Source correction in positron annihilation lifetime spectroscopy

N Djourelov and M Misheva

Faculty of Physics, Sofia University, 5 J Bourchier Boulevard, Sofia 1126, Bulgaria

Abstract. The positron lifetimes in a ^{22}NaCl source have been measured. The fraction of positrons annihilated in ^{22}NaCl salt and in the Kapton supporting foils has been determined as a function of the atomic number Z of the sample. All of the experimental results have been compared with calculated values obtained from a computer program based on a Monte Carlo algorithm. An algorithm has been proposed for the determination of the source correction in lifetime spectra in positron annihilation lifetime spectroscopy.

1. Introduction

In the majority of positron annihilation measurements a 'source' consisting of the radioactive material (very often dried ^{22}NaCl solution) packed between two thin foils is used. The accurate determination of the lifetimes and fractions of the positrons annihilating in the radioactive material and its supporting foils is of great importance for correct interpretation of the results from positron lifetime measurements. For this reason there are many publications on this subject, e.g. [1–4].

As the same source is usually used for studying different samples over a long period of time it is worthwhile to examine the question of the source corrections in positron annihilation lifetime (PAL) spectroscopy once more.

Our interest in this problem arose in connection with positron studies of layered materials. The paths of positrons in a material can in principle be traced. Schrader [5] suggested using the Monte Carlo method for determination of positron annihilation fractions in multilayered materials. In [6, 7] a simple Monte Carlo algorithm for the same purpose was suggested. We used this algorithm to write a Pascal 6.0 code (called Layer) for computation of the fraction of positrons annihilating in each layer of a layered medium provided that the absorption and backscattering coefficients are known. This program was used to determine the positron absorption coefficient in Kapton and to suggest an algorithm for making the source correction to the positron lifetime spectra of samples with different average atomic numbers measured with a source consisting of ^{22}NaCl salt between two Kapton foils. The algorithm was verified by positron lifetime measurements on a series of annealed metal samples.

2. Experimental procedure

The positron lifetime spectrometer used was of the fast–fast coincidence type. It provided a time resolution of 250 ps FWHM. One characteristic spectrum containing 10^6 counts

was collected for 15 000 s. Four spectra were recorded for each source–samples sandwich arrangement. The spectra were analysed with the POSITRONFIT EXTENDED program [8]. The lifetimes and intensities shown in table 1 are each weighted means over four values.

The positron source with activity 13 ± 1 μCi was made by evaporating a few drops of ^{22}NaCl solution on a 7 μm ($\sim$1 mg cm^{-2}) thick Kapton foil and covering the salt with another identical foil. The polyamide Kapton was chosen because of its very suitable properties for a positron-source-supporting foil. It is commercially available in the form of thin foil with sufficiently good mechanical strength. Its positron properties are very suitable because it shows only a single component that does not vary with temperature [9]. For the measurements of the absorption coefficient of Kapton the method proposed by Hansen *et al* [10] was applied. The samples consisted (figure 1) of different numbers of Kapton foils stacked on the top of indium discs whose thickness was sufficient to absorb all incident positrons. Indium (99.99%) was chosen because it can be assumed that, due to its low melting temperature, there are no defects in it at room temperature. To study the foil thickness effect 7 μm and 25 μm Kapton foils were used. The total Kapton thickness of the stacks was changed from 7 μm to 639 μm.
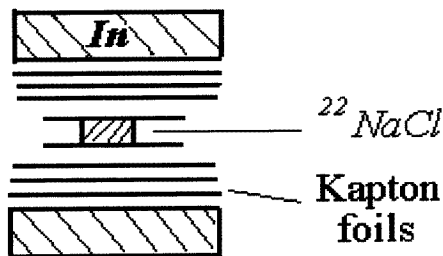

Figure 1. A schematic drawing of the source–sample set-up.

The parameter that allowed us to discern the fraction of positrons annihilating in Kapton and In was the lifetime of positrons in these materials.

3. Results and discussion

3.1. Positron lifetimes in ^{22}NaCl radioactive material

The mean lifetime of positrons annihilating in ^{22}NaCl salt itself was determined making use of ^{22}NaCl which was dried from solution directly onto an In plate and covered with another identical plate. Two-component analysis of spectra yielded $\tau_1 = 196 \pm 1$ ps and $\tau_2 = 415 \pm 3$ ps. The shorter lifetime is close to the bulk positron lifetime in indium [11]. The longer lifetime was ascribed to positron annihilation in ^{22}NaCl salt and the value of 415 ps was used in this study.

