Romanian Reports in Physics, Vol. 60, No. 3, P. 453–469, 2008

Dedicated to Prof. Ioan-Iovitz Popescu's 75th Anniversary

ON SOME EXPERIMENTAL OBSERVATIONS OF QUANTAL BEATS IN POSITRONIUM THREE- AND TWO-GAMMA ANNIHILATION IN MAGNETIC FIELD

M. APOSTOL, I. VATA

Institute for Atomic Physics,
Magurele-Bucharest MG-6, POBox MG-35, Romania
E-mail: apoma@theory.nipne.ro

(Received February 2, 2008)

Abstract. Local magnetic interactions in matter may give rise to transitions between orto- and parapositronium populations. The quantum beats generated by an external magnetic field in the three-gamma annihilation rate of orthopositronium can be transferred onto the two-gamma annihilation rate of parapositronium (511 keV radiation) via such a coupling. The oscillations in the two-gamma decay of parapositronium could, in principle, be seen experimentally over the orthopositronium lifetime. However, their amplitude is, typically, much smaller (by one to three orders of magnitude) than the amplitude of the oscillations in the orthopositronium three-gamma decay rate, which gives little hope to be seen experimentally. At the same time, the coupling between ortho- and parapositronium populations may change the rate constant of the orthopositronium three-gamma decay, while the component constant in time of the local magnetic interactions may give rise to a shift in the quantum beats frequency.

Key words: positronium, gamma disintegration, quantum beats in magnetic field.

PACS: 36.10.k; 42.50.Md; 76.80.+y

INTRODUCTION

There has been shown recently [1–4] that the three-gamma (3γ) annihilation cross-section of orthopositronium (triplet state T, spin $S = 1$) exhibits quantal beats in moderate magnetic fields ($H \sim 500$ Gs), as a consequence of the mixing in the spin-polarization matrix of the spin states $|1 \pm 1\rangle$ ($S = 1$, $m = \pm 1$, ground-state hyperfine energy $W_1 \simeq 4.7 \cdot 10^{-4}$ eV) and a fraction of the spin state $|10\rangle$ ($S = 1$, $m = 0$). This fraction is determined by the magnetic field from the competition with the parapositronium (singlet state S, spin $S = 0$) state $|00\rangle$ ($S = 0$, $m = 0$, ground-state hyperfine energy $W_0 \simeq -3.5 \cdot 10^{-4}$ eV). The beat frequency is $\sim \Delta W (4\mu H/\Delta W)^2/4\hbar \simeq 2.6 \cdot 10^8 H^2$ [kGs] ($\Delta W = W_1 - W_0 \simeq 8.3 \cdot 10^{-4}$ eV and $\mu =$

$= e\hbar/2mc \simeq 9.3\cdot 10^{-24}$ J/Ts is the Bohr magneton), or $\sim 6.5\cdot 10^7$ s^{-1} for a magnetic field $H = 500$ Gs, high enough to be seen as 6–7 cycles during triplet positronium lifetime $\tau_t \simeq 10^{-7}$ s, and sufficiently low to lie within the detector resolution $\sim 10^9$ s^{-1}.

There has been rumoured recently [5] that similar oscillations, with the same frequency, could have been observed in the parapositronium 2γ-annihilation cross-section. The present note is devoted to analyzing this point.

Positronium has a few particularities. [6] Its energy levels are those of a hydrogenoid atom $E_n = me^4/4\hbar^2 n^2$ with the relative mass $m/2$. These levels are splitted by relativistic corrections, arising from both orbital, spin-orbit interaction and spin-spin interaction. They bring about a fine structure of the levels, and a hyperfine structure, the latter coming from the spin-spin interaction. To the first order, these corrections are of the order of the fine-structure constant $\alpha = e^2/\hbar \simeq 1/137$ squared. Since the electromagnetic interaction conserves the combined charge-inversion parity for this neutral pair, the total spin S is a "good" quantal number in this case. In addition, the total momentum j conserves too, so the energy levels can be classified by S and j. Orthopositronium corresponds to $S = 1$ (triplet spin-state T), and parapositronium corresponds to $S = 0$ (singlet spin-state S). Angular momentum l takes the values $l = j$, $j \pm 1$. For the ground state $n = 1$, $l = 0$ the energy has small fine-structure corrections in α^2 and a hyperfine splitting $W_1(S=1) - W_0(S=0)$, as indicated above, also proportional to α^2. It is assumed that positronium is mostly in its ground state in matter, its excited states obeying a rapid relaxation. Another particularity of positronium is its vanishing of the orbital magnetic momentum, while its spin magnetic momentum is not proportional to the spin S. Consequently, the Zeeman effect in a magnetic field mixes up the orthopositronium and the parapositronium states, specifically $|10\rangle$ and $|00\rangle$, and the dependence on the magnetic field is quadratic for low fields, and linear for high fields [7].

Positronium decays by electron-positron annihilation with gamma (γ) emission. Since the relative (internal) momentum of positronium vanishes it only decays by releasing at least 2γ's. However, the total angular momentum of a pair of photons cannot be one, [6] therefore ortopositronium in the ground state cannot decay by releasing 2γ's. Parapositronium in the ground state decays, however, by releasing 2γ's. In fact, the charge parity of orthopositronium is negative, so, by Furry's theorem, it cannot release an even number of photons [6]. Orthopositronium decays only by releasing an odd number of photons. In contrast, parapositronium has a positive charge parity, so it cannot release an odd number of photons. Parapositronium only decays by releasing an even number of photons. The main annihilation processes are therefore 3γ-decay for orthopositronium, and 2γ-decay for parapositronium. (It is worth recalling in this context that the 3γ-decay amplitude squared is by a fine-structure constant α factor smaller that the 2γ-decay

amplitude squared). These processes determine the lifetimes of the two positronium varieties. The cross-sections of these processes can be related to those of the annihilation of a free electron-positron pair in the non-relativistic limit. The singlet lifetime (of parapositronium) is $\tau_s \simeq 10^{-10}$ s (too short to be seen experimentally by usual detection resolution 10^9 s^{-1}), while the orthopositronium (triplet) lifetime is $\tau_t \simeq 10^{-7}$ s. They are much longer than the period of the hyperfine energy levels ($\hbar/\Delta W \simeq 10^{-12}$ s), which justifies the existence of these spin varieties of positronium.

