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2000 J. Phys.: Condens. Matter 12 137

(http://iopscience.iop.org/0953-8984/12/2/304)

J. Phys.: Condens. Matter **12** (2000) 137–141. Printed in the UK PII: S0953-8984(00)03216-1

The diffusion model for the trapping and detrapping of positrons in grain boundaries

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Received 7 April 1999, in final form 19 October 1999

Abstract. A general solution of the model for positrons which diffuse inside a grain and then can be trapped and detrapped at a grain boundary of arbitrary shape is presented. The closed-form relations for the mean positron lifetime and the positron lifetime spectrum are obtained for a grain of spherical shape. The obtained results slightly differ from those presented in 1996 *Phil. Mag.* **73** 1489 where a similar problem was considered and they extend the consideration given in that paper in *J. Phys.: Condens. Matter* **10** L547.

In the literature of recent years there can be found the exact solutions of the diffusion–transition model for the trapping and annihilation of positrons in grain boundaries. These solutions are important because they open new applications for the positron annihilation spectroscopy, e.g., for the study of fine grained samples. In the model which we will call the diffusion trapping model (DTM) it is assumed that positrons randomly walk in a perfect grain where they can also annihilate with the rate $\lambda_f = 1/\tau_f$, where τ_f is the positron lifetime in the free state. The grain is surrounded by the boundary which act as a perfect sink for positrons where they can also annihilate with the rate $\lambda_b = 1/\tau_b < \lambda_f$. The transition rate from the free state to the localized state at the boundary is described by the α parameter which value is equal to the width of the boundary times the trapping rate. The general solution of this model even when the grain boundary has an arbitrary shape was presented in [1], but in this paper we will add in the model the detrapping process of positrons as well. Including the detrapping process into the DTM seems to be important from two points of view. First, to obtain a more general model to see new predicted phenomena. Second, the detrapping of positrons one could expect from the low angle grain boundaries created by dislocations which act as the shallow positron traps. The multilayer system could be also studied by positron annihilation techniques using this model.

Let us assume that a positron which has been localized at the boundary can escape with the rate β to the free state in the grain interior and start to diffuse again. We denote the number of trapped positrons as n_b and their local concentration in the grain interior as $C(\boldsymbol{r}, t)$. Both functions fulfil the following set of equations:

$$\begin{cases} \dfrac{\partial}{\partial t} C(\boldsymbol{r}, t) = D_+ \nabla^2 C(\boldsymbol{r}, t) - \lambda_f C(\boldsymbol{r}, t) + \beta n_b(t) T(\boldsymbol{r}) \\ \dfrac{\mathrm{d}}{\mathrm{d}t} n_b(t) = \alpha \oiint_\Sigma \mathrm{d}S\, C(\boldsymbol{r}, t) - (\lambda_b + \beta) n_b(t) \\ D_+ \oiint_\Sigma \mathrm{d}\boldsymbol{S} \cdot \nabla C(\boldsymbol{r}, t) + \alpha \oiint_\Sigma \mathrm{d}S\, C(\boldsymbol{r}, t) = 0 \end{cases} \tag{1}$$

where Σ is the grain surface, D_+ is the diffusion coefficient of positrons and $T(\boldsymbol{r})\,\mathrm{d}V$ is the probability that the positron which escapes from the boundary will be localized in the element $\mathrm{d}V$ at the place of which the coordinate is equal to $\boldsymbol{r}$. The positrons which have returned to the interior of a grain take part in the diffusion process again as noted in the last term of the time dependent diffusion equation of the set (1). The second equation is the kinetic equation for the number of positrons trapped at boundary and the third one exhibits the fact that only the positrons which pass through the boundary are able to be localized there. The presented set of equations differs from that which was taken into consideration in [2] where the same model was examined. However, in this paper the detrapping process was not taken in the adequate way by modification of the second and third equations in (1) only. We argue that detrapped positrons modify in some way the concentration of positrons in the grain interior what should be reflected in the time dependent diffusion equation.

We should note that the explicit formula of the function $T(\boldsymbol{r})$ is not important because in our calculations we will search only the total number of positrons at the grain boundary and in the interior. During calculations we assume that this function fulfils the condition: $\oiiint_\Omega \mathrm{d}V\,T(\boldsymbol{r}) = 1$, where Ω is the volume of the grain, which means that the positron after escaping from the boundary appears in the free state somewhere in the interior with probability equal to unity. Certainly the exact solution of (1) depends on the $T(\boldsymbol{r})$ function but in the experiment we are able to detect only the total number of positrons which annihilate in the two states.