3.2. Positron lifetimes in Kapton

The lifetime of positrons annihilating in Kapton was determined with samples each consisting of 50 Kapton foils 25 μm thick. The full thickness is enough for absorption of all incident positrons. Two-component analysis of the lifetime spectra failed to converge because of the very similar values of positron lifetimes in ^{22}NaCl and Kapton. So, the value

$\tau = 382 \pm 3$ ps, obtained from one-component analysis of the spectra, is somewhat higher (in our estimation, about 1–4 ps depending on the amount of ^{22}NaCl salt) than the 'real' value of the positron lifetime in Kapton. The value $\tau = 382$ ps is in good agreement with the published data [2, 9].

To check the foil thickness effect, some of the 25 μm foils were replaced by 7 μm foils. This had no effect provided that the full thickness of the Kapton foils was the same. This means that there is no measurable fraction of positrons that have diffused back after implantation in the foils to the surface where they can annihilate with another lifetime.

3.3. Kapton absorption coefficients

The analysis of the lifetime spectra when an ensemble of a stack of Kapton foils and an In plate (figure 1) was used as the sample was performed with two lifetime components without any source correction. When the positron lifetime in In was fixed at $\tau_2 = 196$ ps, the value of the first component varied between 369 ps and 396 ps with the mean value $\tau_1 = 382 \pm 1$ ps. The value of the intensity of the second component I_2 was interpreted as the relative intensity of the positrons passing through Kapton foils. Figure 2 shows the relative transmitted intensity I_2 as a function of the total thickness of Kapton foils.

As has been pointed out recently by many investigators (see, e.g., [2, 5–7]) the positron implantation profile is not of purely exponential type. Because of this we made use of the program Layer for numerical analysis of the experimental results from positron annihilation in **In**–Kapton–^{22}NaCl–Kapton–**In** multilayers. By an iterative procedure varying the absorption coefficient α_{Kapton} of positrons in Kapton and the fraction I_{NaCl} of positrons annihilating in ^{22}NaCl (as its thickness is unknown), a best fit (the continuous curve in figure 2) for the experimental points was obtained for $I_{NaCl} = (4.9 \pm 0.4)$% and $\alpha_{Kapton} = 28.8 \pm 0.5$ cm^{-1}. The intensity I_{NaCl} corresponds to a ^{22}NaCl salt thickness of about 1.4 ± 0.1 μm.
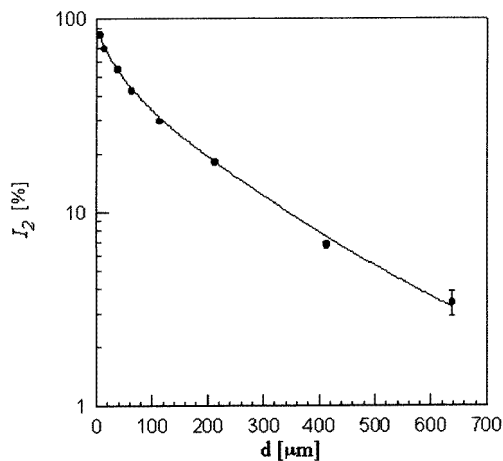

Figure 2. The fraction of positrons transmitted through Kapton foils as a function of total Kapton thickness.

The necessary backscattering coefficients were calculated from the formula given by Schrader *et al* [12]. The absorption coefficient for ^{22}NaCl was calculated from [13]

$$\alpha = 31.24 \rho Z^{0.0878} \tag{1}$$

where ρ is the mass density and Z is the average atomic number of the relevant material ($Z_{Kapton} = 4.2$).

The experimental value of α_{Kapton} is less than that calculated from (1). The main reason for this difference is most probably that while the value given by Arifov *et al* is for positrons injected in a 4p geometry, our value is rather related to the absorption for positrons moving in a certain direction. The value of α_{Kapton} is very close to those given in [2].

3.4. Positron implantation in Kapton source-supporting foils

We used the program Layer to calculate the fraction of positrons that annihilate in radioactive material coating foils (two 7 μm Kapton foils) using α_{Kapton} given in section 3.3 for the absorption coefficient of Kapton (if values for the absorption coefficient for Kapton given by Plotkowski *et al* are used as α_{Kapton}, I_{Kapton} is increased by about 1%). The ratio $I_{NaCl}/I_{Kapton} = 0.34$ obtained for In as described in section 3.3 was kept constant over the whole range of Z. The results of these calculations are shown in figure 3.
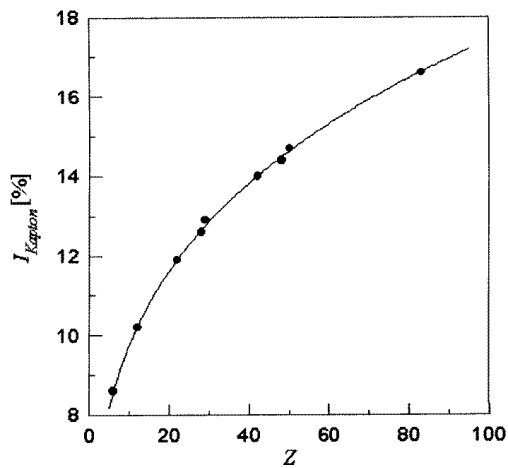

Figure 3. The fraction of positrons annihilated in the source-supporting Kapton foils as a function of the atomic number, Z, of the sample, calculated using the program Layer.