THE RATE OF MIXING UP ORTHO- AND PARAPOSITRONIUM

Let

$$\chi_0 = \frac{1}{\sqrt{2}}(\alpha_+\beta_- - \alpha_-\beta_+), \qquad \chi_1 = \frac{1}{\sqrt{2}}(\alpha_+\beta_- + \alpha_-\beta_+)\ , \qquad \chi_2 = \alpha_+\alpha_-, \quad \chi_3 = \beta_+\beta_- \tag{1}$$

be the spin states of the positronium, where α, β stand for spin up and, respectively, spin down, and $\pm$ refer to positron and, respectively, electron. The unperturbed hamiltonian for the hyperfine splitting can be represented as

$$H_0 = W_0 + \frac{1}{2}(W_1 - W_0)S^2\,, \tag{2}$$

where $\mathbf{S} = \boldsymbol{\sigma}_+ + \boldsymbol{\sigma}_-$ is the total spin, so that $H_0\chi_0 = W_0\chi_0$ and $H_0\chi_{1,2,3} = W_1\chi_{1,2,3}$. The Zeeman interaction is given by

$$V = -\mu_z H = -\mu(\sigma_{z+} - \sigma_{z-})H\,, \tag{3}$$

where μ is the Bohr magneton and H is the magnetic field aligned along the z-axis (the spin magnetic momentum of the positronium is given by $\boldsymbol{\mu} = \boldsymbol{\sigma}_+ + \boldsymbol{\sigma}_-$). It mixes up the states $\chi_{0,1}$, $V\chi_0 = -2\mu H\chi_1$, $V\chi_1 = -2\mu H\chi_0$, and is diagonal for the other two states, $V\chi_{2,3} = 0$. The hyperfine energy levels of positronium and their Zeeman splitting in magnetic field are shown in Fig. 1.

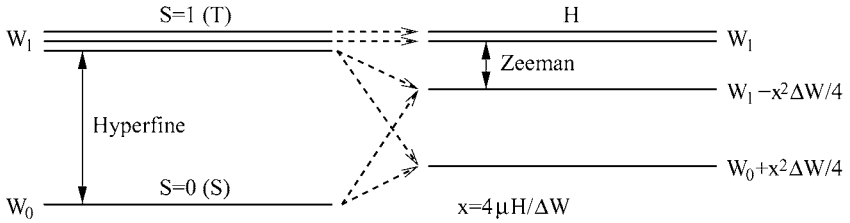

Fig. 1 – Hyperfine energy levels of positronium and their Zeeman splitting in magnetic field.

Such a Zeeman-like interaction can be viewed as a time-dependent perturbation; if introduced suddenly, the transition probability between χ_0 and χ_1 is given by

$$w_{01} = V_{01}^2/\Delta W^2 = 4\mu^2 H^2/\Delta W^2 = x^2/4, \tag{4}$$

where $x = 4\mu H/\Delta W$. For $H = 500$ Gs the parameter x acquires the value $x = 0.014$. The transition probability above suggests the perturbed states

$$\tilde{\chi}_0 = (1 - x^2/8)\chi_0 + \frac{x}{2}\chi_1, \quad \tilde{\chi}_1 = (1 - x^2/8)\chi_1 - \frac{x}{2}\chi_0, \tag{5}$$

and the energies

$$E_0 = W_0 + x^2\Delta W/4, \quad E_1 = W_1 - x^2\Delta W/4. \tag{6}$$

If the perturbation is viewed as being time independent, then, introduced at time zero, it gives the transition probability

$$w_{01} = V_{01}^2 \frac{4\sin^2(\Delta W t/2\hbar)}{\Delta W^2}, \tag{7}$$

which, for short times $\Delta W t/\hbar \leq 1$, becomes $w_{01} \simeq V_{01}^2 t^2/\hbar^2 \simeq V_{01}^2/\Delta W^2$, as in (4). It follows that, indeed, over very short times of the order of $\tau \sim \hbar/\Delta W \sim 10^{-12}$ s ortho- and parapositronium mix up together, to a certain extent as given by (4), under the action of a perturbation of the Zeeman type. This time scale is too short to have any effect on the positronium annihilation and detection of quantal beats in gamma emission, other than leading very quickly to equilibrium (stationary) population mixing of ortho- and parapositronium.

Indeed, the Zeeman-type interaction given by (3) is in fact exactly diagonalized, for eigenfunctions and energies as those given by (5) and, respectively, (6). The eigenstates are expressed as

$$\tilde{\chi}_0 = (1 - x^2/8)\chi_0 + \frac{x}{2}\chi_1, \quad \tilde{\chi}_1 = (1 - x^2/8)\chi_1 - \frac{x}{2}\chi_0, \quad \tilde{\chi}_{2,3} = \chi_{2,3}, \tag{8}$$

and the energies are given by

$$E_0 = W_0 + x^2\Delta W/4, \quad E_1 = W_1 - x^2\Delta W/4, \quad E_{2,3} = W_1. \tag{9}$$

This Zeeman-like splitting $E_{2,3} - E_1 = x^2\Delta W/4$ of the triplet states in magnetic field is much smaller than the hyperfine splitting ΔW.

