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TO THE MEASUREMENTS OF $3\gamma/2\gamma$ RATIO FOR POSITRON ANNIHILATION IN MATTER USING ANNIHILATION ENERGY SPECTRUM

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Processes of the summation of cascade γ-quanta usually neglected, when registering the annihilation energy spectrum for the determination of the three-photon annihilation probability $P_{3\gamma}$ of positronium in samples, are studied. The deviations of the actual $P_{3\gamma}$ value from that determined without allowance made for the summation processes are 56% and 25% for NaI(Tl) and Ge detectors placed at the distance of 3 cm from a positron source, respectively.

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1. Introduction

As it is known (see, for example [1, 2]) there are two ground states of positronium (Ps) depending on the total moment of the electron and positron forming the atom. They are the singlet state *para*-Ps (*p*-Ps) and the triplet state *ortho*-Ps (*o*-Ps). The statistical weight of the triplet Ps state is three times as large as that of the singlet state, yielding a three times higher formation probability of *o*-Ps compared with *p*-Ps. *o*-Ps annihilates into three photons with the continuous energy spectrum ranging from 0 to 0.511 MeV while *p*-Ps annihilates into two photons each flying in the opposite direction with the fixed energies of 0.511 MeV. The lifetime of *p*-Ps in vacuum is $\tau_s^0 = 1.25 \times 10^{-10}$ s whereas that of *o*-Ps is $\tau_t^0 = 1.4 \times 10^{-7}$ s.

As a rule, quenching processes (pick-off-annihilation, *ortho-para*-conversion, and chemical interactions) lead to an essential decrease (by one order of magnitude and more [3–6]) of τ_t, the *o*-Ps lifetime in matter, compared with τ_t^0.

The probability of the three-photon decay of Ps, $P_{3\gamma}$, is one of the most important parameters characterizing Ps in matter [7–11]. Therefore, its precise experimental measurement is ultimately important to know the structure of matter under investigation.

2. Methods of $P_{3\gamma}$ determination

The probability of the Ps formation in matter, P, is concerned with the probability $P_{3\gamma}$ of its 3γ-annihilation through the known expression [1, 12]:

$$P_{3\gamma} \cong 0.75P\frac{\tau_t}{\tau_t^0} + \frac{(1-P)}{372}. \tag{1}$$

The basic methods of $P_{3\gamma}$ determination are described in [13]. They are the lifetime [14–18] and magnetic quenching [19] methods, the angular-correlation method [20], and the three-photon yield method. The latter one is based on using a triple-coincidence apparatus, or upon the measurements of the annihilation radiation energy spectrum which contains the 511 keV line corresponding to two-photon annihilation and the continuous spectrum of γ-rays produced in three-photon annihilation events [21, 22].

$P_{3\gamma}$ can be obtained by means of the comparison of experimental energy spectra measured in a sample under investigation and in a sample in which Ps is certainly not formed (for example, pure (99.999%) Al) so that the probability of the three-photon decay does not exceed 1/372, the 3γ-annihilation probability of a free positron. This is the basis of the one more method of the $P_{3\gamma}$ experimental determination similar to that described in [21, 22]. This method was called the "peak–peak" method and discussed in detail in [23].

The essence of the method is as follows. The majority of investigations of solids by positrons are done with ^{22}Na positron sources. The emission of a positron by the decaying nuclei ^{22}Na is accompanied by the emission of the 1.275 MeV nuclear γ-quantum (100% gamma-rays per positron). Let ε_1 be the efficiency of the 0.511 MeV annihilation quantum into the total absorption peak, i.e. the product of the physical efficiency by the solid angle from the "positron source + sample" system to the detector, ε_2 be the efficiency of the nuclear quantum into the corresponding total absorption peak. Then, in view of the fact that the three-photon annihilation of a free positron is negligibly small (1/372), one has for the standard Al sample

$$(S_{511})_{\mathrm{Al}} = 2\varepsilon_1 Q, \tag{2}$$

$$(S_{1275})_{\mathrm{Al}} = \varepsilon_2 Q, \tag{3}$$

where $(S_{511})_{\mathrm{Al}}$ and $(S_{1275})_{\mathrm{Al}}$ being the count rates of the annihilation and nuclear quanta, respectively, Q is the activity of a positron source. The same quantities for a sample under investigation are

$$(S_{511})_{\mathrm{Ps}} = 2\varepsilon_1(1 - P_{3\gamma})Q. \tag{4}$$

The 1.275 MeV-line intensity does remain unchanged since it does not depend on the structure of a sample being investigated, i.e.

