Some remarks on positron/positronium diffusion models

Wacław Świątkowski

Abstract Positrons thermalized near the sample boundary (as well as Ps atoms formed in the sample) can reach the boundary as a result of diffusive movement. Observations of such effects as positron reemission or positron surface trapping as well as positronium emission allow one to calculate respective diffusion lengths. From the presented analysis it follows that some models used in such calculations give quite wrong results.

Key words diffusion • diffusion length • positron • positronium

W. Świątkowski
Institute of Experimental Physics,
University of Wrocław,
9 M. Borna Sq., 50-204 Wrocław, Poland,
Tel.: +48 71/ 375 93 12, Fax: +48 71/ 328 73 65,
e-mail: swat@ifd.uni.wroc.pl

Introduction

Positron is a very useful probe for investigation of bulk solids and surfaces. The positron is the lightest positive particle and, moreover, it is an antimatter particle in our ordinary matter world, therefore it is free from exchange forces. Positron injected into a solid losses its initial kinetic energy and reaches the thermal energy in a relatively short time (as compare with its common lifetime). After the thermalization stage the further displacement of positron has a diffusive character. Although the positron is a stable particle it does not move diffusively very long because the annihilation and the capture in a localized state can break its migration. That is why both the diffusion constant D_+ and the mean time of diffusive movement τ_{diff} are an important characteristics of the diffusion stage of positron life. With this two parameters one can define a new parameter, positron diffusion length $[L_+ = (D_+\tau_{\text{diff}})^{1/2}]$ characterizing the distance that thermalized positron can travel before annihilation or capturing.

In nonmetals diffusive migration of positron can be preceded by forming a positronium atom (Ps – the positron–electron bound state). In such a case one should consider the positronium diffusion constant D_{Ps} and the positronium diffusion length L_{Ps}. Positron (Ps atom) diffusion length can be determined from the experiments where the effects connected with the possibility of reaching by the positron (Ps atom) of a surface or an interface are observed. Here we present formulas based on the respective diffusion equations, and related to the problem, for two different geometrical situations. These formulas are further compared with those obtained on the basis of two models being not good approximation of the problem.

Escape coefficient for a thin layer sample

Let us consider a thin layer, of thickness l, in which positrons are uniformly implanted as thermalized with the intensity of implantation S. It means that the equilibrium concentration of positrons c can be treated as dependent only on one coordinate, say x, being perpendicular to layer boundaries and that c fulfils the equation

$$(1) \qquad \frac{\mathrm{d}^2 c(x)}{\mathrm{d}x^2} - \frac{c(x)}{L_+^2} + \frac{S}{D_+} = 0$$

where: L_+ is the positron diffusion length; and D_+ the respective diffusion constant. If one assumes that the possibility of leaving the sample through the boundary (or of trapping at the boundary) is determined by the relation (see e.g. [9])

$$(2) \qquad J_{\mathrm{Esc}}\left(x_0\right) = \alpha \cdot c\left(x_0\right) = \pm D_+ \left. \frac{\mathrm{d}c(x)}{\mathrm{d}x}\right|_{x=x_0}$$

where: $J_{\mathrm{Esc}}(x_0)$ is the density of escaping positron current and $c(x_0)$ the equilibrium positron concentration at the boundary (situated at $x = x_0$). The sign in front of D_+ should be the same as that of the derivative $\mathrm{d}c(x)/\mathrm{d}x$. In the considered case x_0 has two values, say $x_{\min}$ and $x_{\max} = x_{\min} + l$, and the relation $J_{\mathrm{Esc}}(x_{\min}) = J_{\mathrm{Esc}}(x_{\max}) \equiv J_{\mathrm{Esc}}$ is obvious. With taking this into account, a solution of Eq. (1) under the condition (2) gives for J_{Esc} the formula

$$(3) \quad J_{\mathrm{Esc}} = SL_+ \frac{e^{l/2L_+} - e^{-l/2L_+}}{e^{l/2L_+} + e^{-l/2L_+} + \frac{D_+}{\alpha L_+}\left(e^{l/2L_+} - e^{-l/2L_+}\right)}.$$

Using this formula one can obtain the following formula for the probability K that a positron implanted in a layer leaves the layer interior (escapes from it being surfacely trapped or/and reemitted) [2]:

$$(4) \qquad K = \frac{n_{\mathrm{Esc}}}{n_{\mathrm{Impl}}} = \frac{2J_{\mathrm{Esc}}}{Sl}$$

$$= \frac{2L_+}{l} \frac{e^{l/2L_+} - e^{-l/2L_+}}{e^{l/2L_+} + e^{-l/2L_+} + \frac{D_+}{\alpha L_+}\left(e^{l/2L_+} - e^{-l/2L_+}\right)}$$

where: n_{Impl} denotes the number of positrons implanted in the layer; n_{Esc} stands for the number of the implanted positrons which escape from the layer interior (through both layer surfaces); and l is the layer thickness. Further, the quantity K will be called the escape coefficient.