TRANSITION RATE BETWEEN ORTHO- AND PARAPOSITRONIUM

Positronium in matter is affected by local magnetic interactions, which vary in time, in general. Such interactions can be represented by a magnetic field h, and

they may acquire various expressions, like, for instance, μh, or $a_{ij}\mu_i h_j$ for an anisotropy tensor a_{ij}, or $\sim(\mu_i h_j)^2$, such that their matrix elements are of the order of μh. Magnetic interactions in matter may originate in spin-orbit or spin-spin coupling, like, for instance, dipolar magnetic interactions which are responsible for the linewidth of the paramagnetic resonance (in which case the magnetic field h is represented as a local magnetic momentum μ divided by an atomic scale volume a^3). During collisions with atoms in matter, such interactions vary in time and their spectrum may include a large range of frequencies. The pickoff process is well documented for orthopositronum in matter. During such a process, the positron in orthopositronium, whose wavefunction overlaps neighbouring electrons wavevfunctions, picks up an electron with antiparallel spin and decay through 2γ-emission. Such a process is obviously an ortho- to parapositronium transition (of course, the reverse process exists too, though the parapositronium lives too shortly to affect appreciably the orthopositronium population).

The local magnetic interaction in matter must be much smaller than the Zeeman splitting of the positronium triplet states in the external magnetic field H, $\mu h \ll x^2\Delta W/4$, *i.e.* $h \ll xH$, in order to preserve the quantal beats observed in the 3γ-annihilation rate of orthopositronium. The component constant in time of this interaction of positronium with matter shifts slightly the Zeeman splitting of the hyperfine energy levels, which amounts to replacing the parameter $x = 4\mu H/\Delta W$ introduced above by $\overline{x} = x + 4\mu h/\Delta W$. Since the quantal beats in the 3γ-annihilation cross-section of orthopositronium occurs at frequency

$$\Omega = (E_{2,3} - E_1)/\hbar = x^2\Delta W/4\hbar \tag{10}$$

(approximately $6.5\cdot10^7$ s^{-1} for H = 500 Gs), the effect of the sample magnetization can be seen, in principle, as a small shift in this frequency; this shift can be estimated to $\sim 2x\%$ at most. The experimental observation of these quantal beats is limited by the detector resolution 10^9 s^{-1}, so, under these circumstances (setting up an upper limit $\Omega = 10^9$ s^{-1}), the parameter x may acquire the value $x \simeq 0.05$ at most (corresponding to an external magnetic field $H \sim 1.5$ kGs; the magnetic energy associated with this upper limit is $\mu h \sim 10^{-7}$ eV). It may shift the frequency Ω of the quantal beats by 10%, at most. However, as shown below, the amplitude of the quantal beats decreases on increasing the beat frequency, which may render these oscillations unobservable. Under the usual working circumstances, corresponding to $x \simeq 0.014$ (for H = 500 Gs), this shift is about 3% at most. It is neglected in what follows.

The time-dependent part of the interaction with matter can mix up all the states $\tilde{\chi}_m$, in principle, where m = 0, 1, 2, 3. The transition probability is given by

$$w_{nm} = V_{nm}^2 \frac{4\sin^2[(\Delta E_{nm} - \hbar\omega)t/2\hbar]}{(\Delta E_{nm} - \hbar\omega)^2}, \tag{11}$$

where ω is the frequency of the local field h and V_{nm} are the matrix elements of the interaction. The only explicitly non-vanishing transition probabilities are, however, w_{01}, $w_{02,3}$ and $w_{12,3}$. For long times (11) gives a transition rate

$$g_{nm} \simeq \frac{2\pi}{\hbar^2}(\mu h)^2 \delta(\Delta E_{nm}/\hbar - \omega). \tag{12}$$

The energy levels of the positronium can be viewed as immersed in a continuum of excitations of the local magnetic interaction represented by the field h, so the transition rate becomes

$$g_{nm} \simeq \frac{2\pi}{\hbar\delta E}(\mu h)^2, \tag{13}$$

where $1/\delta E$ is the density of excitations; the energy δE may compare in magnitude with the uncertainty in the energy splitting ΔE_{nm}, which, in turn, is of the same order of magnitude as μh (the uncertainty in energy is of the order of $(\mu h)^2/\delta E$, which in turn is δE, so $\delta E \sim \mu h$); the local magnetization field in the sample smears out the Zeeman splitting of the hyperfine energy level of positronium, leading to a weak transition rate between them. Therefore,

$$g_{nm} \simeq 2\pi(\mu h)/\hbar \tag{14}$$

can be viewed as a generic transition rate between ortho- and parapositronium. For $H = 500$ Gs, $x = 0.014$ and from $h = xH$ at most, the transition rate given above is approximately 10^7–10^8 s^{-1}, *i.e.* a frequency that may compare with the rate constant of the 3γ-annihilation of orthopositronium. It corresponds to a magnetic field $h \sim 1$–10 Gs, which is the typical linewidth of the paramagnetic resonance in weak magnetic samples. However, for a higher external magnetic field H this rate may be enhanced up to cca 10^9 s^{-1}, which is the maximum Zeeman-splitting allowed by the detection limit at which the quantal beats can still be observed, in principle. It corresponds to a magnetic field $h \sim 75$ Gs. Such magnetic interactions can also be present in samples with complex magnetic properties, as it may be indicated by the linewidth of the paramagnetic resonance. It is worth noting that higher sample magnetic interactions can destroy completely the Zeeman splitting of the hyperfine energy levels of positronium, causing no observable effect anymore, 3γ-decay quantal beats included. A similar effect has higher external magnetic fields, with respect to the 3γ-detector resolution (10^9 s^{-1}) and counts statistics. It is also worth noting that an external field with the frequency in the range of $\Delta W/\hbar \sim 10^{12}$ s^{-1} (terahertz) can affect the transition rate between the

ortho- and parapositronium states, either directly, or by its coupling to the local internal field h (exciting, for instance, magnons in the sample), though a reasonable density of such an external field is difficult to be achieved yet.

A spin-flip transition rate between ortho- and parapositronium has been discussed recently in connection to the quenching rate of positronium in Xe, or Kr, as caused by the spin-orbit interaction [8]. Positronium spin-flip as caused by electron scattering at low energies has also been estimated [9]. Spin-flip scattering of positronium on Li [10] as well as on other matter atoms [11,12] has also been studied. The cross-section estimates for these processes are consistent with a transition rate in the range indicated above. Long time ago, Telegdi *et al.* [13] suggested an upper limit $g \simeq 10^8\ \mathrm{s}^{-1}$ for the mutual conversion rate of ortho- and parapositronium, at least in some amorphous insulators.