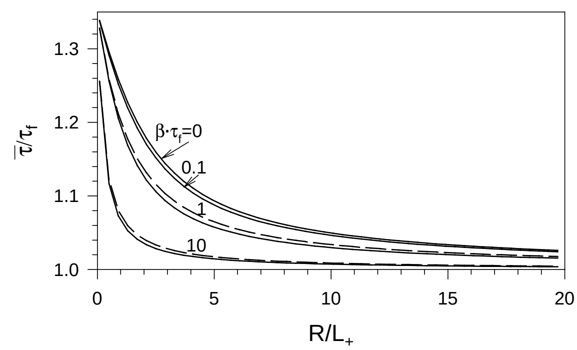

Figure 1. The mean positron lifetime (17) (solid lines), normalized to τ_f, versus R/L_+ calculated from the DTM for a spherical shape grain. The calculations were performed for four values of the $\beta\tau_b$ parameter: 0, 0.1, 1 and 10. The parameter of $\alpha\tau_f/L_+$ was equal to 1. For comparison the dashed lines present the same calculation but using the relation which was found by the authors of [2].

Thus for solution of the DTM we have to find the time dependent total number of positrons which is described as a sum:

$$n(t) = n_b(t) + \oiiint_\Omega \mathrm{d}V\,C(\boldsymbol{r},t). \tag{2}$$

From this the average positron lifetime, $\bar{\tau} = \int_0^\infty \mathrm{d}t\,n(t)$ or the positron lifetime spectrum, $-\mathrm{d}n(t)/\mathrm{d}t$ can be evaluated. Because in [1] the solution procedure of the equations (1), with $\beta = 0$, was presented in detail now we will present only by the final results. The key function in the solution is the function which is defined as follows:

$$\tilde{F}(\boldsymbol{r},s) = (\lambda_f + s)V_\Omega \tilde{C}(\boldsymbol{r},s) \tag{3}$$

where $\tilde{C}(\boldsymbol{r}, s) = \int_0^\infty \mathrm{d}t\, \mathrm{e}^{-st} C(\boldsymbol{r}, t)$ is the Laplace transform of the function which describes the local concentration of the positrons. The function (3) fulfils the following equation:

$$\nabla^2 \tilde{F}(\boldsymbol{r}, s) - \gamma(s)^2 \tilde{F}(\boldsymbol{r}, s) = -\gamma(s)^2 \tag{4}$$

where $\gamma(s) = \sqrt{[(\lambda_f + s)/D_+](\lambda_b + \beta + s)/(\lambda_b + s)}$. The Laplace transform of the function which describes the change of the total number of positrons can be deduced from (1), (2) and (3) as follows:

$$\tilde{n}(s) = \frac{1}{\lambda_b + s}\left[1 - \frac{\lambda_f - \lambda_b}{\lambda_f + s}\frac{1}{V_\Omega}\oiiint_\Omega \mathrm{d}V\, \tilde{F}(\boldsymbol{r}, s)\right]. \tag{5}$$

The mean positron lifetime is described by:

$$\bar{\tau} = \frac{1}{\lambda_f} + \left(\frac{1}{\lambda_b} - \frac{1}{\lambda_f}\right)\left[1 - \frac{1}{V_\Omega}\oiiint_\Omega \mathrm{d}V \tilde{F}(\boldsymbol{r}, 0)\right]. \tag{6}$$

The positron lifetime spectrum after inverting the Laplace transform of (5) and then taking the time derivative is expressed as:

$$\begin{aligned}-\frac{\mathrm{d}n(t)}{\mathrm{d}t} = \operatorname*{res}_{s=-\lambda_b}&\left(\frac{\lambda_b}{\lambda_b + s}\left[1 - \frac{\lambda_f - \lambda_b}{\lambda_f + s}\frac{1}{V_\Omega}\oiiint_\Omega \mathrm{d}V\, \tilde{F}(\boldsymbol{r}, s)\right]\mathrm{e}^{-\lambda_b t}\right)\\ &+ \sum_{i=0}^{\infty}\operatorname*{res}_{s=-\lambda_i}\frac{\lambda_i}{\lambda_b + s}\left[1 - \frac{\lambda_f - \lambda_b}{\lambda_f + s}\frac{1}{V_\Omega}\oiiint_\Omega \mathrm{d}V\, \tilde{F}(\boldsymbol{r}, s)\right]\mathrm{e}^{-\lambda_i t}.\end{aligned} \tag{7}$$

