Enhanced Ps-Ps Interactions due to Quantum Confinement

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Enhanced Ps-Ps interactions due to quantum confinement

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Slow positrons implanted into a porous silica film may efficiently form positronium (Ps) atoms that diffuse through a network of interconnected pores. At high Ps densities, the long lifetime of ortho-positronium atoms is reduced due to Ps-Ps spin dependent interactions at a rate that implies an effective free-space scattering cross section, $\sigma_e = (3.4 \pm 0.5)\times 10^{-14}$ cm^{-2}, at least 25 times larger than the theoretical value. This enhanced interaction rate may be explained if the quantum confinement of Ps results in inter-pore tunnelling rates that depend critically on the distribution of pore sizes, so that rather than uniformly sampling the porous matrix Ps diffusion is limited to a small subset of the pores.

Although positronium (Ps) scattering with atoms and molecules may be successfully accomplished using beam techniques [1], the density of Ps beams is presently too low to observe Ps-Ps scattering in this way [2]. However, interactions between Ps atoms may be detected by creating a "gas" of Ps in a suitable container, and then measuring scattering via mechanisms that affect the Ps decay rate [3]. The cross section, σ_0, for this type of Ps-Ps scattering has been calculated by Ivanov, Mitroy and Varga using the stochastic variational method [4], and is therefore quite accurately known. Our previous attempts to measure this cross section [5, 6] have consistently yielded values at least an order of magnitude higher than this calculation would suggest. However, this discrepancy was not thought to be indicative of anything other than the inaccuracy of our measurements. This was in part due to uncertainties in the Ps thermalization rate and a lack of information concerning Ps diffusion in the porous matrix used to contain the atoms.

We report here new measurements made using a porous film similar to those previously used, except that there is no capping layer so that the Ps is able to escape from the sample into vacuum. This allows us to directly measure the Ps cooling rate in and emission time from the sample [7, 8], and to measure the Ps-Ps interaction rate for different effective Ps densities. Taken together, all of these measurements make it possible for us to determine the effective Ps-Ps scattering cross section, σ_e, with much higher accuracy than was previously possible. We find that not only does the discrepancy persist, but that the effective cross section we measure is in fact even larger than was implied by our prior experiments, being about 25 times higher than the calculated value. We suggest that this is due to effects associated with the quantum confinement [9] of Ps in a porous film, primarily an enhanced effective Ps density due to variations in the tunnelling rates between pores of different sizes restricting the atoms to a small subset of the pores.

The experiments were conducted using a positron accumulator [10] that is able to produce ~ 1 ns wide positron pulses with a central areal density n_{2D} of up to 1 × 10^{11} cm^{-2}. The density of the positron beam may be precisely controlled via the frequency of a rotating wall electric field [11], applied to the positrons while they are in the accumulator [12]. The positrons were implanted into a ~ 600 nm thick film of porous silica with pore sizes of around 5 nm diameter [13], as verified by the shift of the Lyman-alpha frequency for Ps atoms within the pores [14]. The decay rate of Ps atoms subsequently produced was measured using the technique of single shot positron annihilation spectroscopy [15]. This allows us to generate lifetime spectra, from which we determine the "delayed fraction" parameter f_d, defined as the fraction of the total spectrum in the interval 50-300 ns after the prompt peak [7]. This parameter is essentially proportional to the fraction of incident positrons that form long lived positronium.

The basis of our measurement of Ps-Ps scattering is the fact that when two oppositely spin polarised triplet Ps atoms interact with each other, they may undergo a process referred to as spin exchange quenching (SEQ) [16, 17] in the following way [5]:

$$Ps_{m=1} + Ps_{m=-1} \rightarrow 2(Ps_{m=0} + E_{hfs})\,. \qquad (1)$$

Here m is the magnetic quantum number of Ps and E_{hfs} is the hyperfine energy difference between the incoming and outgoing states [18]. In the quenching process, long lived triplet atoms, which have parallel spins and a mean lifetime of 142 ns, are converted to short lived singlet and triplet $m = 0$ atoms, which have anti-parallel spins and decay in a few ns in our 2.3 T magnetic field [19]. The cross section for the two processes described in Eq. 1 in the zero velocity limit is $\sigma_{SEQ} = 2\sigma_0 = 1.28\times 10^{-15}\,\text{cm}^2$ (since the cross sections for scattering into pairs of singlet or triplet m = 0 atoms are the same [4]). The large difference between the decay rates of the m=0 and $|m|$=1 states means that such interactions may be detected via changes in (single shot) lifetime spectra.