A FEW REMARKS UPON THE DENSITY MATRIX AND THE STATISTICAL MATRIX

Let $\psi(x) = \sum a_n \varphi_n(x)$ be a wavefunction expanded in some eigenfunctions φ_n. Then $a_n^* a_n$ is the probability of the state n, any average is given by $\sum f_{nm} a_n^* a_m$ for any quantity f, so that the matrix $\rho_{nm} = a_n^* a_m$ provides a full description, like $\psi(x)$ itself. In addition, $tr\rho = 1$ and $\rho^2 = \rho$. For a mixture not described by a wavefunction, the matrix ρ with these properties, *i.e.* $tr\rho = 1$ and $\rho^2 = \rho$, provides description for any average $tr(\rho f)$ and the probability ρ_{nn}. It is called the density matrix.

Similarly, a statistical average of the type $\sum \rho_n f_{nn}$, with $\sum \rho_n = 1$, gets $\sum \rho_{nm} f_{nm}$ under a change of states representation, so it leads to the statistical matrix ρ, corresponding to the statistical weigths $\rho_{nn} = \rho_n$, or (mean) populations of states. The statistical matrix ρ is such that $tr\rho = 1$ and the averages are given by $tr(\rho f)$. It is distinct from the density matrix, though often denoted by the same ρ. An instance is the statistical matrix $e^{-\beta E_n}/Z$, where β is the inverse of the temperature, E_n are eigenenergies, and Z is the canonical partition function. Another instance of a statistical matrix is the spin-polarization matrix, which for spin 1/2 is diagonal with elements $(1/2)(1 \pm p)$, where p denotes the polarization, or

$$\rho = (1/2)\begin{pmatrix} 1 + p\cos\theta & p\sin\theta e^{-i\varphi} \\ p\sin\theta e^{i\varphi} & 1 - p\cos\theta \end{pmatrix}, \tag{15}$$

when rotated by the polar angle θ and azimutal angle φ.

The electrons in matter are, usually, not polarized, so their spin-polarization matrix ρ_- is 1/2-diagonal. On the contrary, positrons are polarized, as described by the spin-polarization matrix ρ_+ given by (15). It follows that positronium is described by the spin-polarization matrix $\rho = \rho_-\rho_+$ over the electron and positron spin states. By making use of (1) and (5) it is a straightforward matter to get the spin-polarization matrix ρ_{nm} of positronium over the hyperfine χ-states and the spin-polarization matrix $\tilde{\rho}_{nm}$ over the Zeeman-splitted $\tilde{\chi}$-states introduced above. The matrix elements ρ_{nm} of the positronium spin-polarization matrix with respect to the hyperfine χ-states are given in **Appendix** 1. Similar results are obtained in the reverse case, where the positrons are not polarized, while the electons in matter are polarized.

The time-dependence of the spin-polarization matrix has two components, in general. One component is the stationary part, of the form $\sim \rho_{nm} e^{i(E_n - E_m)t/\hbar}$, governed by the hamiltonian, where E_n are the eigenenergies. The other component is the kinetic part, of the form $\sim \rho_{nm} e^{-(\gamma_n + \gamma_m)t/2}$, governed by kinetic equations, where γ_n are the damping rates of the states n. These rates are usualy small, in comparison with the observation time, and the kinetic part is usually not relevant in dynamics governed quantities. In addition, the kinetic part is irrelevant for computing transition rates, as the latter are precisely the corresponding damping rates. However, they govern the kinetics of populations.

3γ-ANNIHILATION OF ORTHOPOSITRONIUM

The 3γ-annihilation amplitudes $M_n = \langle 3\gamma | \chi_n \rangle$, n = 1, 2, 3 for the triplet states of (ortho) positronium are easily calculated by standard methods (this amplitude vanishes for the singlet state, $M_0 = 0$). [6] It is then easy to get these amplitudes for the Zeeman-splitted states $\tilde{\chi}_n$, n = 0, 1, 2, 3. They are given by

$$\widetilde{M}_0 = (x/2)M_1, \quad \widetilde{M}_1 = (1 - x^2/8)M_1, \quad \widetilde{M}_{2,3} = M_{2,3}. \tag{16}$$

The squared amplitude of 3γ-annihilation in magnetic field is given by

$$M^2 = \sum \tilde{\rho}_{nm} \widetilde{M}_n^* \widetilde{M}_m \, e^{i(E_n - E_m)t/\hbar}, \tag{17}$$

where summation over polarizations of the gamma quanta is included. First, it is useful to average over high frequencies in (17), as they are beyond the usual detection resolution. This way, only the frequency

$$\Omega = (E_{2,3} - E_1)/\hbar = x^2 \Delta W/4\hbar \tag{18}$$

is left in (17). Secondly, it is also useful to keep in (17) only the main contributions in magnetic field, *i.e.* zero-order contributions in parameter x (the decay amplitude has a significant dependence on strong magnetic fields). The squared amplitude reads then

$$M^2 = A + B\sin\Omega t, \tag{19}$$

where A is the squared amplitude of 3γ-annihilation in the absence of the magnetic field (corresponding to free orthopositronium), and B is the small amplitude of oscillations in magnetic field. The squared amplitude A does not depend on polarization p, in contrast to the coefficient B which is proportional to p. In addition, B is anisotropic, it depends on both the positronium polarization and the orientation of the plane of propagation of the three quanta of gamma radiation. The maximum value of the amplitude squared is reached for this plane rotated by $\pi/4$ about the magnetic field (z-axis) and parallel with the positronium polarization, directed along the x-axis $(\theta = \pi/2, \varphi = 0;$ in addition the angles between the gamma counters is 120°) [2]. For its maximum value the squared amplitude (properly normalized) reads

$$M^2 = \gamma_t[1 + (p/4)\sin\Omega t], \tag{20}$$

i.e. it is proportional to the rate constant γ_t of the 3γ-annihilation of free orthopositronium. The oscillating term in (20), corresponding to the quantal beats, is the contribution of the magnetic field. The geometric arrangement of the experimental setup [2] is shown in Fig. 2.

