependence exhibited by $I_2(\lambda_1 - \lambda_2)$ suggests that μ_D is weakly dependent on temperature. Also, as suggested by the solid line, this temperature dependence appears to scale with that of λ_B , which is explicable in terms of thermal expansion.¹⁸ If positron trapping is phonon mediated, one expects the increase in phonon population with increasing sample temperature to effect a positive contribution to the slope of $\mu_D(T)$. On the other hand, as the sample temperature increases, so does the energy of the thermalized positron, which may lead to a decrease in $\mu_D(T)$ with T . These effects were both considered in the work of Smedskjaer, Manninen, and Fluss⁶, who calculated

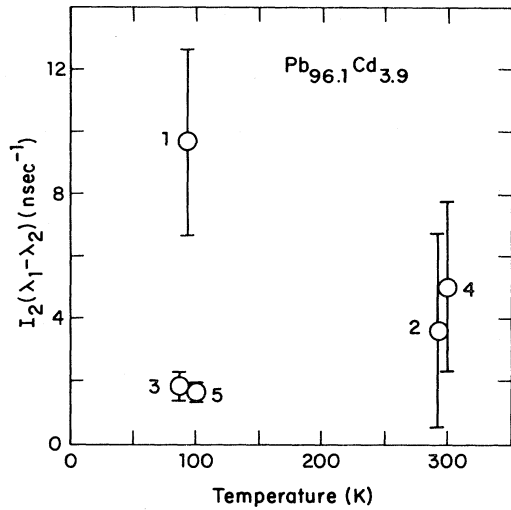


FIG. 3. Course of the trapping rate $I_2(\lambda_1 - \lambda_2)$ during heating and cooling cycles in quenched $\text{Pb}_{96.1}\text{Cd}_{3.9}$. For each point, the sample remained at the indicated temperature for a minimum of 24 h.

$\mu_D(T)$ for different positron-dislocation-trap binding energies (they did not include the relatively small effects of thermal expansion). For a binding energy of 1 eV, they found that μ_D is weakly temperature dependent (see curve C in Fig. 3 of their work), in rough agreement with the present result.

B. The quenched specimen

Point 1 in Fig. 3 is the value of the trapping rate in the as-quenched specimen at about 100 K. When the sample was heated to room temperature, a lower trapping rate (point 2) was observed. The sample exhibited the value shown by point 3 after being cooled back to 100 K. Heating the specimen again to room temperature and cooling it back to 100 K resulted in trapping rates indicated by points 4 and 5. The lifetimes τ_2 resolved from the spectra corresponding to these five points are shown in Fig. 1 as solid squares; the solid square with error bars corresponds to point 1. Note that even though the trapping rate indicated by point 1 in Fig. 3 is far greater than that indicated by points 3 or 5, the lifetimes corresponding to these three points are approximately equal. Hence it is the number and not the nature of the traps that has changed. Also, both the magnitude and the temperature dependence of τ_2 for the quenched sample are in close agreement with what was observed for the as-rolled sample. It appears therefore that positron trapping in the quenched specimen also is due predominantly to the presence

of dislocations. That is, the thermal treatment described in Sec. II apparently results in the formation of dislocation loops by vacancy condensation. If so, the decrease in the trapping rate from points 1 to 3 (Fig. 3) would be explicable in terms of a coarsening or ripening of the loops, which may have occurred during the first heating cycle. When, after quenching the sample, we shortened the time to dry and cool the aluminum capsule from 20 to 3–5 sec (see Sec. II), lifetimes were observed which are considerably longer than those characteristic of positron annihilation at dislocations; presumably these longer lifetimes are due to positron trapping at vacancy aggregates that have not yet collapsed to form loops.

Because the trapping rates indicated by points 4 and 5 approximately equal those indicated by points 2 and 3, respectively, it appears that any additional coarsening or recovery effected after the first heating cycle was not significant. Hence any change in $I_2(\lambda_1 - \lambda_2)$ from points 3 and 5 to points 2 and 4