When positrons are implanted into our porous film the resulting instantaneous Ps density will depend on the incident positron beam areal density as well as the mean implantation depth, which is given by [20] $\bar{z} = A\rho / K^{\nu}$, where K is the beam energy in keV, $\nu \approx 1.7$, A=2.81 μg/cm^2 [21], and ρ=1.35

g/cm^3 is the average target density. Following positron implantation into the film, Ps atoms will form quickly (within ~ 10 ps) and then diffuse in the bulk material until they either annihilate or are emitted into the voids with ~ 1 eV kinetic energy [22]. If they are created near the surface they may then rapidly escape from the sample. On the other hand, Ps atoms created deeper in the sample will cool by collisions with the pore walls until they can only move between pores by tunnelling. These atoms are far more likely to encounter other Ps atoms as they spend much more time in the sample.

Using single shot lifetime and laser spectroscopy of atoms emitted from a sample nominally identical to that used here, we have measured Ps thermalization as a function of the incident beam energy [7] as well as the Ps emission time and diffusion coefficient [8]. These data all support the idea that hot Ps will initially diffuse rapidly and escape from the sample, but may cool down and remain inside for longer if implanted deep enough. We can also see evidence for this in the energy dependence of Ps-Ps scattering, as shown in fig 1, which shows the quenching parameter Q for different positron beam areal densities and implantation energies. This parameter is defined as

$$Q(n_{2D}^0) \equiv f_d(n_{2D}^0)/f_d(0), \tag{2}$$

where n_{2D}^0 is the central areal density of the $|m|$=1 Ps just after implantation of the positron pulse into the sample. The slopes of the curves of fig 1 indicate the extent to which Ps-Ps interactions occur. It is evident that there is almost no quenching effect at all if the beam is implanted at 1.17 keV. This is hardly surprising since at this implantation energy the mean emission time is less than the ~ 1 ns positron beam width. Thus, Ps atoms leave the sample even before others have been created, and their effective density is greatly reduced. As the beam energy is increased and Ps atoms spend more time in the sample, the amount of quenching increases.

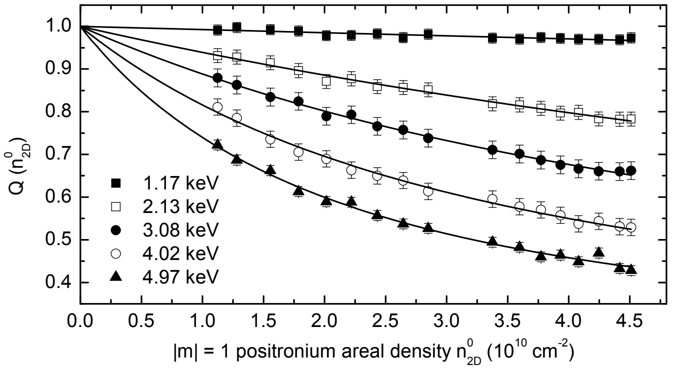

Fig 1. Normalized Ps quenching data for various positron implantation energies, with fits of eq (4) as described in the text. Since the polarization p_0 cannot be determined from quenching data that does not have any degree of saturation, p_0 = (31 ± 3) %, as determined from the highest beam energy data, was used as a non-variable parameter in all of the other fits.

Because the primary positron beam is partially spin polarized we have an asymmetric population of Ps spin states. This leads to a saturation of the quenching effect at high density when the minority spin states become depleted [19]. The Ps decay is described by a pair of coupled equations:

$$dn_{\uparrow(\downarrow)}/dt = -\gamma n_{\uparrow(\downarrow)} - \gamma\beta n_{\uparrow} n_{\downarrow}\,. \tag{3}$$