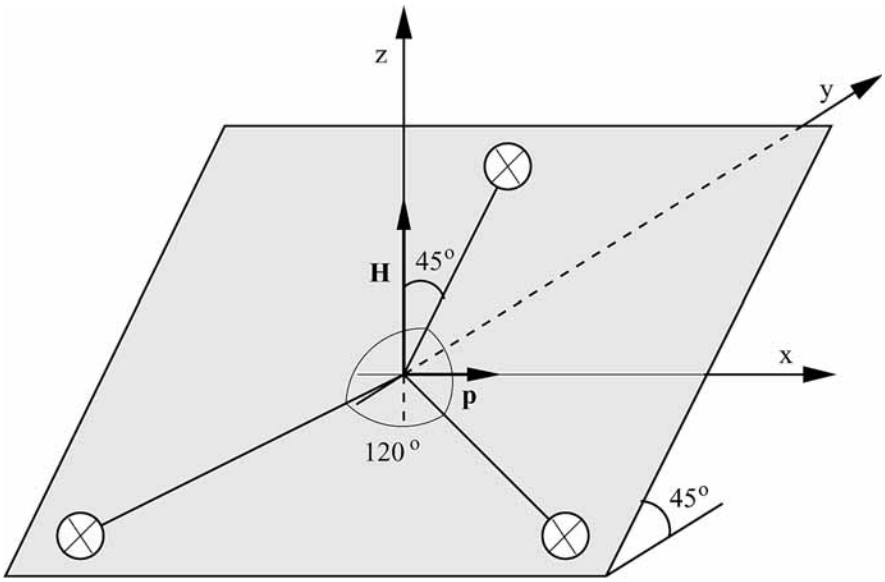

Fig. 2 – The geometric arrangement of the experimental setup as described [2].

Indeed, the number of (triple-coincidence) gamma counts N_t (*i.e.* the cross-section, properly normalized)) is proportional to the fraction $\rho_{11}+\rho_{22}+\rho_{33}=3/4$ of triplet population, which in the presence of the magnetic field conserves, *i.e.* $|(\tilde{\chi}_1,\chi_1)|^2\rho_{11}+|(\tilde{\chi}_0,\chi_1)|^2\rho_{11}+\rho_{22}+\rho_{33}=\rho_{11}+\rho_{22}+\rho_{33}$. It varies in time according to the kinetic equation

$$\frac{\partial N_t}{\partial t}=-\gamma_t N_t-(\gamma_t p/4)N_t\sin\Omega t, \tag{21}$$

whose solution is given by

$$N_t=N_{0t}e^{-\gamma_t t-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)}, \tag{22}$$

or, since $\gamma_t p/2\Omega\ll 1$,

$$N_t=N_{0t}e^{-\gamma_t t}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)], \tag{23}$$

which is seen experimentally [1–4]. A qualitative picture of such oscillations is shown in Fig. 3. The phenomenon of quantal beats in orthopositronium 3γ-annihilation has been predicted much earlier [14] and it was termed "positronium spin rotation" [1] by analogy with "muon spin rotation" [15]. Similar quantal beats appear also in other averages involving off-diagonal elements of the spin-polarization matrix, like, for instance, the positronium mean spin (hence the term "spin rotation").

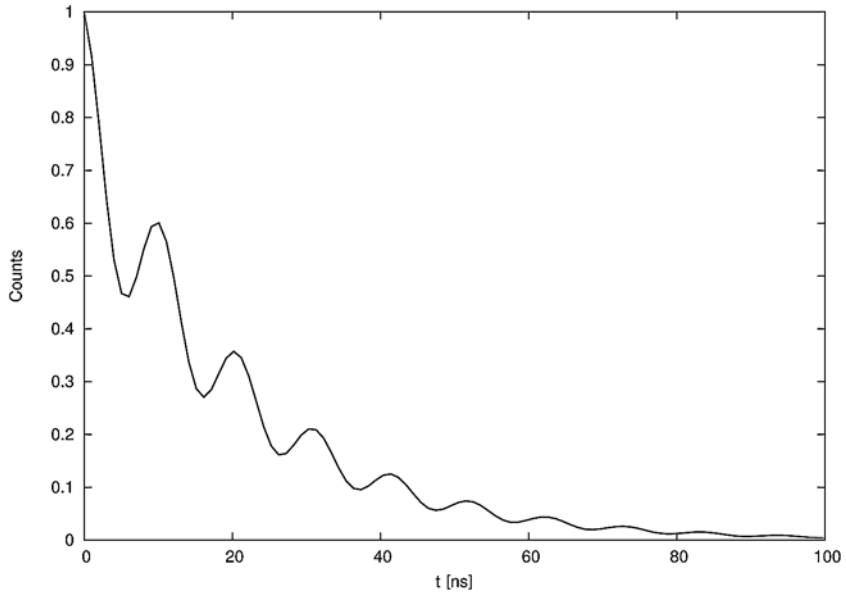

Fig. 3 – Qualitative picture of the quantal beats in the orthopositronium 3γ-annihilation in magnetic field.

It is worth noting that reversing the direction of the magnetic field amounts to reversing polarization in (23), which is exploited experimentally. In addition, the cross-section of one gamma out of three also oscillates in magnetic field.

2γ-ANNIHILATION OF PARAPOSITRONIUM

The 2γ-annihilation amplitude of positronium is associated exclusively to the singlet state (parapositronium), such that $M_0 \neq 0$ and $M_{1,2,3} = 0$ (gamma radiation of 511 keV). In magnetic field these amplitudes become $\widetilde{M}_0 = (1 - x^2/8)M_0$, $\widetilde{M}_1 = -(x/2)M_0$ and $\widetilde{M}_{2,3} = 0$. The 2γ-annihilation amplitude squared is given by an equation which is similar to (17). It is easy to see that quantal beats (whose amplitude is linear in the magnetic field) occurs for the frequency $\Omega_{01} \simeq \Delta W/\hbar$, which, however, is too high to be detected experimentally.