The solution of equation (4) one can express as follows:

$$\tilde{F}(\boldsymbol{r}, s) = A(s)\tilde{f}(\boldsymbol{r}, s) + \tilde{g}(\boldsymbol{r}, s) \tag{8}$$

where:

$$\nabla^2 \tilde{f}(\boldsymbol{r}, s) - \gamma^2 \tilde{f}(\boldsymbol{r}, s) = 0. \tag{9}$$

The function $A(s)$ we can evaluate from the third equation of the set (1):

$$A(s) = -\frac{(\lambda_f + s)(\lambda_b + \beta + s)\oiiint_\Omega \mathrm{d}V[\tilde{g}(\boldsymbol{r}, s) - 1] + \alpha(\lambda_b + s)\oiint_\Sigma \mathrm{d}S\, \tilde{g}(\boldsymbol{r}, s)}{(\lambda_f + s)(\lambda_b + \beta + s)\oiiint_\Omega \mathrm{d}V\, \tilde{f}(\boldsymbol{r}, s) + \alpha(\lambda_b + s)\oiint_\Sigma \mathrm{d}S\, \tilde{f}(\boldsymbol{r}, s)}. \tag{10}$$

In our consideration important role play only the following expression:

$$\begin{aligned}1 - \frac{1}{V_\Omega}&\oiiint_\Omega \mathrm{d}V\, \tilde{F}(\boldsymbol{r}, s)\\ &= \frac{1}{1 + \frac{(\lambda_f + s)}{\alpha}\frac{(\lambda_b + \beta + s)}{(\lambda_b + s)}B(\gamma)}\left[1 - \left(1 - \frac{B(\gamma)}{D(\gamma)}\right)\frac{1}{V_\Omega}\oiiint_\Omega \mathrm{d}V\, \tilde{g}(\boldsymbol{r}, s)\right]\end{aligned} \tag{11}$$

where

$$B(\gamma) = \frac{\oiiint_\Omega \mathrm{d}V\, \tilde{f}(\boldsymbol{r}, s)}{\oiint_\Sigma \mathrm{d}S\, \tilde{f}(\boldsymbol{r}, s)} \tag{12}$$

and

$$D(\gamma) = \frac{\oiiint_\Omega \mathrm{d}V \tilde{g}(\boldsymbol{r}, s)}{\oiint_\Sigma \mathrm{d}S \tilde{g}(\boldsymbol{r}, s)} \tag{13}$$

and

$$\gamma \equiv \gamma(s). \tag{14}$$

If we know the $B(\gamma)$ function then we could evaluate the value of λ_i as follows. First we have to solve the transcendental equation:

$$\xi_i^2 B(\xi_i) + \frac{\alpha}{D_+} = 0 \tag{15}$$

and then the second order equation:

$$\lambda_i^2 + \lambda_i[D_+\xi_i^2 - (\lambda_b + \lambda_f + \beta)] + \lambda_f(\lambda_b + \beta) - D_+\xi_i^2\lambda_b = 0. \tag{16}$$

The presented above equations are valid for a grain of arbitrary shape. Let us assume that the grain is a sphere of radius R. The functions $B(\gamma)$ and $D(\gamma)$ can be written in an analytical form: $B(\gamma) = (1/\gamma)L(\gamma R)$, where $L(z) = \coth(z) - 1/z$ is the Langevin function, $D(\gamma) = R/3$ and $\tilde{g}(\boldsymbol{r}, s) = 1$, [1]. From (6) and (11) we can obtain the value of the mean positron lifetime:

$$\bar{\tau} = \tau_f + \frac{3L_+(\tau_b - \tau_f)}{R\sqrt{1+\tau_b\beta}} \frac{L(R\sqrt{1+\tau_b\beta}/L_+)}{1 + (L_+\sqrt{1+\tau_b\beta}/\alpha\tau_f)L(R\sqrt{1+\tau_b\beta}/L_+)} \tag{17}$$

