Positron trapping model including spatial diffusion of the positron

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The trapping model for the annihilation rate of positrons implanted in solids has been combined with a diffusion model for the spatial motion of the implanted positrons prior to annihilation. Results are given for a simple system having traps only at the surface of a sample and allowing no detrapping, and for a general case where both effects are included. Numerical results are given for the example of positrons trapping in an image-potential well (as well as forming positronium) at a surface.

I. INTRODUCTION

In virtually all the work that has been done on the measurement of positron lifetimes in various solids, the theoretical model used to fit the data has been based on the simple two-state trapping model.[1] Generalizations of this model have been made to include more than two states, more complicated trapping and detrapping modes, and other effects.[2,3] In all cases, however, explicit spatial variations of the positron density have been neglected or treated only very approximately; only time dependences have been considered carefully. In many cases of uniform implantation of the positrons in more or less homogeneous samples, spatial considerations are not in fact significant. However, a number of systems would benefit from a more careful treatment.

One such system in which spatial effects cannot be neglected is that of the lifetime of positrons bound in image-potential wells at the surfaces of solids. A first measurement of this lifetime has recently been completed, and in the analysis of the data from this measurement, a more general model for the lifetimes of positrons in materials, including spatial diffusion of the implanted positrons as well as the usual trapping and annihilation rates, has been derived.[4] In addition to its usefulness in the case of low-energy positrons implanted near a surface by a positron beam, this result should have significance for a variety of more traditional measurements in bulk systems as well. Even for homogeneous samples a non-insignificant number of positrons will reach the surface, and there become trapped or form positronium. Moreover, most samples are not truly very homogeneous. The concentrations of large voids, grain boundaries, dislocations, and other such macroscopic defects are often sufficiently small that diffusion of the implanted positrons can occur for a significant period before they encounter one and are potentially able to trap.[5,6] Even for microscopic defects such as monovacancies, spatial effects could be significant for small concentrations.[7] In all of these cases the results to be presented here for a semi-infinite solid may prove useful, either as given, or modified to deal with a different geometry.

The motion of thermalized positrons in crystalline solids has been treated successfully using a simple, classical diffusion equation.[8,9] We use this equation to describe the motion of positrons implanted in a semi-infinite medium with a given implantation profile. To simplify the situation we consider only a one-dimensional problem, neglecting concentration variations parallel to the surface of the sample. The method of solution is to solve the diffusion equations subject to the boundary condition of perfect absorption of those positrons which reach the surface. (We make no assumptions about the fate of those positrons at this point, but only assume that they are removed from the diffusion process.) This allows us to obtain a rate $N(t)$ of positrons reaching the surface. Using this function we can set up rate equations similar to those found in the usual trapping model. The solution of these rate equations yields the desired annihilation rate $R(t)$.

We have actually solved the rate equations for a fairly general trapping model allowing various types of detrapping and any number of bulk and surface traps. To illustrate the method though, we first do a simple case with only the lattice annihilation rate in the bulk (no bulk traps) and without any detrapping permitted. This is done for two elementary implantation profiles. After the simple case is completed, we return to the more general problem.

II. CASE OF A PERFECT SEMI-INFINITE SOLID

The one-dimensional diffusion equation, including annihilation in the bulk but no trapping, is given by

$$D\frac{\partial^2 u}{\partial x^2}-\frac{\partial u}{\partial t}=\kappa_0 u \ , \tag{1}$$

where $u(x,t)$ is the density of positrons at x and t, D is the positron diffusion coefficient, and κ_0 is the bulk annihilation rate. The equation is to be solved subject to the boundary conditions

$$\begin{aligned} &u(0,t)=0 \ \ (\text{absorbing boundary}) \ , \\ &u(x,0)=C_0(x) \ \ (\text{implantation profile}) \ , \end{aligned} \tag{2}$$

and the desired rate $N(t)$ of positrons reaching the surface is given by

$$N(t)=D\frac{\partial u}{\partial x}(0,t)\ . \tag{3}$$

Equation (1) can easily be solved as a Fourier sine transform

$$u(x,t)=\int_0^\infty A(q)\sin(qx)e^{-(Dq^2+\kappa_0)t}dq\ , \tag{4}$$

where

$$A(q)=\frac{2}{\pi}\int_0^\infty C_0(x)\sin(qx)dx$$

from which we obtain $N(t)$ as

$$N(t)=D\int_0^\infty qA(q)e^{-(Dq^2+\kappa_0)t}dq\ . \tag{5}$$

We consider a system having some number m of processes occurring at the surface, all fed by the rate $N(t)$. Examples of these processes would be trapping in a surface image-potential well and formation of orthopositronium or parapositronium. Each process has associated with it an intrinsic annihilation rate κ_i and a branching ratio ϵ_i (where $\sum_{i=1}^m \epsilon_i \equiv 1$). The rate equations are then given by