$$(S_{1275})_{\mathrm{Ps}} = \varepsilon_2 Q. \tag{5}$$

To experimentally determine the value of $P_{3\gamma}$ it is necessary to compare the two energy spectra (the spectrum in the standard Al sample and that in a sample being investigated) under the natural assumption of the equality of positrons emitted in both cases. Thus, the energy spectra must contain the nuclear γ-radiation total absorption peaks of the same area under the assumption of due regard for the background count rate, i.e.

$$(S_{1275})_{\mathrm{Al}} = (S_{1275})_{\mathrm{Ps}}. \tag{6}$$

Based on this fact the spectra are normalized so that the areas under the nuclear radiation total absorption peaks are the same. Then the quantity experimentally observed is

$$\Delta = (S_{511})_{\mathrm{Al}} - (S_{511})_{\mathrm{Ps}} = 2\varepsilon_1 P_{3\gamma} Q, \tag{7}$$

whence

$$P_{3\gamma} = \frac{\Delta}{2\varepsilon_1 Q}. \tag{8}$$

3. Summation processes of the cascade γ-radiation in $P_{3\gamma}$ determination

Incorrect background as well as random coincidences of quanta from different decays are out of the scope of the present paper and, therefore, will not be considered. Below, physical effects occurring when registering the cascade γ-radiation will be discussed for the decays described above.

Let $\varepsilon_1^{\mathrm{c}}$ and $\varepsilon_2^{\mathrm{c}}$ be the efficiency of the 0.511 MeV annihilation photon and 1.275 MeV nuclear photon scattered by means of the Compton scattering, respectively, $\varepsilon_3^{\mathrm{t}}$ be the total efficiency of the photon originated from the *o*-Ps decay (average probability that a γ-photon from *o*-Ps decay will be counted in the full energy spectrum). Then, for the standard Al sample, in view of the fact that the positron and 1.275 MeV photon are emitted within the same cascade, additional processes occur in the detector which are resulted from the summation of the quanta (originated from the same nuclear decay) coming into the detector simultaneously:

(1) registration of the annihilation quantum and coincident nuclear quantum scattered via Compton scattering each, the count rate of the process is $2\varepsilon_1^{\mathrm{c}}\varepsilon_2^{\mathrm{c}}Q$;

(2) registration of the Compton scattered annihilation quanta along with the nuclear quanta of the total absorption peak — $2\varepsilon_1^{\mathrm{c}}\varepsilon_2 Q$;

(3) registration of the annihilation quantum of the total absorption peak along with the Compton scattered nuclear quantum — $2\varepsilon_1\varepsilon_2^{\mathrm{c}}Q$;

(4) registration of the annihilation quantum of the total absorption peak and coincident nuclear quantum of the total absorption peak — $2\varepsilon_1\varepsilon_2 Q$.

Processes (3) and (4) lead to the variation of the intensity of the 0.511 MeV total absorption peak, while processes (2) and (4) lead to the variation of the intensity of the 1.275 MeV total absorption peak. Therefore, one has for 0.511 Mev-line and 1.275 MeV-line

$$(S_{511})^*_{\mathrm{Al}} = 2\varepsilon_1 Q(1-\varepsilon_2^{\mathrm{t}}), \tag{9}$$

$$(S_{1275})^*_{\mathrm{Al}} = \varepsilon_2 Q(1-2\varepsilon_1^{\mathrm{t}}), \tag{10}$$

where $\varepsilon_2^{\mathrm{t}} = \varepsilon_2 + \varepsilon_2^{\mathrm{c}}$ and $\varepsilon_1^{\mathrm{t}} = \varepsilon_1 + \varepsilon_1^{\mathrm{c}}$ are the total efficiencies of the 1.275 MeV nuclear quantum and 0.511 MeV annihilation quantum, respectively.

