

Distribution of Positron Annihilation Lifetime Components

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Abstract

The intensities of the positron annihilation lifetime components are shown to be related to the free volume model, and the theoretical value of the ratio between the singlet to the triplet state intensities, described by the ratio η , is shown to follow an empirical model. An application of the model on some experimental data shows that the calculated value of the ratio η approaches the theoretical value, which is 1/3.

Furthermore, the relation between the free volume hole fraction, F_h , with the free volume V_h described by the constant C is related to τ_3 . Therefore, the deviation of the constant C and the ratio η that are reported and differ from the theoretical values for similar researches could be corrected. When data are collected by means of positron annihilation lifetime spectroscopy (PALS), the spread-intensity model is suggested to explain this correction.

Keywords: positron annihilation in solids, annihilation intensity.

Introduction

The physical properties of the positron are identical to those of the electron except the sign of its charge, for the positron it is a unit positive charge, $+e$. If the electron meets the positron, they annihilate emitting usually two photons after a certain lifetime, which is called the positron lifetime. The electron - positron pair may also form a bounded system, the Positronium atom (Ps), before annihilation event. The Ps atom may exist in either the para-state (denoted by *p-Ps*), or the ortho-state (*o-Ps*). The lifetime of the p-Ps in vacuum is 125 picoseconds (ps), and that of the o-Ps is 140 nanoseconds (ns). In matter, the existence of many electrons around the Ps atom changes its lifetime significantly, specially in the o-Ps system where the lifetime is reduced from 140 ns to 2 - 5 ns. Any process that is responsible for such effect is called *Quenching Process*.

There are many types of quenchers such as [1]:

1- pick-off: a process that includes the annihilation of the Ps positron with an external electron.

2- spin conversion: the collision of the Ps atom with a paramagnetic atom or molecule of matter may convert the p-Ps (o-Ps) to o-Ps (p-Ps).

3- ionization: the Ps atoms binding energy is $I_{Ps} = -6.802$ eV. If the Ps atom goes through some energetic reaction where it receives an amount of energy above I_{Ps} it may be ionized returning into a free electron and positron.

4- oxidation: a second electron may join the Ps system and becomes bounded to it, performing electron-positron-electron (or Ps) bounded system.

5- Ps-molecule compound formation: an intermediate reaction that could end by any of the earlier reactions.

The formation of the Ps atom is a process that usually occurs in molecular media. In heavier materials, such as metals, the Ps formation is not possible

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due to the high electron density. Any molecular material contains at least one type of quenchers.

The correlation between the positron system's lifetime and the properties of the material provides important information about the internal structure of that material. Such a correlation is usually studied by means of *Positron Annihilation Lifetime Spectroscopy* (PALS).

The i^{th} positron system is characterized by a lifetime, τ_i , and a corresponding intensity, I_i . It is customary to refer to the p-Ps by $i=1$, to the free positron by $i=2$, and for the o-Ps by $i=3$.

When the Ps atom is formed in some material, there should be a relation between the o-Ps and p-Ps intensities. The statistical weight of the triplet state is three times larger than single state, i.e., \bar{I}_1/\bar{I}_3 equals to 1/3. Let us denote this ratio by $\bar{\eta}$, which is the ratio between the theoretical ratios of p-Ps to the o-Ps intensities. In molecular materials, there exist at least three positron systems; those are: p-Ps ($\tau_1 \approx 100$ -200 ps), the free positron ($\tau_2 \approx 300$ -500 ps), and the o-Ps ($\tau_3 \approx 2000$ -5000 ps).

The measurement of the o-Ps pick-off lifetime, τ_3 , can be used to calculate the free volume size, V_h , of the unit cell of the material [2], by using the following semi-empirical formula [3]:

$$\tau_3 = 0.5 \left[1 - \frac{R_h}{R_h + dR} + \frac{1}{2\pi} \sin\left(\frac{2\pi R_h}{R_h + dR}\right) \right]^{-1} \quad (1),$$

where R_h is the free volume hole radius and dR is the electron layer thickness, its value was found to be 0.1665 nanometer (nm) for solid and 0.1830 nm for liquid molecular materials [3]. Formula (1) is generally accepted to represent the free volume radius [4,5,6]. V_h is then given as $V_h = (4\pi R_h^3)/3$ in a spherical geometry approximation. Another quantity can be measured from τ_3 and \bar{I}_3 , that is the free volume hole fraction, F_h , given as:

$$F_h = C \bar{I}_3 V_h \quad (2)$$

where C is an empirical constant of a value between 1 and 2 nm^{-3} , when V_h is given in nm^3 and \bar{I}_3 in percentage.