$$\begin{aligned} n_0'(t)&=-\kappa_0 n_0(t)-N(t)\ ,\\ n_i'(t)&=-\kappa_i n_i(t)+\epsilon_i N(t)\ , \end{aligned} \tag{6}$$

where $n_0(t)$ is the probability that a positron is found in the bulk at time t, $n_i(t)$ is the probability that a positron is found in the ith surface associated component, and primes denote differentiation with respect to t. Writing k as a subscript which ranges from 0 to m, and defining $\epsilon_0 \equiv -1$ (so that $\sum_{k=0}^m \epsilon_k \equiv 0$), we can combine Eq. (6) into a single equation given by

$$n_k'(t)+\kappa_k n_k(t)=\epsilon_k N(t)\ . \tag{7}$$

This may readily be solved to yield

$$n_k(t)=\left[n_k(0)+\epsilon_k\int_0^t N(\tau)e^{\kappa_k\tau}d\tau\right]e^{-\kappa_k t}\ . \tag{8}$$

Thus combining Eq. (5) with Eq. (8), we find the desired expression for the probability densities

$$n_k(t)=\left[n_k(0)+D\epsilon_k\int_0^\infty \frac{qA(q)}{Dq^2+\mu_k}\times(1-e^{-(Dq^2+\mu_k)t})dq\right]e^{-\kappa_k t}\ , \tag{9}$$

where $\mu_k \equiv \kappa_0-\kappa_k$. The initial conditions are $n_0(0)=1$ and $n_i(0)=0$.

We shall consider two implantation profiles $C_0(x)$ here. The simplest is just an exponential profile

$$C_0(x)=\frac{1}{x_0}e^{-x/x_0}\ , \tag{10a}$$

which leads to

$$A(q)=\frac{2qx_0}{\pi(1+x_0^2q^2)}\ . \tag{10b}$$

A more correct expression for the actual profile from a monoenergetic positron beam may possibly be given by the derivative of a Gaussian[10]

$$C_0(x)=\frac{2x}{x_0^2}e^{-(x/x_0)^2}\ , \tag{11a}$$

which leads to

$$A(q)=\frac{qx_0}{\sqrt{\pi}}e^{-(qx_0/2)^2}\ . \tag{11b}$$

Using these expressions for $A(q)$ in Eq. (9), we can obtain (after some algebra) results for the $n_k(t)$ appropriate to these two profiles. We shall actually quote the corresponding overall annihilation rate $R(t)\equiv\sum_{k=0}^m \kappa_k n_k(t)$. In both profiles, this can be expressed in the same general form by defining functions $\rho_k(t)$ such that

$$R(t)=\sum_{k=0}^m A_k\rho_k(t)e^{-\kappa_k t}\ , \tag{12}$$

where

$$A_0=\kappa_0-\sum_{i=1}^m \epsilon_i\kappa_i\rho_i(0)\ ,$$

$$A_i=\epsilon_i\kappa_i\ .$$

The functions $\rho_k(t)$ differ for the two profiles. For the exponential profile, we find

$$\begin{aligned} \rho_0(t)&=e^{\kappa_D t}\,\mathrm{erfc}[(\kappa_D t)^{1/2}]\ ,\\ \rho_i(t)&=\frac{\kappa_D}{\kappa_D-\mu_i}\left[1-\left[\frac{\mu_i}{\kappa_D}\right]^{1/2}\mathrm{erf}[(\mu_i t)^{1/2}]\right]\ , \end{aligned} \tag{13}$$

where $\kappa_D \equiv D/x_0^2$ is a rate determined by the average time required for the positrons to diffuse to the surface. For the differential Gaussian profile, we have

$$\begin{aligned} \rho_0(t)&=\frac{1}{(1+4\kappa_D t)^{1/2}}\ ,\\ \rho_i(t)&=1-\left[\frac{\pi\mu_i}{4\kappa_D}\right]^{1/2}e^{\mu_i/4\kappa_D}\left\{\mathrm{erf}\left[\left[\frac{\mu_i}{4\kappa_D}\right]^{1/2}\right]-\mathrm{erf}\left[\left[\frac{\mu_i}{4\kappa_D}+\mu_i t\right]^{1/2}\right]\right\}\ . \end{aligned} \tag{14}$$

Using these results for the $\rho_k(t)$ in Eq. (12) yields the desired annihilation rate including the effect of the surface. We find that Eq. (12) has a fairly straightforward form. It is the usual sum of exponentials found in trapping model results, but with two changes. First, the decay rates in the exponentials are precisely the intrinsic annihilation rates of the various states, not combinations of intrinsic annihilation rates and trapping rates as in more usual cases. As we shall see, this is a consequence of the special case we have done, without bulk traps on detrapping.