In the case of the "Ps-forming" sample under investigation, o-Ps annihilates into three γ-quanta with the energy smaller than 0.511 MeV each. These quanta come into the Compton part of the spectrum rather than into the total absorption peak. Therefore, one has the following processes similar to those described above:

(1) $[2\varepsilon_1^{\mathrm{c}}(1-P_{3\gamma}) + 3\varepsilon_3^{\mathrm{t}} P_{3\gamma}]\varepsilon_2^{\mathrm{c}} Q$,

(2) $[2\varepsilon_1^{\mathrm{c}}(1-P_{3\gamma}) + 3\varepsilon_3^{\mathrm{t}} P_{3\gamma}]\varepsilon_2 Q$,

(3) $2\varepsilon_1(1-P_{3\gamma})\varepsilon_2^{\mathrm{c}} Q$,

(4) $2\varepsilon_1(1-P_{3\gamma})\varepsilon_2 Q$.

Hence, one has

$$(S_{511})^*_{\mathrm{Ps}} = 2\varepsilon_1 Q(1-2\varepsilon_2^{\mathrm{t}})(1-P_{3\gamma}), \tag{11}$$

$$(S_{1275})^*_{\mathrm{Ps}} = \varepsilon_2 Q\left[1-2\varepsilon_1^{\mathrm{t}} + P_{3\gamma}(2\varepsilon_1 - 3\varepsilon_3^{\mathrm{t}})\right]. \tag{12}$$

Let us now analyze consequences of the requirement of the equality of the areas under the nuclear γ-quantum total absorption peaks, which is commonly used when comparing spectra. Such a requirement yields the following normalization coefficient to be multiplied by the spectrum in the Al standard sample:

$$\frac{(S_{1275})^*_{\mathrm{Ps}}}{(S_{1275})^*_{\mathrm{Al}}} = \frac{[1-2\varepsilon_1^{\mathrm{t}} + P_{3\gamma}(2\varepsilon_1 - 3\varepsilon_3^{\mathrm{t}})]}{(1-2\varepsilon_1^{\mathrm{t}})}. \tag{13}$$

In this case, one gets

$$P^*_{3\gamma} = \frac{\Delta^*(1-2\varepsilon_1^{\mathrm{t}})}{2Q\varepsilon_1(1-\varepsilon_2^{\mathrm{t}})(1-3\varepsilon_3^{\mathrm{t}})}, \tag{14}$$

whence it follows that the actual 3γ-annihilation probability differs by the factor of

$$k = \frac{(1-2\varepsilon_1^{\mathrm{t}})}{(1-\varepsilon_2^{\mathrm{t}})(1-3\varepsilon_3^{\mathrm{t}})} \tag{15}$$

from that defined by Eq. (8).

4. Results

The estimated values of the coefficient k are shown in the Table for detectors of some types commonly used. To obtain the estimates, experimental data were used for the NaI(Tl) (150 × 100 mm) and high efficiency Ge detector GC8021 of volume 200 cm^3. For simplicity, the value of ε_3^t was taken to be equal to the total registration efficiency of the 0.392 MeV quantum.

TABLE

Dependence of the coefficient k for the NaI(Tl) (150 × 100 mm) and high efficiency Ge detector GC8021 of the volume 200 cm^3. ε_1 and ε_2 are the efficiency of the 0.511 MeV annihilation quantum and the 1.275 MeV nuclear quantum into the corresponding total absorption peak; ε_1^c and ε_2^c are the efficiency of the 0.511 MeV photon and 1.275 MeV photon scattered by means of the Compton scattering, respectively; ε_3^t — the total registration efficiency of the 0.392 MeV quantum; R — distance from the source and sample to the surface of the detector.