Although the value of the constant C is thought to be strongly dependent on the type of the investigated material, many researches reported different values for similar types of the investigated materials under identical conditions. In addition, there are many confusing measurements of the ratio η , where its value was found, in general, $\bar{\eta} \neq 1/3$.

A general remark can be deduced is that, if the mathematical representation and the computational method used for analyzing the annihilation lifetime spectra are accurate, the obtained information *should* be the same if the same material was studied under similar conditions, within an accepted range of error.

A possible reason of the deviation of the results may be due to the nature of the studied samples or the technical effects. When PALS is used to study polymers, the deviations of the studied material's properties can add some effects. Impurities and defects may change the positron lifetime. In pure materials, such as metals and alloys, there is a universal matching of the measured lifetimes in such a way that the positron's lifetime in metals can be assumed as a reference lifetime for calibrating experimental apparatus [7, 8].

For molecular materials, the differences in the lifetime measurements are so wide that they may not be due to the samples properties only. To explain this let us take an example.

Yu et al. [5] have studied phase transition of the polystyrene polymer (PS) of different molecular weights, ($M_w = 40 \times 10^3$, 9.2×10^3 , 25×10^3 and 400×10^3) under temperature range from 10°C to 130°C; and Liu et al. [6] also studied PS polymer of $M_w = 105 \times 10^3$, under the temperature range from 25°C to 130°C.

Furthermore, both groups used PATFIT program for analyzing the lifetime spectra into three discrete lifetime components, and the characteristics of the experimental system for both groups were almost identical, where the full width at half maximum of the time resolution was 260 ps for both experiments [5, 6].

For comparison, we take from Yu et al. [5] the results of the polymer samples of $M_w = 25 \times 10^3$ and those of $M_w = 400 \times 10^3$, where the selection is chosen so that the results of Liu et al. [6] can be compared to them approximately. In Table 1, these results are listed for the

temperature range from 110°C to 130°C, where this range is chosen in order to ignore the structural and thermal history effects on the lifetime and intensity measurements. In between the results of Yu et al., one expects that there is an acceptable matching with Liu et al., within the reported experimental error given in both groups. However, the results of the lifetime measurements showed only behavior matching, whereas intensity measurements showed neither numerical nor behavior matching (*see* Table 1).

Table 1. The results of references [5] and [6] for polystyrene polymer (PS) in the temperature range 110°C to 130°C. In both references, τ_1 was fixed to 120 ps.

M_w $\times 10^3$	τ_2 (ps)	τ_3 (ps)	\bar{I}_1 (%)	\bar{I}_2 (%)	\bar{I}_3 (%)	Temperature °C
Yu et al. [5]						
25	Not reported	2500	Not reported	Not reported	37.5	110
25	Not reported	2600	Not reported	Not reported	37.0	130
400	374±4	2342±8	26.22±0.49	37.6:1:0.42	26.12±0.16	110
400	368±4	2482±8	26.26±0.49	36.9:1:0.42	36.82±0.16	130
Liu et al. [6]						
105	398±6	2080±9	24.31±0.44	34.79±0.36	40.90±0.24	110
105	396±6	2106±9	24.36±0.43	34.36±0.36	40.70±0.25	130

From the results of Yu et al. [4], one observes that the lifetime of the o-Ps annihilation increases as the molecular weight decreases. The behavior of τ_3 is in accordance to the free volume model [9]. However, this does not show reasonable matching to the results of Liu et al. because the molecular weight of their samples was $25 \times 10^3 < 105 \times 10^3 < 400 \times 10^3$; therefore, one expects that the value of τ_3 reported by, Liu et al. should be $2500 < \tau_3 < 2600$ ps, whereas it was 2100 ps. This mismatching could be either due to:

- 1- The differences of the examined samples properties.
- 2- The differences of the technical methods.

The first reason can change the

numerical value of the measured lifetimes and intensities due to the different reactions and formation probabilities, while the second one may not affect the lifetime sufficiently especially the o-Ps lifetime due to its long lifetime. This reason may highly affect the intensities of the positron systems, because the intensities measurement depends on the features of the computational method used for analyzing the lifetime spectra, such as its stability and the number of required iteration for the error to converge.

This can result in high deviation of the measured values of intensities for similar materials and causes deviation of the ratio $\bar{\eta}$ from 1/3. Mogensen et al. [10-12]

studied 36 pure liquids, and found that there are many reasons affecting the positron systems, which may cause such measurements, such as [10,11]:

1- methods of data analysis that may fail in subtracting the intensities correctly especially the p-Ps intensity (\bar{I}_1) due to its short lifetime.