The second difference between Eq. (12) and earlier forms is the presence of the functions ρ of time multiplying the simple exponentials. These functions are the real difference here, and contain all the details of the diffusion effects that we set out to derive. As we have seen, the only influence of differing implantation profiles is to modify the ρ's. Moreover, for sufficiently shallow implantation we can write $\kappa_D \gg \kappa_k$ for all k; thus for $t \gg \kappa_D^{-1}$, we can approximate $\rho_0 \sim (\pi\kappa_D t)^{-1/2}$ for the exponential profile, $\rho_0 \sim (4\kappa_D t)^{-1/2}$ for the differential Gaussian profile, and $\rho_i \sim 1$ for both. Equation (12) becomes precisely a sum of pure exponentials in this approximation, and as expected, the bulk annihilation rate κ_0 does not enter.

III. A MORE GENERAL CASE

A more general situation than the simple case treated above is shown schematically in Fig. 1. There we show a system having one freely diffusing state and m nondiffusing states (traps, positronium formation, etc.). Each has an intrinsic annihilation rate written as κ_0 or κ_i. The nondiffusing states (which we index by i and j, both varying from 1 to m) can be either surface associated or bulk associated. These states are fed by two separate mechanisms, depending on their classification: Those associated with the surface are fed by $N(t)$ as before, with branching ratios ϵ_i. The trapping rates σ_i vanish in their case. The nondiffusing states associated with the bulk are fed instead by trapping rates σ_i multiplied by the bulk occupation number $n_0(t)$; here, the branching ratios ϵ_i are taken as zero. To simplify our notation we always write the overall input rate from the diffusing state to a nondiffusing state as $\sigma_i n_0(t)+\epsilon_i N(t)$. It is always understood, however, that one or the other of these terms vanishes for any given nondiffusing state.

In addition to the transition rates described above, we permit arbitrary transitions among the nondiffusing states, specified by the transition rates κ_{ij} (from state j to state i). In order to retain the ability to deal with the diffusion equation and the rate equations separately though, we cannot allow any return of positrons back to the diffusing state from any of the nondiffusing states, i.e., we neglect the effect of shallow traps in the bulk. This restriction should not in general prove to be significant

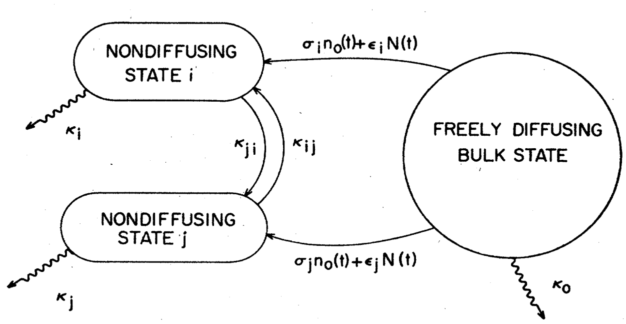

FIG. 1. Schematic diagram of states to be considered in a general case of the diffusion-trapping model. One diffusing state and m nondiffusing states are considered. Arbitrary trapping and detrapping rates are included with the single exception that detrapping back into the diffusing state from a nondiffusing state is not permitted.

since bulk trapping is usually a one way process, in the direction of lower potential energy. (Thus usually at least one of a pair κ_{ij} and κ_{ji} will vanish for a given i and j.)

The rate equations in this general case are given by

$$n_0' = -\kappa_{00} n_0 - N(t) ,$$
$$n_i' = -\kappa_{ii} n_i + \sum_{\substack{j=1 \\ (j\neq i)}}^{m} \kappa_{ij} n_j + \sigma_i n_0 + \epsilon_i N(t) , \tag{15}$$

where

$$\kappa_{00} \equiv \kappa_0 + \sum_{j=1}^{m} \sigma_j ,$$
$$\kappa_{ii} \equiv \kappa_i + \sum_{\substack{j=1 \\ (j\neq i)}}^{m} \kappa_{ji} .$$

The equation for the bulk state (subscript 0) above is identical to that in Eq. (6) with the change $\kappa_0 \rightarrow \kappa_{00}$. Moreover, the results given in Eq. (5), (10), and (11) for $N(t)$ also remain valid here with the same substitution. So all we need do is deal with the equations for the n_i. To do this we resort to matrix notation.