NaI(Tl) 15 × 10 cm						
R [cm]	ε_1	ε_1^c	ε_2	ε_2^c	ε_3^t	k
3	0.11	0.046	0.07	0.083	0.16	1.56
10	0.037	0.015	0.023	0.028	0.052	1.12
20	0.013	0.005	0.008	0.0095	0.017	1.035
Ge 200 cm^3						
3	0.036	0.072	0.019	0.069	0.103	1.25
10	0.009	0.017	0.005	0.015	0.026	1.05
20	0.0025	0.0065	0.0016	0.006	0.010	1.02

The results of experimental investigation of the $P_{3\gamma}$ values in SiO_2-samples with 150 × 100 mm NaI(Tl) detector using the method described in [23] are in agreement with the data in the Table.

5. Summary

As it is seen in the Table, the neglect of the summation processes described leads to the increase in the actual $P_{3\gamma}$ if the distance from the source to the detector does not exceed 10 cm. It must be emphasized that the summation processes are important regardless of the value of the lower energy threshold set when registering spectra (evidently, the maximal value of the energy threshold cannot exceed 0.5 MeV).

Note that the correction of experimental spectra to take the summation processes into account is unnecessary if the solid angle from the sample onto the detector does not exceed 2% of 4π steradian, or the positron beam is used without the concomitant γ-radiation.

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References

[1] V.I. Goldansky, *Physical Chemistry of Positron and Positronium*, Nauka, Moscow 1968, p. 13.

[2] A. Rich, *Rev. Mod. Phys.* **53,** 127 (1981).

[3] A. Seeger F. Banhart, *Phys. Status Solidi A* **102,** 171 (1987).

[4] J. Dryzek, *Acta Phys. Pol. A* **95,** 527 (1999).

[5] M. Bokor, T. Marek, K. Süvegh, Z.S. Böcskei, J. Buschmann, A. Vértes, *Acta Phys. Pol. A* **95,** 469 (1999).

[6] K. Ciesielski, A.L. Dawidowicz, T. Goworek, B. Jasinska, J. Wawryszczuk, *Chem. Phys. Lett.* **289,** 41 (1998).

[7] K. Ciesielski, A.L. Dawidowicz, T. Goworek, *Acta Phys. Pol. A* **95,** 483 (1999).

[8] P. Kindl, G. Reiter, *Phys. Status Solidi A* **104,** 707 (1987).

[9] J. Serna, *Phys. Status Solidi A* **122,** 489 (1990).

[10] H.-E. Schaefer, *Phys. Status Solidi A* **102,** 47 (1987).

[11] K. Jerie, A. Baranowski, J. Gliński, K. Orzechowski, *Acta Phys. Pol. A* **95,** 562 (1999).

[12] R.N. West, *Adv. Phys.* **22,** 263 (1973).

[13] A. Dupasquier, in: *Positron Solid-State Physics*, Eds. W. Brand, A. Dupasquier, North-Holland, Amsterdam 1983, p. 530.

[14] W. Osoba, *Nukleonika A* **42,** 181 (1997).

[15] J. Kansy, *Nucl. Instrum. Methods Phys. Res. A* **374,** 235 (1996).

[16] J. Serna, J.Ch. Abbe, G. Duplatre, *Phys. Status Solidi A* **115,** 389 (1989).

[17] V.G. Baryshevsky, O.N. Metelitsa, V.V. Tikhomirov, S.K. Andrukhovich, A.V. Berestov, B.A. Martsynkevich, E.A. Rudak, *Phys. Lett. A* **136,** 428 (1989).

[18] V.V. Baranov, A.V. Berestov, B.A. Martsynkevich, E.A. Rudak, A.S. Talat, *Instrum. Exp. Tech.* **38,** 18 (1995).

[19] G. Consolati, F. Quasso, *Nukleonika A* **42,** 75 (1997).

[20] C. Rybka, T. Goworek, K. Ciesielski, *Nukleonika A* **42,** 205 (1997).

[21] S. Marder, V.W. Hughes, C.S. Wu, W. Bennett, *Phys. Rev.* **103,** 1258 (1956).

[22] K.G. Lynn, D.O. Welch, *Phys. Rev. B* **22,** 99 (1980).

[23] P.U. Arifov, S.N. Vasserman, A.R. Grupper, S.A. Tishin, in: *Methods of Positron Diagnostics and Decipher of Positron Annihilation Spectra*, Ed. P.K. Khabybullaev, Fan, Tashkent 1985, p. 34.