Commonly it is assumed that for perfectly not reflecting boundary one can take $\alpha = \infty$. A validity of such approximation was discussed by Dębowska and Świątkowski [7]). We will follow this assumption.

For $\alpha = \infty$ the formula (4) takes the form [11]:

$$(5) \qquad K = \frac{2L_+}{l}\tanh\left(\frac{l}{2L_+}\right).$$

More general treatment of the considered here problem (for different geometries), which leads as well to the formula (5), one can find in the paper of Brandt and Paulin [6].

Probability of escaping (e.g. reemission) of positrons through a flat boundary of a thick sample

Let us now consider the possibility of leaving of a thick uniform sample through its flat boundary by positrons being firstly implanted in the sample. Taking in Eq. (1) S as being different from zero only for the distance from the boundary equal to z, one can (with condition (2) and $\alpha = +\infty$) obtain the probability $p(z)$ of remission of positrons as equal

$$(6) \qquad p(z) = \exp(-z/L_+).$$

If $P(z)$ denotes the implantation profile, then the global probability of reemission (the escape coefficient) is given by the commonly used formula (see, e.g. [8]):

$$(7) \qquad K = \int\limits_0^{+\infty} P(z)p(z)\mathrm{d}z$$

with $p(z)$ given by Eq. (6).

The formulas (4)–(7) may be used not only for positrons, but as well for other objects with limited diffusion time (e.g. for Ps atoms) implanted/created and then diffusing in the sample (for Ps atoms parameters D_+ and L_+ should be replaced with D_{Ps} and L_{Ps}).

As it was shown, for geometrically simple situations one can easily obtain respective escape coefficients (K) without using any "simplified" models. Unfortunately, in some cases such models were used leading to very wrong results. Two of them will be discussed below.

The geometrical model used by SERNA

In a paper by Serna [10], the so called geometrical model was proposed to obtain the formula giving the escape coefficient for Ps atoms (related there to the transition of the atoms from crystalline to amorphous phase), created uniformly in a flat layer of thickness l and characterized by the diffusion length L_{Ps}. According to the model, a Ps atom starting its diffusive movement at a given point moves up to reaching the distance L_{Ps} from the starting point. If in a choosen direction the sample ends at the distance less than L_{Ps} from the point, then the Ps atom escapes from the layer. It means that in the model it is assumed that the diffusive movement of the Ps atom is rather close to the α-particle movement than to the real diffusive movement. Using this model, Serna obtained the formula

$$(8) \qquad K = \begin{cases} 1 - \dfrac{l}{2L_{\mathrm{Ps}}} & \text{for } l < L_{\mathrm{Ps}} \\[2ex] \dfrac{L_{\mathrm{Ps}}}{2l} & \text{for } l \geq L_{\mathrm{Ps}} \end{cases}$$

instead of Eq. (5). Some results obtained with formula (5) and formula (8) are presented in Fig. 1 for comparison. As
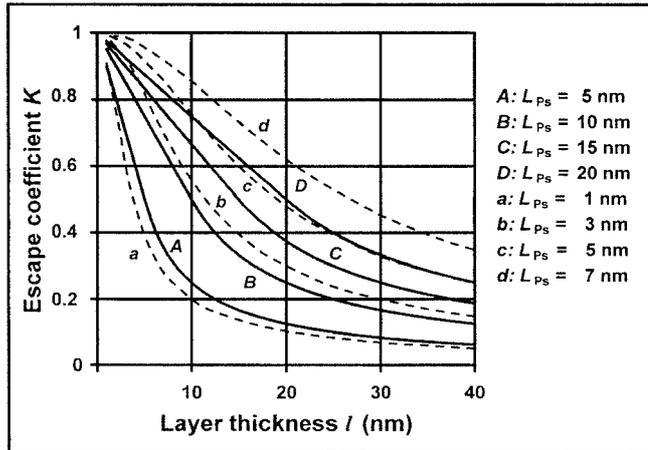
Fig. 1. The dependence of the escape coefficient K on the layer thickness obtained with formula (8) used by Serna (smooth lines) and with formula (5) (dashed lines). The smooth lines *A, B, C* and *D* correspond to L_{Ps} equal 5, 10, 15 and 20 nm, respectively; the dashed lines *a, b, c* and *d* are related to L_{Ps} equal to 1, 3, 5 and 7 nm, respectively.