The black dots, 'experimental data', on the figure were calculated with 106 positrons. The curve represents a least-squares fit of the experimental data with a power function:

$$F(Z) \equiv I_{Kapton}(Z) = a + bZ^c \qquad (2)$$

with $a = -0.42$, $b = 5.83$ and $c = 0.242$.

Very often it is worth simplifying the data analysis by ascribing only one lifetime component to the source. From the formula

$$\tau_m = (\tau_1 I_1 + \tau_2 I_2)/(I_1 + I_2) \qquad (3)$$

for the determination of the mean lifetime, a value $\tau_{source} = 391$ ps was obtained using $t_{NaCl} = 415$ ps, $I_{NaCl} = 4.9\%$ and $t_{Kapton} = 382$ ps, $I_{Kapton} = 14.6\%$ (for $Z = 49$).

So, the total source contribution to lifetime spectra for In plates as samples is $I_{source} = I_{NaCl} + I_{Kapton} = 19.5\%$ and $\tau_{source} = 391$ ps. We consider that the ratio $f = I_{source}/I_{Kapton}$ is a characteristic feature of a given positron source. Its value for the positron source under consideration is $f = 1.34$.

To obtain the fraction of positrons that annihilate in the same source when samples with average atomic number different from $Z = 49$ are under study it is necessary to calculate $I_{Kapton}(Z)$ from formula (2) and to multiply the results by $f = 1.34$.

To verify the method for source correction of lifetime spectra, the positron lifetimes in a number of pure well-annealed metals have been measured.

Annealing was done in argon atmosphere for one hour at a temperature of $T = 0.75T_m$, where T_m is the melting temperature of the corresponding metal. The cooling down of the samples to room temperature was very slow (10 hours). When necessary, chemical polishing of the samples after annealing was performed.

Each lifetime spectrum was analysed in terms of two components without any source correction. The second component was fixed at 391 ps. Its intensity I_2 was interpreted as a source contribution I_{source} to the spectrum. The results are presented in table 1 and figure 4. The last column of the table contains the published values of bulk lifetimes for comparison.

Table 1. The lifetime τ_1, source intensity $I_{source} \equiv I_2$ and bulk lifetime τ_b.

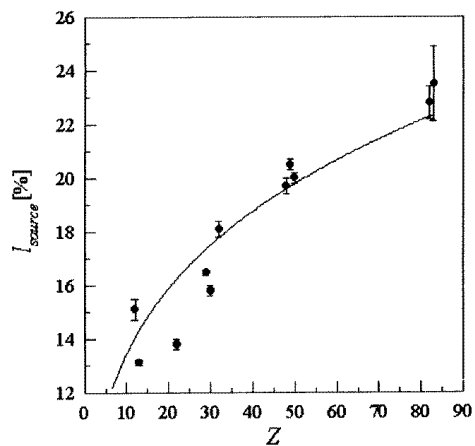| Element	Z	τ_1 (ps)	I_{source} (%)	τ_b (ps)
Mg	12	224 ± 1	15.1 ± 0.4	225 [4, 14]
Al	13	165 ± 1	13.1 ± 0.2	166 [4], 163 [14]
Ti	22	149 ± 1	13.8 ± 0.2	154 [4], 147 [14]
Cu	29	121 ± 1	16.5 ± 0.1	110 [14]
Zn	30	156 ± 1	15.8 ± 0.2	175 [14], 154 [4]
Ge	32	231 ± 1	18.1 ± 0.3	228 [4]
Cd	48	185 ± 1	19.7 ± 0.3	190 [14], 184 [4]
In	49	191 ± 1	20.5 ± 0.2	199 [4], 192 [11], 193 [14]
Sn	50	195 ± 1	20.0 ± 0.2	201 [14]
Pb	82	207 ± 2	22.8 ± 0.6	204 [4], 194 [14]
Bi	83	249 ± 1	23.5 ± 1.4	241 [4]

Figure 4. The fraction of positrons annihilated in the source as a function of the atomic number, Z, of the sample.

The line in figure 4 represents the least-squares fit for the experimental points with a function of the type $F'(Z) = gF(Z)$, where g is a fitting parameter and $F(Z)$ is given

by formula (2). Obviously, if the suggested algorithm for source correction is correct, the value of g must coincide with the ratio f defined in section 3.4. In fact, the value obtained for g is $g = 1.3 \pm 0.1$.