2- the o-Ps - to - p-Ps interference, which may be due to linear combination of both systems intensities in the observed annihilation spectrum.

3- greater probability of o-Ps pick-off annihilation comparing to that of the p-Ps.

4- greater probability of o-Ps ionization comparing to that of the p-Ps.

5- greater probability of o-Ps oxidation comparing to that of the p-Ps.

6- spin conversion of the Ps atom at the terminal spur might be significant.

7- possible interactions of the Ps with the positive ions in the terminal spur.

The participation of the reasons 3 to 7 thought not to affect the measured intensities as sufficient as the reasons 1 and 2 [10, 11], because of the small probability of the reactions leading to reasons 3 - 7.

In the present paper, a suggested explanation for the deviation of the ratio $\bar{\eta} = \bar{I}_1 / \bar{I}_3$ from 1/3 is given. In addition, it is shown that the deviation of the value of the constant C (equation 2) can be explained accordingly.

The Spread-Intensity Model

The contradiction of the measurements of the ratio $\bar{\eta}$ can be explained on the base of the reasons 1 and 2 of Mogensen et al. given above. This is achieved by further assuming that:

1- There are only three positron systems, those are of the p-Ps (lifetime, τ_1 , intensity \bar{I}_1), the free positrons (τ_2, \bar{I}_2) and the o-Ps (τ_3, \bar{I}_3).

2- The pure p-Ps (unmixed) intensity is

assumed to be slightly affected by quenching processes.

3- The free positrons intensity is less spread over the shorter lifetime component (of the p-Ps).

4- The o-Ps intensity is spread over the whole spectrum.

5- The p-Ps intensity is not spread at all upon any other lifetime component.

Then one can formalize the relation between the measured intensities, \bar{I}_i in relation to the actual ones I_i , where $i=1, 2, 3$; as:

$$\bar{I}_1 = I_1 + a_{1,2} I_2 + a_{1,3} I_3 \quad (3-a)$$

$$\bar{I}_2 = a_{2,2} I_2 + a_{2,3} I_3 \quad (3-b)$$

$$\bar{I}_3 = a_{3,3} I_3 \quad (3-c)$$

or in the general form,

$$\bar{I}_i = \sum_{j=1}^3 a_{i,j} I_j \quad (4)$$

with: $a_{1,1} = 1$, and: $a_{1,2} = a_{3,1} = a_{3,2} = 0$; where \bar{I}_i is the i^{th} measured intensity, and I_i is the i^{th} real intensity; and $a_{i,j}$ are numerical coefficients, normalized in such a way that:

$$\left. \begin{aligned} a_{1,2} + a_{2,2} &= 1 \\ a_{1,3} + a_{2,3} + a_{3,3} &= 1 \end{aligned} \right\} \quad (5-a)$$

and,

$$\sum_{j=1}^3 I_j = \sum_{i=1}^3 \bar{I}_i \quad (5-b)$$

Formulae (3-a to 3-c) means that the observed intensities, \bar{I}_i represent a mixture of the actually occurring intensities, I_i .

These formulae are written in this form in order to explain the possible interfering of the o-Ps -to- the free positron -to- the p-Ps systems. Such a phenomenon could cause the failure of the accurate separation of the real intensities.

Furthermore, let us define,

$$\eta = I_1 / I_3 \quad (6)$$

this is the relation between the *real* intensities I_1 and I_3 , then, the following formulae could be deduced from formulae (3), (4), (5), and (6):

$$\begin{aligned} \bar{I}_1 &= a_{1,2} + \frac{\bar{I}_3}{a_{3,3}}(a_{2,2} \eta + a_{1,3} - a_{1,2}) \\ &= a_{1,2} + \frac{\bar{I}_3}{a_{3,3}}(\eta + a_{1,3} - a_{1,2}(1 + \eta)) \end{aligned} \quad (7)$$

similarly

$$\bar{I}_2 = a_{2,2} - \frac{\bar{I}_3}{a_{3,3}}(a_{2,2}(\eta + 1) - a_{2,3}) \quad (8)$$

which represents the relation between two of the measured intensities with the coefficients $a_{i,j}$ as unknowns. These formulae do not depend on the real intensities.