Fig. 2. The dependence of the escape coefficient K on the Ps diffusion length as calculated using the accurate formula (curve 1), the spherical segment model (curve 2), and Serna's model (curve 3).

it is seen there is a great discrepancy between the results obtained using Serna's model (formula (8)) and formula (5). According to Serna, one series of his experimental points could be well fited by the line corresponding to L_{Ps} = 10 nm. As it is seen in Fig. 1, a similar shape to Serna's curve for L_{Ps} = 10 nm could have the curve obtained with formula (5) and corresponding to L_{Ps} being somewhat less than 3 nm. Thus the model used by Serna and his estimation of L_{Ps} value cannot, anyway, be treated as some approximation of reality; instead of L_{Ps} close to 3 nm the L_{Ps} obtained by him is close to 10 nm.

The model of spherical segment

In a paper by Anwand *et al.* [1], for the calculation of the fraction of Ps atoms leaving a sample another model, called by the authors "*the model of spherical segment*", was used. According to the model, a Ps atom created at a given point can reach due to its diffusive movement any place in the sphere of radius L_{Ps} (the point is a center of this sphere) with the same probability. According to these authors "*the Ps gets chance to leave the sample in the case when its diffusion length is longer than its implantation depth*". It means that they proposed to calculate the escape coefficient using formula (7) with $p(z)$ not given by Eq. (6) but equal to

$$(9) \qquad p(z) = \begin{cases} \dfrac{(L_{Ps}-z)^2 \cdot (2L_{Ps}+z)}{4L_{Ps}^3} & \text{for } z \leq L_{Ps} \\ 0 & \text{for } z > L_{Ps} \end{cases}.$$

In addition, in calculations done in paper [1] the Makhov implantation profile was used as $P(z)$. Here, taking from [1] the parameters related to synthetic alpha-quartz A, the dependence of the escape coefficient on diffusion length was calculated both with formula (6) and formula (9). The results are shown in Fig. 2. Additionaly, in Fig. 2 a similar result obtained with the use of Serna's model is shown.

As it is seen, there is a great discrepancy between the results obtained with formula (9) and with formula (6). The
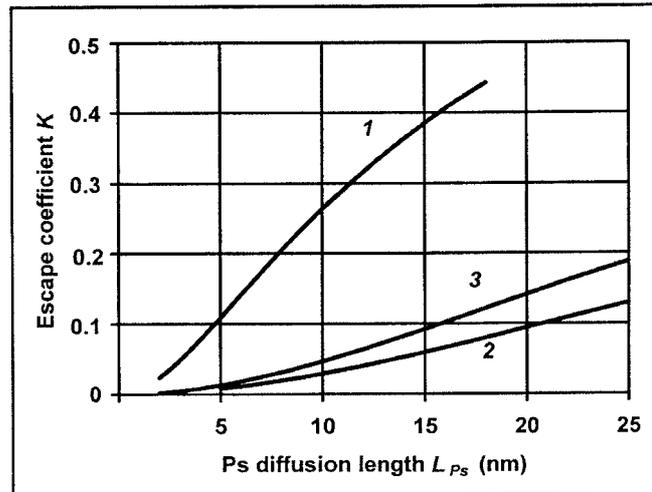
experimental escape probability for Ps atoms created in synthetic alpha-quartz A estimated in the paper [1], related to the implantation of monoenergetic positrons of 1.03 keV, was close to 0.1.

Thus it follows from Fig. 2 that according to the spherical segment model $L_{Ps} \cong 20$ nm, whereas accurate calculations give $L_{Ps} \cong 5$ nm; Serna's model gives for this case $L_{Ps} \cong 15$ nm. It should be mentioned that in paper [1] L_{Ps} for alpha-quartz A was found as being equal to 18 nm. The small difference between this value (18 nm) and that obtained in this work 20 nm may be a result of using in Makhov formula different parameters. Nevertheless, it is quite clear that the spherical segment model (as well as Serna's model) is very wrong.