The equations for the n_i for $i\neq 0$ can be written in matrix form as

$$\vec{n}\,' + \tilde{\kappa}\,\vec{n} = \vec{\sigma} n_0(t) + \vec{\epsilon} N(t) , \tag{16}$$

where

$$\vec{n} \equiv \begin{bmatrix} n_1 \\ n_2 \\ \cdot \\ \cdot \\ \cdot \\ n_m \end{bmatrix} , \quad \vec{\sigma} \equiv \begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \cdot \\ \cdot \\ \cdot \\ \sigma_m \end{bmatrix} , \quad \vec{\epsilon} \equiv \begin{bmatrix} \epsilon_1 \\ \epsilon_2 \\ \cdot \\ \cdot \\ \cdot \\ \epsilon_m \end{bmatrix} ,$$

$$\tilde{\kappa} \equiv \begin{bmatrix} \kappa_{11} & -\kappa_{12} & \cdots & -\kappa_{1m} \\ -\kappa_{21} & \kappa_{22} & \cdots & -\kappa_{2m} \\ \cdot & \cdot & & \cdot \\ \cdot & \cdot & & \cdot \\ \cdot & \cdot & & \cdot \\ -\kappa_{m1} & -\kappa_{m2} & \cdots & \kappa_{mm} \end{bmatrix} .$$

To solve this equation we assume that $\tilde{\kappa}$ is diagonalizable and write λ_{ii} for its eigenvalues and $\tilde{M}$ for the transformation matrix needed to bring $\tilde{\kappa}$ to diagonal form. Thus designating the transformed matrix and vectors by $*$'s, we can write

$$\tilde{\kappa}^* \equiv \tilde{M}\,\tilde{\kappa}\,\tilde{M}^{-1} ,$$
$$\vec{n}^* \equiv \tilde{M}\,\vec{n} , \quad \vec{\sigma}^* \equiv \tilde{M}\,\vec{\sigma} , \quad \vec{\epsilon}^* \equiv \tilde{M}\,\vec{\epsilon} . \tag{17}$$

Here $\tilde{\kappa}^*$ is a diagonal matrix of the λ_{ii}'s. Transforming Eq. (16), we can write

$$(n_i^*)' + \lambda_{ii} n_i^* = \sigma_i^* n_0(t) + \epsilon_i^* N(t) . \tag{18}$$

These equations are very similar in form to Eq. (6) and can be solved for a given $N(t)$ in an analogous way.

The solution to Eq. (18) corresponding to Eq. (9), the solution of Eq. (6), is

$$n_i^* = \left[\int_0^\infty \frac{\epsilon_i^* Dq^2 + \sigma_i^*}{Dq^2 + \mu_{ii}} \frac{A(q)}{q} (1 - e^{-(Dq^2+\mu_{ii})t}) dq \right] e^{-\lambda_{ii} t} , \tag{19}$$

where $\mu_{ii} \equiv \kappa_{00} - \lambda_{ii}$. [We have implicitly used the initial conditions $n_i^*(0) = n_i(0) = 0$.] Using this result, we can again generate expressions for $R(t)$ for any given implantation profile. We only give the results for the differential Gaussian profile here. They are

$$R(t) = \sum_{k=0}^{m} A_k \rho_k(t) e^{-\lambda_{kk} t} , \tag{20}$$

where $\lambda_{00} \equiv \kappa_{00}$. The bulk term is similar to that in Eqs. (12) and (14) with

$$A_0 = \kappa_0 - \sum_{i=1}^{m} \epsilon_i \kappa_i \rho_i(0) , \qquad \rho_0(t) = \frac{1}{(1+4\kappa_D t)^{1/2}} . \tag{21}$$