INDUCED QUANTAL BEATS OF THE PARAPOSITRONIUM 2γ-ANNIHILATION IN MATTER

The hyperfine states of positronium undergo quantal transitions in matter as a consequence of the local magnetic interactions. The transition rate between positronium triplet and singlet states can be represented as

$$g \simeq 2\pi(\mu h)/\hbar, \tag{24}$$

according to (14). Its magnitude should be ~ 10^7 s^{-1} at most, for an external field H = 500 Gs, or 10^9 s^{-1} at most (which is the detection limit), for an external magnetic field H = 1.5 kGs. Such a range of local magnetic interactions in matter corresponds to the linewidth of the paramagnetic resonance (an internal magnetic field h ~ 1–75 Gs). For higher magnetic interaction, or external fields, the quantal beats can be spoiled out, or they may lie outside the detector resolution. This transition rate is preserved in an external magnetic field which splits the hyperfine energy levels of positronium, providing the inequality $h \ll xH$ is satisfied, where H is the external magnetic field. The quantal beats in the ortopositronium 3γ-decay occurs at the fequency $\Omega = x^2\Delta W/4\hbar$, which amounts to ~ $6.5 \cdot 10^7$ s^{-1} for an external magnetic field H = 500 Gs. These beats are observable during the orthopositronium lifetime $\tau_t = 1/\gamma_t \simeq 10^{-7}$ s, by usual γ detectors with resolution 10^9 s^{-1}. The frequency of these oscillations may be enhanced by applying a higher magnetic field, of up to H = 1.5 kGs, such that the beat frequency reaches the detection limit 10^9 s^{-1}, in principle; although the amplitude of oscillations

decreases considerably in this case, up to the point of spoiling out the counts statistics. Since the transition rate must satisfy the inequality $g \ll \Omega$, one can set therefore an upper limit of cca 10^9 s^{-1} for this transition rate. In any case, the rate constant g is much smaller than the 2γ-annihilation rate $\gamma_s \simeq 10^{10}\,\mathrm{s}^{-1}$ of parapositronium (similarly with the 3γ-annihilation rate γ_t of orthopositronium, which satisfies the inequality $\gamma_t \ll \gamma_s$).

The kinetic equations for the triplet N_t and singlet N_s populations of positronium in matter and in an external magnetic field can therefore be written as

$$\begin{aligned} \partial N_t/\partial t &= -\gamma_t(t)N_t - gN_t + gN_s, \\ \partial N_s/\partial t &= -\gamma_s N_s - gN_s + gN_t, \end{aligned} \tag{25}$$

where $\gamma_t(t) = \gamma_t(1 + p/4\sin\Omega t)$ is the 3γ-annihilation rate of orthopositronium in magnetic field, quantal beats included, as given by (20). These equations are solved by making use of the small, perturbational parameter g, assuming $g \ll \gamma_t \ll \Omega \ll \gamma_s$. The solution can be represented as a series expansion in powers of g. Its first order terms are given in **Appendix 2**. The lowest-order solution reads

$$\begin{aligned} N_t &\simeq N_{0t}e^{-\gamma_t t}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)], \\ N_s &\simeq N_{0s}e^{-\gamma_s t} + N_{0t}\frac{g}{\gamma_s}e^{-\gamma_t t}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)]. \end{aligned} \tag{26}$$

One can see that the quantal beats produced by the magnetic field in the 3γ-annihilation rate of orthopositronium are induced, via the coupling g brought about by the local magnetic interactions in matter, into the 2γ-annihilation rate of parapositronium, with precisely the same frequency Ω. Moreover, while the free parapositronium 2γ-decay cannot be seen (since it is too fast for the detector resolution), the induced oscillations can be seen over the orthopositronium lifetime γ_t^{-1} as given by

$$N_s \simeq N_{0t}\frac{g}{\gamma_s}e^{-\gamma_t t}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)]. \tag{27}$$

Their amplitude is smaller than the amplitude of the 3γ-rate oscillations, by the ratio

$$N_s^{oscill}/N_t^{oscill} \simeq g/\gamma_s\,. \tag{28}$$

For $g \sim 10^7$ s^{-1} at most, as estimated above for weak magnetic matter, the ratio given by (28) is 10^{-3}.

For transition rate g of the same order of magnitude as γ_t, or greater, the solution reads

$$N_s \simeq N_{0s} e^{-\gamma_s t} + N_{0t} \frac{g}{\gamma_s} e^{-(\gamma_t + g)t} [1 - (\gamma_t p/2\Omega) \sin^2(\Omega t/2)], \tag{29}$$

(for $g,\ \gamma_t \ll \Omega \ll \gamma_s$), *i.e.* the triplet damping rate γ_t is strongly renormalized, (to the limit of detection resolution), but the ratio of the two oscillations amplitudes is left unchanged, as given by (28). In the most favourable case $g \sim 10^9$ s^{-1}, as estimated above, this ratio is about 1/10. The solution of (25) in this case is sketched in **Appendix 3**.

EXPERIMENTAL

Positrons are produced usually by radioactive ^{22}Na with energy 1–2 eV, corresponding to a velocity ~ $5 \cdot 10^7$ cm/s, and polarization $p = 0.7$. A small aperture (of approximatelly 2π) collects the positrons flow and average their polarization to $p \sim 0.25$ and velocity to $v \sim 10^7$ cm/s. Typically, the rate of emission is 10^4 s^{-1} (*i.e.* 10^4 Bq, or 0.27 μCi activity) [4]. Positrons are injected in noble gases, usually, with a flow ~ 10^3 cm^{-2}s^{-1}, where they experience direct annihilation, inelastic and elastic collisions, approach thermalization, and form ortho- and parapositronium. The latter suffer their own 3γ- and 2γ-annihilation processes, respectively, including the pickoff annihilation by electrons in atoms. The collisions of positronium by matter atoms can be represented generically by a cross-section $\sigma \sim 10^{-16}$ cm^2, so the colliding rate $vn\sigma \sim 10^{11}$ s^{-1}, for a gas density $n \sim 10^{20}$ cm^{-3} (mean-free path ~ 10^{-4} cm). The relativistic effects, which are responsible for spin-flip interactions, are typically effective by a Z^{-2} factor, where Z is the atomic number, so a reduction factor of 10^4 applied to the rate estimated above is reasonable. The spin-flip rate of positronium can therefore be estimated, very qualitatively, as $g \sim 10^7$ s^{-1}, which agrees with other estimates [8–13].