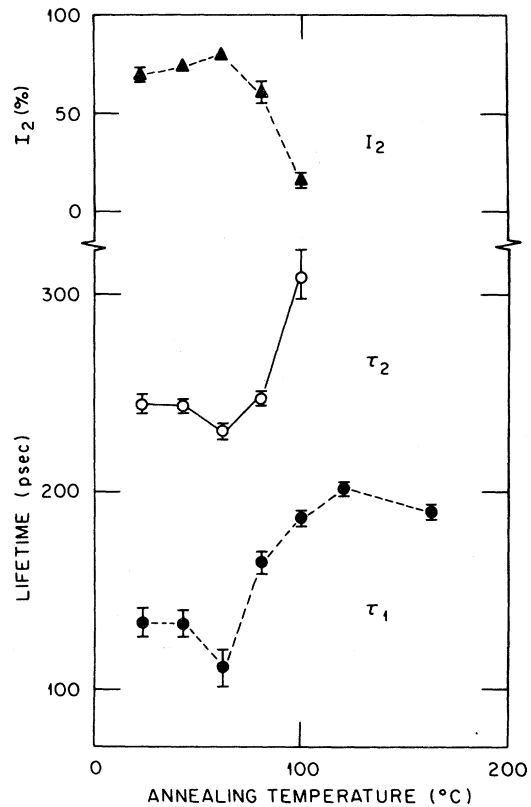


FIG. 4. Results of POSFIT analyses for as-rolled $\text{Pb}_{96.1}\text{Cd}_{3.9}$, showing the resolved lifetimes and intensity I_2 vs annealing temperature. The measurements were made at a sample temperature of about 100 K, and the annealing time was 1 h for each point. The quenched specimen showed similar behavior. The lines have been added to guide the eye.

should reflect the temperature dependence of $\mu_D(T)$. As seen, this quantity appears to increase with T . A similar increase, somewhat reduced, is also present in P_D (not shown). Though this temperature dependence is not strong, these findings do appear to be inconsistent with the behavior exhibited by the as-rolled specimen (Fig. 2). However, since only four data points (points 2–5) concentrated about two sample temperatures are involved, and since there are relatively large uncertainties associated with the data for $T \approx 300$ K, it is not clear that the apparent increase with T is significant. In any case, for both the as-rolled and the quenched specimen, it can be concluded that neither μ_D nor P_D is strongly temperature dependent.

C. ISOCHRONAL ANNEALING

The results of isochronal-annealing measurements on the sample in both the as-rolled and quenched conditions are similar. In both cases, the total trapping rate $I_2(\lambda_1 - \lambda_2)$ decreases with additional annealing, going to approximately zero for annealing temperatures $T_A \gtrsim 120^\circ\text{C}$. That is, for $T_A \gtrsim 120^\circ\text{C}$, we were unable to resolve two lifetimes in addition to the source-time lifetime. Figure 4 shows the resolved lifetimes and I_2 plotted versus annealing temperature for the as-rolled specimen at 100 K. Both the decrease in I_2 to zero and the increase in τ_1 to τ_{WA} (for a sample temperature of 100 K) are attributed to the rapid decrease in the trapping rate with T_A [see Fig. 5 and Eqs. (1) and (5)]. This drop in trapping rate also was accompanied by a drop in the mean lifetime, while, except for the one datum for $T_A = 100^\circ\text{C}$ in Fig. 4, little or no change was observed in τ_2 . This behavior, which was exhibited by the sample in both as-rolled and quenched conditions, is similar to that already discussed in reference to points 1, 3, and 5 of Fig. 3 and their respective lifetimes, and we attribute it to sample recovery. Notice that the apparent anomaly exhibited by τ_2 at $T_A = 100^\circ\text{C}$ does not appear in the corresponding value of the trapping rate shown in Fig. 5. The statistical uncertainty of $I_2(\lambda_1 - \lambda_2)$ is less than that of either λ_1 or λ_2 , because the variations in these rates are correlated with the variations in I_2 ; hence the relatively large value of τ_2 at $T_A = 100^\circ\text{C}$ probably is due to an error in the POSFIT analysis, owing to the small value in I_2 .

V. CONCLUDING REMARKS

We have described an investigation of the temperature dependence of positron lifetimes in a $\text{Pb}_{96.1}\text{Cd}_{3.9}$ alloy in both its as-rolled and quenched conditions. The e^+e^- annihilation spectra were

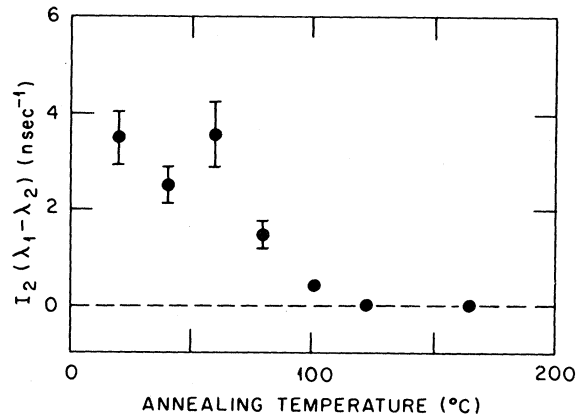


FIG. 5. Variation of the trapping rate $I_2(\lambda_1 - \lambda_2)$ with annealing temperature for as-rolled $\text{Pb}_{96.1}\text{Cd}_{3.9}$, computed using the data in Fig. 4. The quenched specimen showed similar behavior.

measured with a delayed coincidence system and were resolved into two unconstrained lifetimes in addition to a source-term lifetime. Positron trapping in the as-rolled specimen most likely is due to the presence of dislocations pinned by Cd precipitates. Because the annihilation behavior in the quenched sample is similar to that in the as-rolled sample, it appears that trapping in the quenched specimen also is predominantly at dislocations, presumably loops formed by vacancy condensation. Therefore, in both cases the longer of the two lifetimes is attributed to positrons localized by traps associated with dislocations.