Using of the product ($\tau_i \bar{I}_i$) is another possible way to have similar equations to formulae (3-a to 3-c), where if one assumes that:

$$\tau_1 \bar{I}_1 = \tau_1 I_1 + b_{1,2} \tau_2 I_2 + b_{1,3} \tau_3 I_3 \quad (9-a)$$

$$\tau_2 \bar{I}_2 = b_{2,2} \tau_2 I_2 + b_{2,3} \tau_3 I_3 \quad (9-b)$$

$$\bar{I}_3 = b_{3,3} I_3 \quad (9-c)$$

which by defining the average lifetime, τ_{ave} , as

$$\tau_{ave} = \frac{\sum_{i=1}^3 \tau_i \bar{I}_i}{\sum_{i=1}^3 \bar{I}_i} = \sum_{i=1}^3 \tau_i \bar{I}_i \quad (10)$$

and using a similar normalization condition to formula (5-a) for $b_{i,j}$ and (5-b), then the solution will be :

$$\tau_1 \bar{I}_1 = b_{1,2} \tau_2 + \frac{\tau_3 \bar{I}_3}{b_{3,3}} \left(\eta \frac{\tau_1}{\tau_3} - b_{1,2} \frac{\tau_2}{\tau_3} (\eta + 1) + b_{1,3} \right) \quad (11)$$

$$\begin{aligned} \tau_2 \bar{I}_2 &= \tau_{ave} - b_{1,2} \tau_2 - \frac{\tau_3 \bar{I}_3}{b_{3,3}} \times \\ &\left(\eta \frac{\tau_1}{\tau_3} - b_{1,2} \frac{\tau_2}{\tau_3} (\eta + 1) - b_{2,3} + 1 \right) \end{aligned} \quad (12)$$

Applications

The usefulness of this simple correction model (formulae (7) and (8), or (11) and (12)) is in the numerical coefficients value. For now, let us discuss formulae (7) and (8) only. Formulae (11) and (12) will be left for future investigations.

By comparing the experimental data of the measured intensities with that of formulae (7) and (8), one can notice that the contribution of the of the i^{th} real intensity to the measured one is higher than its contribution in the j^{th} measured intensity, where $i \neq j$, i.e., in general, $a_{1,2} < a_{2,2}$; $a_{1,3} < a_{3,3}$ and $a_{1,3} < a_{2,3}$.

The ratio η is kept as unknown in order to include any possibility that its real value deviate from 1/3. However, the exact values of the entire numerical coefficient cannot be found using formulae (7) and (8) directly. Instead, the values of $a_{1,2}$ and $a_{2,2}$ can be found by, for example, fitting formulae (7) or (8), and using the identities (5-a and 5-b). Then the value of η can be found from the equation:

$$\eta = \frac{a_{3,3} \times slope + a_{1,2} - a_{1,3}}{1 - a_{1,2}}$$

where the fit was assumed as, from equation (7),

$$\bar{I}_1 = a_{1,2} + (slope) \times \bar{I}_3.$$

For example, we take the last five points from Yu et al. [5] and fit them to the formulae (7). Let's assume initially that $a_{1,3} \approx 0$, and that the intensity \bar{I}_3 is corrected as in equation (3-a). Furthermore, let equation (2) be given as:

$$C \bar{I}_3 = k \frac{\bar{I}_3}{a_{3,3}}, \text{ i.e., } C = \frac{k}{a_{3,3}} \quad (13)$$

where k is a proportionality constant. Assuming that $k = 1 \text{ nm}^{-3}$ (same units of C) then $C=1/a_{3,3}$, and one can find that the slope = -0.13604, then: $a_{1,2}= 0.3129$, $a_{2,2} = 0.6871$, and one gets $\eta \approx 0.3368 = 1/2.9688$, where $a_{3,3}$ was taken as 1/1.67 [5].

Similarly we take the last two points

of Liu et al. (for M_w values 400×10^3 only) [6], and treating them as before, then the slope = -0.2500, leading to: $a_{1,2} = 0.3453$, $a_{2,2} = 0.6542$, and one gets $\eta \approx 0.2547 = 1/3.926$, where $a_{3,3} = 1/1.400$ [6]. The fitted data were chosen for the temperature range $T=110 - 130^\circ\text{C}$.

The deviation of η from $1/3$ can be more improved if one can calculate the value of the coefficient $a_{1,3}$. For if $a_{1,3} = 0.002$, then $\eta \approx 1/3.07$ (data from [5]) and if $a_{1,3} = -0.05$ then $\eta \approx 1/3.02$ (data from [6]). Thus we see that, though the assumption of $a_{1,3} \approx 0$ is acceptable, its value may highly change the results. This coefficient is playing the rule of determining the illness condition of the present set of equations, at least for the given values from the two examples taken.