Some remarks related to the interpretation of diffusion length

The models discussed above indicate that even experienced authors do not well interpret the diffusion length. The physical quantity, called the diffusion length, is in common use not only in positron problems but also in neutron and semiconductor physics. Especially many problems, related to neutron diffusion, for different geometries (see, e.g. [4]), were solved. In each case the diffusion length L (see Eq. (1)) is defined by the relation

$$(10) \qquad L = \sqrt{D \cdot \tau_{diff}}$$

where D is the diffusion constant and τ_{diff} denotes the mean time of the diffusion. Unfortunately, in positron physics this formula is not commonly acceptable. For example, in papers by Brandt [5] and Bergersen *et al.* [3] instead of formula (10) another relationship,

$$(11) \qquad L = \sqrt{2D \cdot \tau_{diff}}$$

is used. Sometimes you can find the term "mean diffusion length" as equivalent of "diffusion length". It seems thus that it would be suitable to remember certain relations

connected with diffusion length as they are treated in neutron physics.

Solving of diffusion equation

$$(12) \qquad \nabla^2 c(x,y,z) - \frac{c(x,y,z)}{L^2} + s(x,y,z) = 0$$

(Eq. (1) is its special variant) for a point source situated in an infinite uniform material at the center of coordinate ($x = y = z = 0$) one can obtain for the equilibrium neutron/positron concentration c the formula

$$(13) \qquad c(r) \approx \frac{1}{L^2} \frac{e^{-r/L}}{r}.$$

Using this formula it is easy to come to the following conclusions:

1. The mean distance $\bar{r}$ from the source to the place where the diffusive movement of the particle ends is

$$(14) \qquad \bar{r} = 2L.$$

2. The mean value $\bar{r}^2$ of the square of the above distance is

$$(15) \qquad \bar{r}^2 = 6L^2.$$

3. About 26% of positrons end their diffusive movement in the sphere of radius L with the center in the source; for the sphere of radius $2L$ the corresponding amount is 59%.

Final remarks

As it is seen from formula (14), the mean distance from the place where the diffusive movement begins to that where it ends is equal to two diffusion lengths. If one calculates such value basing on the above considered models, the result is $\bar{r} = L$ according to Serna's model and $\bar{r} = (3/4)L$ within the spherical segment model. From each of the two models it follows that all the Ps atoms diffusing in an infinite sample, end their diffusion motion in the sphere (centered at the source) of radius L; as it was pointed out earlier, the accurate prediction is only 26%. At the end, let us turn to the probability $p(z)$. Considering the upper limit of $p(z)$, we have for $z = 0$. The accurate value of $p(0)$ related to formula (6) equals 1, whereas both models gives $p(0) = \frac{1}{2}$. Thus, the discussed models should be treated as very wrong in all aspects.

It is not possible to know how many papers give wrong estimations of diffusion lengths. In many cases it is not precised if we have accurate calculations or some "simplified procedure". Especially in some papers where diffusion length is estimated you can meet Anwand and Brauer (coauthors of paper [1]) as coauthors of the papers. Sometimes it is not clear which procedures were used in these cases.

Thus, one can be very careful in using diffusion length values taken from the papers where the procedures were not enough precisely described.

References

1. Anwand W, Brauer G, Hasegawa M, Dersch O, Rauch F (2001) A study of positron properties in quartz crystals and synthetic silica glass. Acta Phys Pol A 99:321–328
2. Baranowski A, Ewertowski R, Świątkowski W (1991) Positron trapping rate coefficient for thermalized positrons at metal-dielectric interface. Acta Phys Pol A 79:517–523
3. Bergersen B, Pajanne E, Kubica F, Stott MJ, Hodges CH (1974) Positron diffusion in metals. Solid State Commun 15:1377–1380
4. Berkurts KH, Wirtz K (1964) Neutron physics, 2nd ed. Springer Verlag, Berlin–Göttingen–Heidelberg–New York
5. Brandt W (1974) Positron dynamics in solids. Appl Phys 5:1–23
6. Brandt W, Paulin R (1972) Positron diffusion in solids. Phys Rev B 5:2430–2435
7. Dębowska M, Świątkowski W (1999) Some remarks on diffusion model of positron reemission and/or positron surface trapping. Acta Phys Pol A 95:515–520
8. Dupasquier A, Zecca A (1985) Atomic and physics experiments with slow-positron beams. Riv Nuovo Cimento 8:1–73
9. Schultz PJ, Lynn KG (1988) Interaction of positron beams with surfaces, thin films, and interfaces. Rev Mod Phys 60:701–779
10. Serna J (1990) Positronium diffusion in crystalline polyethylene. Phys Status Solidi A 122:489–493
11. Świątkowski W (1974) The possibility of using the metal–metal contact effects to investigate the slow positron diffusion. Nukleonika 19:79–83