We conclude the following:

(1) The lifetime of positrons localized at traps associated with dislocations in $\text{Pb}(\text{Cd})$ decreases with increasing sample temperature T for $90 \leq T \leq 300$ K. We propose that this temperature dependence is principally due to deeper penetration of atoms into the traps as T is raised. We suggest that these atomic incursions also effect a negative contribution to the slope of the line-shape parameter $S(T)$.

(2) The trapping rate per unit dislocation density and the probability of annihilation occurring at dislocation traps for constant dislocation density are both weakly temperature dependent for $90 \leq T \leq 300$ K. This behavior is consistent with a positron-dislocation binding energy of order 1 eV.

(3) The total trapping rate decreases markedly during recovery anneals, while little change is exhibited by the lifetime of the trapped positrons.

ACKNOWLEDGMENTS

S. Berko and D. Turnbull are gratefully acknowledged for invaluable suggestions and stimulat-

ing discussions throughout the course of this work. We also express our gratitude to them and to G. C. Battle, Jr., M. J. Fluss, L. D. Hulet, T. McMullen, and F. A. Modine for many useful comments on the manuscript. This research was supported by the Army Research Office under Contract No. DAAG-

29-77-G-0186, the National Science Foundation under Contract No. DMR80-12933, and the Division of Materials Sciences, Office of Basic Energy Sciences, U. S. Department of Energy, under Contract No. W-7405-eng-26 with Union Carbide Corporation.

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¹For an extensive review on the use of positrons for investigating voidlike defects in metals, see Alfred Seeger, *J. Phys. F* **3**, 248 (1973).

²C.-K. Hu, S. Berko, G. R. Gruzalski, and D. Turnbull, *Solid State Commun.* **31**, 65 (1979).

³This line-shape parameter was first used by I. K. MacKenzie, J. A. Eady, and P. P. Gingerich, *Phys. Lett.* **33A**, 279 (1970). The investigators of Ref. 2 define S as the number of counts in the central 15 channels (82 eV per channel) of the Doppler-broadened profile divided by the total number of counts in the profile (after background subtraction).

⁴P. Rice-Evans, I. Chagler, and F. El Khangi [*Phys. Lett.* **64A**, 450 (1978)] made Doppler profile measurements on pure Pb which had been worked at 77 K to 0, 5, 15, and 25 % thickness reductions. In all cases Rice-Evans *et al.* found that the line-shape parameter F (similar to S) was temperature independent over the range $4 < T < 100$ K. The data in Ref. 2 discussed in the present paper are for sample temperatures from 77 to 350 K.

⁵See, for example, Refs. 6 and 7, and references therein.

⁶Lars C. Smedskjaer, Matti Manninen, and Michael J. Fluss, *J. Phys. F* **10**, 2237 (1980).

⁷B. Bergersen and T. McMullen, *Solid State Commun.* **14**, 421 (1977).

⁸G. R. Gruzalski, D. Turnbull, C.-K. Hu, and S. Berko (unpublished).

⁹S. Berko, S. C. Sharma, and C.-K. Hu (unpublished); for some details on the coincidence system, see Ref. 10.

¹⁰Suresh C. Sharma, Ph.D. thesis, Brandeis University, 1976 (unpublished) (available from University Microfilm, Ann Arbor, MI, USA, No. 77-13, 388).

¹¹P. Kirkegaard and M. Eldrup, *Comput. Phys. Commun.* **3**, 240 (1972).

¹²W. K. Warburton, *Comput. Phys. Commun.* **13**, 371 (1978).

¹³C. L. Snead, Jr., Thomas M. Hall, and A. N. Goland, *Phys. Rev. Lett.* **29**, 62 (1972).

¹⁴Chao-Kun Hu, Ph.D. thesis, Brandeis University, 1979 (unpublished) (available from University Microfilm, Ann Arbor, MI, USA, No. 80-13, 632).

¹⁵T. McMullen (private communication).

¹⁶W. Triftshauser, *Phys. Rev. B.* **12**, 4634 (1975).

¹⁷M. Bertolaccini, A. Bisi, G. Gambarini, and L. Zappa, *J. Phys. C* **4**, 734 (1971).

¹⁸M. J. Stott and R. N. West, *J. Phys. F* **8**, 635 (1978).