Comparing the values of η with the uncorrected values reported in references [5, 6], one finds that the fitted η values are well accepted, (see Table 2).

Table 2. Comparison between experimental and calculated values of η . Calculations were made according to the Spread - Intensity model

	the reported experimental value of η	the value of η calculated according to the SI model	the value of η calculated according to the SI model corrected for $a_{1,3}$
reference [5]	1/3.8330	1/2.9688	1/3.070
reference [6]	1/1.6765	1/3.9260	1/3.021

Discussions

The results of η (of the corrected $a_{1,3}$ and uncorrected) strongly indicate the validity of equation (13). This equation means that equation (2) can be re-written as:

$$F_h = \frac{k I_3 V_h}{a_{3,3}} \quad (14)$$

which means that F_h is proportional to the real o-Ps intensity I_3 . This does not contradict the theory at all [6], and still give the same meaning of the match between the relation of F_h and V_h . Therefore the empirical formula (13)

is still convenient.

The idea of the spread-intensity model is that the probable interfering of the real intensities with each other will appear as a completely new set of intensities. The mathematical representation of PALS spectrum and the method treating it in order to extract lifetime parameters may share a great deal to such effect.

Most of the programs used to analyze PALS spectra depend in one way or in another on iteration procedure. It is well known [13,14] that, in such programs, the results may show change in the output results according to the initial input values. However, this may not affect the results of the measured annihilation lifetime considerably, especially the o-Ps lifetime because of its long lifetime, and the nature of the mathematical form used for describing the annihilation spectrum.

The values of the measured intensities, on the other hand, may suffer from some error due to the same reason, especially due to the presence of the short lifetime component. Thus, instead of measuring the real intensities, a combination of them appears in the results of numeric calculations because spectrum analysis may not distinguish such interfering.

An important difference between this model and the model of Brandt and Spirn [15] is that, the intensity of the second component, I_2 in our model was not assumed to be constant. This seems convenient since that component was thought to vary under certain conditions, such as irradiating a polymer in air and using PALS as a method of study [16]. It should be noted that the value of η does not have value corresponding to theory, in a medium consisting of a paramagnetic atoms. It is known that, in such media, spin conversion of o-Ps to p-Ps becomes significant, so in some cases when $\eta > 1/3$, it may still in accordance to theory.

The small value of the coefficient $a_{1,3}$ indicates that o-Ps component is not contributing significantly in the shortest

lifetime component. Without being wrong, the value of the coefficient $a_{2,3}$ can be given as $a_{2,3} \approx 1 - a_{3,3}$, then $0 \leq a_{2,3} \leq 0.5$ (because $0.5 \leq a_{1,3} \leq 1$), where $1 \leq C \leq 2$.

Conclusions

In conclusion, the technical causes of error of the intensity measurements can be eliminated using simple set of equations, where the contradiction of the ratio η and the value of the constant C can be explained for similar type of polymeric materials of similar properties and similar experimental conditions.

This is achieved using the spread-intensity model. Application of this model on the measured intensity values of PS polymer showed well-accepted results that explain the deviation of the ratio I_1/I_3 from 1/3.

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توزيع مركبات الطيف الزمني لفناء البوزترون

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الخلاصة

لقد تم توضيح العلاقة بين مركبات طيف فناء البوزترون مع نموذج الحجم الحر، وحسبت النسبة النظرية بين شدة الفناء من الحالة الأحادية (Singlet para state) والحالة الثلاثية (Triplet ortho state) والموصوفة بالمعامل η ومقارنتها مع النسبة المحسوبة من النتائج عمليا، والتي تتبع معادلة وضعية. وعند تطبيق هذه المعادلة على نتائج عملية وجد أن النسبة المحسوبة من النتائج العملية تقترب من النسبة النظرية، والتي هي $1/3$. بالإضافة إلى ذلك، فقد تم الربط بين نسبة الحجم الحر، F_h ، والحجم الحر، V_h ، المحسوبة من عمر فناء المركبة الطويلة τ_3 مع الثابت C . وعلى أساس هذا الربط فسرت الأسباب وراء تغير قيم الثابت C وتفاوت قيم المعامل η بين القياسات المختلفة وربط ذلك مع النسبة النظرية للمعامل η . تم اقتراح نموذج انتشار الشدة لتفسير النتائج عند استخدام مطيافية فناء البوزترون لتفسير هذه التصحيحات.