where $L_+ = \sqrt{D_+\tau_f}$ is the diffusion length of positrons inside the grain. For calculation of the positron lifetime spectrum in such a case we should notice that the first term in (7) is equal to zero. After some algebra the final relation for the positron lifetime spectrum is given by:

$$-\frac{\mathrm{d}n(t)}{\mathrm{d}t} = 6(\tau_b - \tau_f)\sum_{i=1}^{\infty}\{\mathrm{e}^{-t/\tau_i}\}\{\tau_i[\tau_b - \tau_f + (1 - \tau_f/\tau_i)^2(R^2/L_+^2)\tau_b/\xi_i^2] \\ \times[1 - L_+^2/\alpha\tau_f R + (L_+^2/\alpha\tau_f R)^2\xi_i^2]\xi_i^2\}^{-1}. \tag{18}$$

ξ_i fulfils the transcendental equation:

$$\xi_i \coth(\xi_i) + \frac{\alpha\tau_f R}{L_+^2} = 1 \tag{19}$$

and τ_i can be calculated from the solution of the equation

$$\tau_f\tau_b\left(\frac{1}{\tau_i}\right)^2 - \left(\frac{1}{\tau_i}\right)\left(\tau_b + \tau_f + \beta\tau_b\tau_f + \xi_i^2\frac{L_+^2}{R^2}\tau_b\right) + (1 + \beta\tau_b) + \xi_i^2\frac{L_+^2}{R^2} = 0. \tag{20}$$

Note that for one value of the ξ_i parameter evaluated from (19) we have two values of τ_i from (20). Let us assume that the detrapping rate β is small enough that it fulfils the following relation:

$$\beta \ll \frac{R^2}{4\tau_f\tau_b^2 L_+^2\xi_i^2}(\tau_b - \tau_f - \beta\tau_b\tau_f + \xi_i^2\tau_b L_+^2/R^2)^2 \tag{21}$$

thus the solutions of (20) have the approximated forms:

$$\frac{1}{\tau_{i,1}} = \frac{1}{\tau_f}\left(1 + \xi_i^2\frac{L_+^2}{R^2}\right) + \varepsilon_i \tag{22}$$

and

$$\frac{1}{\tau_{i,2}} = \frac{1}{\tau_b} + \beta - \varepsilon_i \tag{23}$$

where $\varepsilon_i = 2(\xi_i^2\beta\tau_b L_+^2/R^2)/(\tau_b - \tau_f - \beta\tau_f\tau_b + \xi_i^2 L_+^2\tau_b/R^2)$. Note that if we neglect the detrapping the relations (22) and (23) will reduce to the relations which were presented in [3]. The new feature of the obtained result is that the positron lifetime contains an infinite number of lifetime components of which the values are less than the τ_f value (22) and additionally an infinite number of lifetime components of which the values are less than the τ_b value (23).

In comparison to the results obtained in [2] we should mark two important differences: first, in our expression for the mean positron lifetime (17) the value of L_+ is divided by the factor $\sqrt{1 + \tau_b \beta}$, second, we can find a difference in the transcendental equation (19). Both arise from the more complex relation for the $\gamma(s)$ function which we have obtained than that in [2] (relation (5)).

Figure 1 presents an example of the mean positron lifetime as a function of the R/L_+ ratio for a few values of the $\beta\tau_b$ parameter (0, 0.1, 1 and 10). We can see that increase of the value of this parameter causes a faster decrease of the mean positron lifetime towards the τ_f value. This is easily understood, because an increase of the detrapping rate indicates more positrons in the interior of the grain where the positron lifetime is equal to τ_f. In this figure we have drawn for comparison also the results predicted by the authors of [2] for two values of the $\beta\tau_b$ parameter (1 and 10) marked by the dashed lines. We can see that differences between the mean positron lifetime predicted by the two approaches are not large and they disappear when R/L_+ tends to zero.

In conclusion, it may be stated that the detrapping process of positrons from the boundary to the grain modifies in a relevant way the positron annihilation characteristics. When the grain has a spherical shape the detrapping process reduces the diffusion length parameter in the relation for the mean positron lifetime and adds an additional infinite sum of lifetime components in the relation for the positron annihilation spectrum.

Acknowledgment

The author wishes to thank the Committee of Scientific Research (Poland) for the financial support of this work under grant No 2P03B 027 10.

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