Measurements in stationary flux imply a continuously feeding of positrons at rate Γ and a decay at a generical rate γ, so the equation $\partial N/\partial t = \Gamma - \gamma N$ has solution $N = (\Gamma/\gamma)(1 - e^{-\gamma t})$; in short time the stationary population is Γ/γ and yield Γ is collected at a rate γ, *i.e.* Γ counts per second (actually $\Gamma[1 - (\gamma_t p/2\Omega)\sin^2(\Omega t/2)]$ for oscillations explicitly included). For a sample volume of 1 cm^3, the rate $\Gamma \sim 10^3$ s^{-1} is estimated above. However, a reduction factor of about 10 must be allowed for the fraction of positronium formation, and another reduction by a factor of ~ 100 comes probably from the counters. It follows that a yield of about $10^{-3}\Gamma \sim 1$ is collected in fact, per second. In order to get an accuracy of ~ 1%, a total of ~ 10^4 counts is needed, which gives about 10^4 s of running the measurements, *i.e.* cca 3 hours. However, the oscillations amplitude

$\gamma_p p/2\Omega$ is itself about 1% of the signal, so, in order to get 1% accuracy in oscillations amplitude, another factor 100 is needed, which raises the running time at about 300 hours (cca a couple of weeks).

Identifying, in the same conditions, the small oscillations induced in the parapositronium 2γ-annihilation would require another factor of 10 to 1000; it may be achieved by increasing correspondingly the ^{22}Na-source activity, the feeding flux, by multiplying the counters, or by prolonging the detection time.

Usually, measurements are performed on the decay tail governed by laws like $\sim e^{-\gamma t}$ (delayed coincidence), where the starting time is the emission of one positron from the source (accompanied by gamma radiation), and subsequent decay gamma quanta are recorded during the positronium lifetime. Such measurements imply a similar statistics and running time as those corresponding to a stationary flux.

CONCLUSION

In conclusion, an ortho-parapositronium transition rate g that may originate in the local magnetic interactions in matter can transfer the quantal beats observed in the orthopositronium 3γ-annihilation in magnetic field onto the parapositronium 2γ-annihilation (511 keV gamma radiation). These induced oscillations have the same frequency as the 3γ-annihilation rate oscillations, occur over the same time as the orthopositronium lifetime, but have a much smaller amplitude in comparison with the 3γ-annihilation rate amplitude, in the ratio g/γ_s, where $\gamma_s \simeq 10^{10}$ s^{-1} is the rate constant of the parapositromium 2γ-decay. Qualitative estimations indicate values in the range 1/10 to 1/1000 for this ratio, which impose severe restrictions on the experimental observation of these induced quantal beats. However, providing such experimental constraints are overpassed, these induced beats may give valuable information on probing the local magnetic interactions in matter, suggesting a new method of experimental investigation which may be termed "positronium spin rotation" by analogy with "muon spin rotation". At the same time, the transition rate g may shift the rate constant γ_t of the orthopositronium 3γ-decay into $\gamma_t + g$, and the component constant in time of the magnetic field associated with the local magnetic interactions may produce a shift in the beats frequency of a maximum relative value $2x$.

Acknowledgments. One of the author (MA) is deeply indebted to E. Ivanov, I. Vata, D. Dudu and the team of the Cyclotron Accelerator at Magurele-Bucharest for bringing this problem to his attention and for many valuable discussions.

Appendix 1
Matrix elements of the spin-polarization matrix

$$\begin{aligned}
&(\chi_0, \rho\chi_0) = 1/4 \\
&(\chi_0, \rho\chi_1) = (1/4)p\cos\theta \\
&(\chi_0, \rho\chi_2) = -(1/4\sqrt{2})pe^{i\varphi}\sin\theta \\
&(\chi_0, \rho\chi_3) = (1/4\sqrt{2})pe^{-i\varphi}\sin\theta
\end{aligned} \tag{30}$$

$$\begin{aligned}
&(\chi_1, \rho\chi_1) = 1/4 \\
&(\chi_1, \rho\chi_2) = (1/4\sqrt{2})pe^{i\varphi}\sin\theta \\
&(\chi_1, \rho\chi_3) = (1/4\sqrt{2})pe^{-i\varphi}\sin\theta
\end{aligned} \tag{31}$$

$$\begin{aligned}
&(\chi_2, \rho\chi_2) = (1/4)(1 + p\cos\theta) \\
&(\chi_2, \rho\chi_3) = 0
\end{aligned} \tag{32}$$

$$(\chi_3, \rho\chi_3) = (1/4)(1 - p\cos\theta) \tag{33}$$

The matrix ρ is a symmetrical matrix, $\rho_{mn} = \rho_{nm}$.