Here $n_{\uparrow(\downarrow)}$ refers to the spin up (down) triplet $|m|$=1 Ps atom number density (smoothed over the ~ 10 nm-scale density fluctuations associated with the porous sample), γ is the Ps decay rate in the sample (0.031 ns^{-1} [7]), $\beta\gamma$ is the spin exchange quenching rate per unit density of opposite spin $|m|$=1 Ps atoms, and β is an effective spin exchange interaction volume. The solution to eq. (3) may be written in terms of the quenching parameter Q in the following way [19],

$$Q(\zeta) = p_0[1 + p_0 \tanh\{\tfrac{1}{2} p_0\zeta\}]/[p_0 + \tanh\{\tfrac{1}{2} p_0\zeta\}]\,. \tag{4}$$

Here p_0 is the Ps polarisation (which we assume is the same as that of the positron beam) and the effective density is defined as $\zeta \equiv \beta n_0[1 - \exp\{-\gamma t_e\}]$, with a mean density n_0 and a mean time for Ps to leave the sample t_e [8]. We fit the data in Fig. 1 using the function $Q(\zeta)$ to determine the initial slopes $\partial Q/\partial n_{2D}^0$ from which we will deduce the spin exchange interaction volume β and obtain our estimate of the effective free particle Ps-Ps cross section from the proportionality $\beta\gamma \propto \sigma_e \bar{v}$, where $\bar{v}$ is the average thermal speed of the Ps atoms.

By considering the radial distribution and implantation depth of the positron beam, we find that the initial density of $|m| = 1$ positronium atoms averaged over the density is given by

$$\left\langle n_{3D}^0 \right\rangle = \frac{\pi n_{2D}^0}{4\sqrt{2}\bar{z}}\,. \tag{5}$$

Then it may be shown [23] that

$$\beta = -\frac{8\sqrt{2}\bar{z}}{\pi\gamma(1-p_0^2)} \frac{\partial Q}{\partial n_{2D}^0} \frac{\exp\{-2\gamma t_I\}}{G(t_I)}\,. \tag{6}$$

where t_I is an effective Ps thermalization time and G(t_I) is a function that represents the normalised SEQ yield [23].

Figure 2 shows values of β calculated from eq. 6 using different choices for the (unknown) thermalization times t_I for energies above 3 keV. For completeness we also show data obtained from eq. (6) using the two lowest energy points, even though we do not expect them to be valid due to the fast Ps emission times associated with these energies (~ 50 and 400 ps at 1 and 2 keV respectively) [24]. As is indicated by the dashed line in Fig 2, the optimal choice of t_I that makes β

independent of the positron implantation energy K is $t_I \approx (2.5 \pm 0.7)\,\mathrm{ns}$ for K=3, 4, and 5 keV, with an error estimate obtained from the chi square distribution for fits of horizontal lines to the data of fig 2. Assuming that K-independence of β indicates the correct value of t_I, we then have $\beta = (4.7 \pm 0.7) \times 10^{-14}\ \mathrm{cm}^3$. In evaluating Eq. (6) we have used the positron central density determined from the rotating wall frequency [25] and $f_{Ps}(|m|=1) = 0.40 \pm 0.05$; $\partial Q / \partial n_{2D}^0$ is determined from the initial slopes of the curves in Fig. 1; and $G(t_I)$ by numerical integration [23].

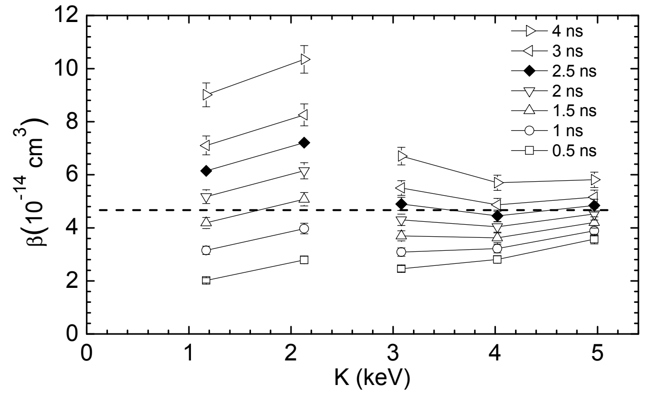

Fig 2. Determinations of β from fits shown in fig 1 and from Eq. 6 using various choices for the effective thermalization times t_I shown in the legend.