For surface associated components, we have the expressions

$$A_i = \sum_{j=1}^{m} \epsilon_i^* m_{ij} \kappa_j ,$$
$$\rho_i(t) = 1 - \left[\frac{\pi \mu_{ii}}{4\kappa_D} \right]^{1/2} e^{\mu_{ii}/4\kappa_D} \left\{ \mathrm{erf} \left[\left[\frac{\mu_{ii}}{4\kappa_D} \right]^{1/2} \right] - \mathrm{erf} \left[\left[\frac{\mu_{ii}}{4\kappa_D} + \mu_{ii} t \right]^{1/2} \right] \right\} , \tag{22a}$$

while for the bulk associated components, we have

$$A_i = \sum_{j=1}^{m} \frac{\sigma_i^*}{\mu_{ii}} m_{ij} \kappa_j$$
$$\rho_i(t) = \left[\frac{\pi \mu_{ii}}{4\kappa_D} \right]^{1/2} e^{\mu_{ii}/4\kappa_D} \left\{ \mathrm{erf} \left[\left[\frac{\mu_{ii}}{4\kappa_D} \right]^{1/2} \right] - \mathrm{erf} \left[\left[\frac{\mu_{ii}}{4\kappa_D} + \mu_{ii} t \right]^{1/2} \right] \right\} . \tag{22b}$$

In these expressions, we have used m_{ij} for the components of the inverse transpose of the transformation matrix $(\widetilde{M}^{-1})^t$.

In Eqs. (20)–(22) then, we have presented the solution to the diffusion-trapping model in a fairly general case. Although these equations may look formidable, they are relatively straightforward to reduce to any particular case one might wish to consider. Since the form of the equations here is the same as we have discussed in conjunction with Eqs. (12)–(14), and since even the ρ_i's here are nearly identical to those given there, the only differences here are in obtaining the exponents λ_{ii} and the coefficients A_i. These are both readily found after the problem of diagonalizing the matrix $\widetilde{\kappa}$ has been solved. It is the diagonalization of this matrix that presents the only potential difficulties in using Eqs. (20)–(22). To make all of this clear, we present below the results for a particular special case as an example.

IV. THE CASE OF A SEMI-INFINITE SOLID WITH BULK TRAPPING AND SURFACE DETRAPPING

The special case we shall do involves a freely diffusing bulk state and three nondiffusing states: State 1 is a bulk defect with intrinsic annihilation rate κ_1, trapping rate σ_1 from the freely diffusing state, and no detrapping. State 2 is a surface image-potential well with annihilation rate κ_2, branching ratio ϵ_2, and detrapping rate $4\kappa_{32}$ into positronium (i.e., κ_{32} into parapositronium and $3\kappa_{32}$ into orthopositronium). We shall treat the orthopositronium decay rate here as being negligibly small, but the $3\kappa_{32}$ detrapping rate into orthopositronium must be included. State 3 is parapositronium with annihilation rate κ_3, branching ratio ϵ_3, and no detrapping (and of course input rate κ_{32} from state 2 as we have described). The bulk annihilation rate is κ_0.

The rate matrix is given in this special case by

$$\widetilde{\kappa} = \begin{bmatrix} \kappa_1 & 0 & 0 \\ 0 & \kappa_2 + 4\kappa_{32} & 0 \\ 0 & -\kappa_{32} & \kappa_3 \end{bmatrix} . \tag{23}$$

The eigenvalues λ_{ii} are given by

$$\lambda_{11} = \kappa_1 , \quad \lambda_{22} = \kappa_2 + 4\kappa_{32} , \quad \lambda_{33} = \kappa_3 \tag{24}$$

and the transformation matrix and its inverse transpose are

$$\widetilde{M} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & \kappa_{32} & \kappa_2 - \kappa_3 + 4\kappa_{32} \end{bmatrix} , \qquad (\widetilde{M}^{-1})^t = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & \dfrac{-\kappa_{32}}{\kappa_2 - \kappa_3 + 4\kappa_{32}} \\ 0 & 0 & \dfrac{1}{\kappa_2 - \kappa_3 + 4\kappa_{32}} \end{bmatrix} . \tag{25}$$

[Normalization of the matrix $\widetilde{M}$ does not effect the quantities defined in Eqs. (20)–(22). We choose an arbitrary normalization for $\widetilde{M}$ above.] With use of these matrices, it is easy to generate expressions for the constants in $R(t)$. They are

$$\lambda_{00} = \kappa_0 + \sigma_1 ,$$
$$A_0 = \kappa_0 - \epsilon_2 \kappa_2 - \epsilon_3 \kappa_3 ,$$
$$A_1 = \frac{\kappa_1 \sigma_1}{\kappa_0 + \sigma_1 - \kappa_1} ,$$
$$A_2 = \epsilon_2 \kappa_2 - \frac{\epsilon_2 \kappa_{32} \kappa_3}{\kappa_2 - \kappa_3 + 4\kappa_{32}} , \tag{26}$$