Appendix 2
Perturbational solution of eq. (25)

For small values of the transition rate g its contribution to the diagonal terms of equations (25) can be neglected. The solution of these equations can be written as a series expansion in powers of g, of the form

$$\begin{aligned}
N_t &= N_{to} + gN_{t1} + g^2N_{t2}\ldots, \\
N_s &= N_{so} + gN_{s1} + g^2N_{s2},
\end{aligned} \tag{34}$$

where $N_{t,s0,1,2,\ldots}$ satisfy equations

$$\begin{aligned}
\partial N_{t0}/\partial t &= -\gamma_t(t)N_{t0}, \\
\partial N_{t1}/\partial t &= -\gamma_t(t)N_{t1} + N_{s0}, \\
N_{t2}/\partial t &= -\gamma_t(t)N_{t2} + N_{s1},
\end{aligned} \tag{35}$$

and, respectively,

$$\begin{aligned}
\partial N_{s0}/\partial t &= -\gamma_s N_{s0}, \\
\partial N_{s1}/\partial t &= -\gamma_s N_{s1} + N_{t0}, \\
\partial N_{s2}/\partial t &= -\gamma_s N_{s2} + N_{t1}.
\end{aligned} \tag{36}$$

The solution is obtained straightforwardly as

$$N_{t0} = N_{0t} e^{-\gamma_t t}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)], \tag{37}$$

as given by (23), $N_{s0} = N_{0s} e^{-\gamma_s t}$, and

$$N_{s1} = N_{0t} e^{-\gamma_s t}\int_0^t dt' \cdot e^{\Delta\gamma t'}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t'/2)], \tag{38}$$

where $\Delta\gamma = \gamma_s - \gamma_t$. Higher-order terms in g, included N_{t1}, are not relevant, so they are not computed here. The integral in (38) is performed straightforwardly, leading to

$$\begin{aligned} N_{s1} = N_{0t} e^{-\gamma_s t}\Big\{&\frac{1}{\Delta\gamma}(1-\gamma_t p/4\Omega)(e^{\Delta\gamma t}-1)+ \\ &+\frac{\gamma_t p/4\Omega}{(\Delta\gamma)^2+\Omega^2}[\Delta\gamma(e^{\Delta\gamma t}\cos\Omega t-1)+\Omega e^{\Delta\gamma t}\sin\Omega t]\Big\}. \end{aligned} \tag{39}$$

Since $\gamma_t \ll \Omega \ll \gamma_s$ it simplifies to

$$N_{s1} \simeq N_{0t}\frac{1}{\gamma_s} e^{-\gamma_t t}[1-(\gamma_t p/2\Omega)\sin^2(\Omega t/2)], \tag{40}$$

which leads to (27). The constants $N_{0t,s}$ are the populations of ortho- and, respectively, parapositronium at time zero.

Appendix 3
Solution of eq. (25)

In matricial notation equations (25) can be written as

$$\partial N/\partial t = A(t)N, \tag{41}$$

whose solution is given by

$$N(t) = \exp\left[\int_0^t dt' \cdot A(t')\right] N_0, \tag{42}$$

where the matrix

$$Bt = \int_0^t dt' \cdot A(t') = \begin{pmatrix} -a & g \\ g & -b \end{pmatrix} t \tag{43}$$

is given by

$$\begin{aligned} a &= \gamma_t + g + \varepsilon(t), \\ b &= \gamma_s + g, \\ \varepsilon(t) &= (\gamma_t p/2\Omega t)\sin^2(\Omega t/2). \end{aligned} \tag{44}$$

The matrix B is diagonalized by

$$C = \begin{pmatrix} 1-\alpha^2/2 & -\alpha \\ \alpha & 1-\alpha^2/2 \end{pmatrix}, \tag{45}$$

$C^{-1}BC = \Lambda$, where $\alpha = g/(b-a)$ and the eigenvalues in Λ are given by

$$\lambda_1 = -a + g^2/(b-a), \quad \lambda_2 = -b - g^2/(b-a) \tag{46}$$

up to contribution of the order $g/(b-a) \ll 1$.

It follows straightfowardly the solution

$$N_t \simeq N_{0t} e^{-(\gamma_t + g)t} [1 - (\gamma_t p/2\Omega)\sin^2(\Omega t/2)], \tag{47}$$

as given by (23), except for the renormalization $\gamma_t \to \gamma_t + g$, and

$$N_s \simeq N_{0s} e^{-\gamma_s t} + N_{0t}(g/\gamma_s) e^{-(\gamma_t + g)t} [1 - (\gamma_t p/2\Omega)\sin^2(\Omega t/2)] \tag{48}$$

as indicated in (29).

REFERENCES

1. V. G. Baryshevsky, O. N. Metelitsa, V. V. Tikhomirov, S. K. Andrukhovich, A. V. Berestov, B. A. Martsinkevich, E. A. Rudak, Phys. Lett., **A136**, 428 (1989).
2. V. G. Baryshevsky, O. N. Metelitsa, V. V. Tikhomirov, J. Phys., **B22**, 2835 (1989).
3. S. Fan, C. D. Beling, S. Fung, Phys. Lett., **A216**, 129 (1996).
4. E. Ivanov, I. Vata, D. Dudu, I. Rusan, N. Stefan, *Applied Surface Science* (in print).
5. E. Ivanov, private communication.
6. V. B. Berestetsky, E. M. Lifshitz, L. P. Pitaevsky, *Quantum Electrodynamics*, *Landau and Lifshitz Course of Theoretical Physics*, 2nd ed, NY, Pergamon 1982.
7. B. I. Goldansky, *The Physical Chemistry of Positron and Positronium*, Moscow, 1969.
8. J. Mitroy, S. A. Novikov, Phys. Rev. Lett., **90**, 183202-1 (2003).
9. V. S. Melezhik, F. R. Vukaylovic, Phys. Rev. Lett., **59**, 641 (1987).
10. M. Mukherjee, A. S. Ghosh, Phys. Rev., **A46**, 2558 (1992).
11. S. Mrowczynski, Phys. Rev., **A33**, 1549 (1986); Phys. Rev., **D36**, 1520 (1987).
12. K. G. Denisenko, S. Mrowczynski, Phys. Rev., **D36**, 1529 (1987).
13. V. L. Telegdi. J. C. Sens, D. D. Yovanovitch, S. D. Warshaw, Phys. Rev., **104**, 867 (1956).
14. V. G. Baryshevsky, Dokl. Akad. Nauk BSSR, **16**, 445 (1966); Phys. Stat. Sol., **b124**, 619 (1984).
15. S. F. J. Cox, J. Phys., **C20**, 3187 (1987).