3.5. The algorithm for determination of the source correction

All considerations above concern just one source of positrons. The only difference between this source and any other source consisting of another quantity of ^{22}NaCl salt packed between two 7μm foils is their different values of the ratio $f = I_{source}/I_{Kapton}$ and, as a result, their different values of τ_{source} calculated from f and formula (3).

The ratio f can be determined as follows.

(i) Positron lifetimes in the well annealed one-component reference sample have to be measured.

(ii) The spectrum has to be analysed using the one-component source correction with $\tau = 382$ ps and I_{Kapton} calculated from formula (2) by substituting in the value of Z for the sample. Two-component analysis $(\tau_1(I_1), \tau_2(I_2))$ has then to be performed, with the second lifetime fixed at 415 ps. The fraction I_{NaCl} (in %) of positrons that annihilate in ^{22}NaCl salt itself is calculated using I_2 from the formula

$$I_{NaCl} = I_2(100 - I_{Kapton})/100 \tag{4}$$

as the sum $I_{NaCl} + I_{sample} + I_{Kapton}$ is equal to 100%.

The fraction of positrons that annihilate in the source is then given by

$$I_{source} = I_{NaCl} + I_{Kapton} \tag{5}$$

and

$$f = I_{source}/I_{Kapton}. \tag{6}$$

This value has to be used thereafter for the measurements with the same source. Further, the source correction, when positron lifetimes in a sample with average atomic number Z are under study, is determined as was described in section 3.4, as we now describe below.

(i) The fraction, I_{Kapton}, of positrons that annihilate in Kapton supporting foils is calculated from formula (2).

(ii) The source component intensity I_{source} is obtained from (6) via the relation

$$I_{source} = f I_{Kapton}.$$

(iii) The source lifetime component τ_{source} is calculated from (3) with $\tau_1 = 382$ ps, $I_1 = I_{Kapton}$, $\tau_2 = 415$ ps and $I_1 + I_2 = I_{source}$ or alternatively τ_1 and τ_2 can be used separately as source components.

To check this algorithm we used a second source with activity 61 ± 1 μCi. As a reference sample we chose two identical annealed Bi plates. The results of two-component analysis of the lifetime spectra when τ_2 was fixed at 415 ps and the source correction was $I_{Kapton} = 16.5\%$ (from formula (2)), and $\tau_2 = 382$ ps, are: $\tau_1 = 246 \pm 2$ ps and $I_2 = 10.3 \pm 0.2\%$. Following the procedure described, a value of $f = 1.5$ for the positron source was determined and $\tau_{source} = 393.5$ ps. Using this value of f, the positron lifetimes in lead and copper were found by measurement to be $\tau_{Pb} = 204 \pm 1$ ps and $\tau_{Cu} = 120$ ps in good agreement with values given in table 1.

The experience with the proposed algorithm for the determination of the source correction in positron lifetime spectroscopy showed that a tolerance of 2 ps in τ_{source} and of 1–2% in I_{source}, depending on the value of Z of the sample, affects the values obtained for the lifetimes and their intensities within their error ranges.

Acknowledgments

References

[1] Bertolaccini M and Zappa L 1967 *Nuovo Cimento* B **52** 487
[2] Plotkowski K, Panek T J and Kansy J 1988 *Nuovo Cimento* D **10** 933
[3] Saocha A, Pedersen N J and Eldrup M 1992 *Mater. Sci. Forum* **105–110** 1971
[4] Monge M A and del Rio J 1994 *J. Phys.: Condens. Matter* **6** 2643
[5] Schrader D M 1988 *Positron and Positronium Chemistry (Studies in Physical and Theoretical Chemistry 57)* ed D M Schrader and J C Jean (Amsterdam: Elsevier) ch 2
[6] Gregory R B and Wen Su 1991 *Nucl. Instrum. Methods Phys. Res.* B **62** 1
[7] Gregory R B and Wen Su 1992 *Mater. Sci. Forum* **105–110** 1875
[8] Kirkegaard P, Eldrup M, Mogensen O E and Pedersen N J 1981 *Comput. Phys. Commun.* **23** 307
[9] MacKenzie I K and Fabian J 1980 *Nuovo Cimento* B **58** 162
[10] Hansen H E, Linderoth S and Petersen K 1982 *Appl. Phys.* A **29** 99
[11] del Rio J, Plazaola F, de Diego N and Moser P 1994 *Solid State Commun.* **89** 913
[12] Schrader D M, Chin S W, Nakanishi H and Rochanakij S 1985 *Positron Annihilation* ed P C Jain, R M Singru and K P Gopinathan (Singapore: World Scientific) p 822
[13] Arifov P U, Grupper A R and Alimkulov H 1982 *Positron Annihilation* (Amsterdam: North-Holland) p 699
[14] Seeger A and Banhart F 1987 *Phys. Status Solidi* a **102** 171