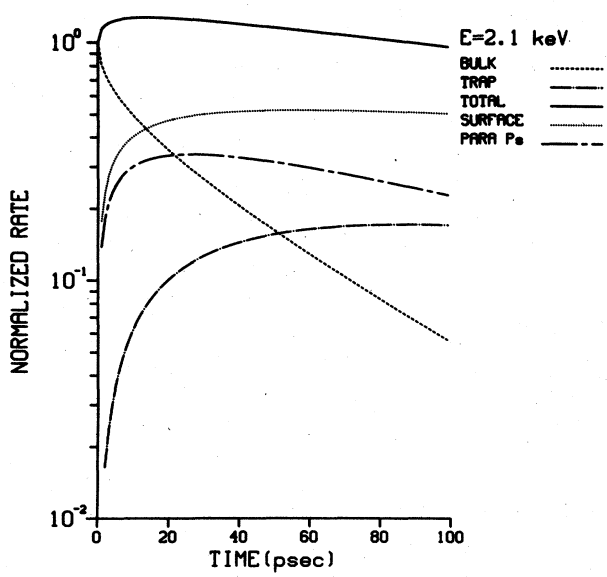

FIG. 2. Plots of $R(t)$ and the four terms contained in it for the case of a single bulk trap, and an image-potential well and positronium formation at the surface. No detrapping is included. Numerical values for the constants in $R(t)$ are taken from Ref. 4 and are given in the text.

$$A_3=\epsilon_3\kappa_3+\frac{\epsilon_2\kappa_{32}\kappa_3}{\kappa_2-\kappa_3+4\kappa_{32}}\ ,$$

$$\mu_{11}=\kappa_0+\sigma_1-\kappa_1\ ,$$

$$\mu_{22}=\kappa_0+\sigma_1-\kappa_2-4\kappa_{32}\ ,$$

$$\mu_{33}=\kappa_0+\sigma_1-\kappa_3\ .$$

The formulas for the ρ's are of course as given in Eqs. (20)—(22). (Remember that $\kappa_D \equiv D/x_0^2$.) Thus we have a solution for the diffusion-trapping model in this particular four-state system.

Plots of the four contributions to $R(t)$ in this case are given as Fig. 2. (In the plots, no detrapping is included so that $\kappa_{32}=0$.) The constants needed to generate the curves are taken from the surface lifetime measurement referred to in the Introduction.[4] They are $\kappa_0=6.13$ ns^{-1}, $\kappa_1=4.07$ ns^{-1}, $\kappa_2=1.71$ ns^{-1}, $\kappa_3=8.00$ ns^{-1}, $\sigma_1=10.0$ ns^{-1}, $\epsilon_2=60\%$, and $\epsilon_3=10\%$. [The remaining 30% of $N(t)$ is orthopositronium formation which is taken to have zero decay rate for the purposes of this calculation.] We also use $D=0.31$ cm^2/s and $x_0=408$ Å which yields $\kappa_D=18.6$ ns^{-1}. The curves in the figure show clearly the deviation from simple exponential behavior for times $t \lesssim \kappa_D^{-1}$. For times $t \gg \kappa_D^{-1}$ (not shown in the figure), the curves all approach pure exponential decays.

V. CONCLUSION

We have presented in this paper a new expression, Eqs. (20)—(22), for the annihilation rate of positrons implanted in a semi-infinite, one-dimensional sample. The expression is obtained from a combination of the diffusion model for the motion of thermalized positrons in solids, and a many-state trapping model. This expression should more correctly describe the annihilation rates in any positron lifetime experiment in which the annihilation occurs with significant likelihood at sites separated from the implantation locations of the positrons. Experiments on lifetimes or trapping rates associated with sample surfaces clearly fall into this category. However, any lifetime measurement in which the average distance between trapping centers is a significant fraction of a positron diffusion length should properly make use of the results given here. Moreover, the effects of the sample surface may not be negligible even for bulk lifetime studies, particularly for small samples. Again, the diffusion picture may be useful. (It should be kept in mind that the diffusion coefficient D depends on temperature; thus diffusion effects may be misinterpreted as actual changes in the trapping rates in some cases.) In conclusion, a number of positron lifetime measurements, both surface and bulk, may well be improved by a more careful consideration of the effects of spatial separation and positron diffusion on the equations for the annihilation rate.

ACKNOWLEDGMENT

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