We may interpret our value of β in terms of free Ps atoms scattering from each other in the available effective free volume of the sample at thermal velocities and quenching at a rate

$$\Gamma = \tfrac{1}{2}(1 - p_0^2)\left\langle n_{3D}^0 \right\rangle \varphi p^{-1} \sigma_e \bar{v}_{th} = \tfrac{1}{2}(1 - p_0^2)\beta\gamma\left\langle n_{3D}^0 \right\rangle, \qquad (8)$$

where $p \sim 0.5$ is the sample porosity and φ is a density enhancement factor that takes into account Ps localization within the pores in the following way; the normalized ground state wave function of a particle in a spherical cavity of radius a is an L=0 spherical Bessel function $\psi(r) = \frac{1}{\sqrt{2\pi a}} \frac{1}{r} \sin\{\pi r / a\}$. The density averaged density, needed for finding two-particle interaction rates, is $\left\langle \psi^2 \right\rangle = \frac{1}{\pi a^2} \int_0^a \frac{1}{r^2} \sin^4\{\pi r / a\} dr$, and the density enhancement factor is thus $\varphi = \frac{4}{3}\pi a^3 \left\langle \psi^2 \right\rangle \approx 2.8$. We can now obtain our cross section, $\sigma_e = \varphi^{-1} p \beta\gamma / \bar{v}_{th} = (3.4 \pm 0.5) \times 10^{-14}\ \mathrm{cm}^2$, which is 25 times larger than the expected zero velocity limit σ_{SEQ} [4]. It may be that scattering into two singlet atoms is suppressed in the confined pore geometry due to the relatively large hyperfine energy difference (~ 2 meV for the singlet and only 0.4 meV for the triplet channel). In this case $\sigma_{\mathrm{SEQ}} = \sigma_0$ and our effective cross section σ_e would then be 50 times larger than σ_{SEQ}.

The free space cross section implies the interaction rate for 2 Ps atoms in a single pore would be $\left\langle \psi^2 \right\rangle \sigma_{SEQ} \bar{v}_{th} \approx (2.5\,\mathrm{ps})^{-1}$ [26]. At this rate the dwell time in one pore for tunnelling from pore to pore in three dimensions, $\tau \approx \lambda^2 / 6D_0 \approx 0.9\,\mathrm{ps}$, would imply only a ~ 35% probability of quenching per 2 particle encounter. However, the cross section may be an unrealistic description of the interactions of two quantum confined particles for which the overlap of the wavefunctions will undoubtedly lead to significant correlation effects. In principle the interaction rate for such particles could be up to three times higher than is implied by the free space cross section (i.e., unit probability of quenching for two |m| = 1 atoms in the same pore), in which case the anomalous quenching cross section would then be ~ 10 rather than 25 times too large. This remaining discrepancy can be explained by considering the dynamics of Ps diffusion between pores. Since quantum confined Ps atoms move primarily via tunnelling [7], the subset of pores that can be visited will depend critically on the tunnelling rates between pores, and hence on their individual dimensions. If there is a distribution of characteristic geometries, the Ps may preferentially travel through a sub-set (e.g ~ 10%) of nearly identical pores at the mean of the distribution [27], which will lead to a concomitant increase in the effective Ps density.

Since the tunnelling rate between pores depends exponentially on the effective energy barrier between them, even small variations in the pore geometry could lead to the preferential selection of only a small fraction of the total pore volume. The quantitative details of any such mechanism will depend strongly on the real sample structure, in particular on the nature of the interconnecting paths between pores and not just their radii [7].

We conclude by remarking that the enhanced Ps-Ps interaction rate inside porous materials is rather serendipitous from the point of view of our "many positron" experiments [28], as it has made it much easier to observe Ps-Ps interactions. Unlike ordinary atoms, Ps can easily be produced in porous structures, whether the geometry is open or closed. This provides a convenient way to study the physics of both interacting [5, 19] and confined atoms [7, 14]. Although Ps formation in porous materials has been extensively studied [E.g., 13, 29] this has previously always been in the single particle limit, usually with the tacit assumption that the pores are uniformly sampled; the ability to produce interacting Ps in a porous medium has shown that this assumption is not correct. It would be instructive to perform similar measurements to those reported here using a large range of pore sizes. For very large pores, for which there will be no quantum confinement at all, it may be possible to measure cross sections approaching the free space limit, although in this case the effective scattering rates would of course be much lower.

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