

# Chemistry and physics with many positrons

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## Abstract

The first study of a high-density collection of positronium (Cassidy et al., 2005) revealed Ps–Ps spin exchange interactions in a porous silica sample. The prospects for further work in this field include forming di-positronium molecules and the Ps Bose–Einstein condensate, and constructing an annihilation gamma ray laser.

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## 1. Introduction

Experiments with many positrons require a suitable source of slow positrons (Mills, 1995c), a means for accumulating and storing positrons (Greaves and Surko, 2000), and devices for the compression of large bursts of positrons in time (Mills, 1980a; Chu et al., 1981) and space (Mills, 1980b; Canter, 1985; Greaves and Surko, 2000). Various ways of achieving these requirements have been available for many years and have recently been combined into a single apparatus for the purpose of studying Ps–Ps interactions (Cassidy et al., 2005). We now have sufficient positron densities to make Ps<sub>2</sub> molecules and need only a few improvements of our apparatus to make a Ps Bose–Einstein condensate (Platzman and Mills, 1994). Thanks to the invention of positron trapping and accumulation, the experiments so far could be done using a relatively small (20 mCi) <sup>22</sup>Na source.

An experiment that would demonstrate the stimulated annihilation necessary for a gamma ray laser would require a much stronger positron source, with <sup>13</sup>N appearing to be the best candidate (Mills, 1995a; Cassidy et al., 2002) since its potentially high specific activity should yield a slow positron beam many orders of magnitude brighter than the only alternatives, namely

positrons obtained from pair production, either from Bremsstrahlung radiation (Howell et al., 1983; Cassidy et al., 2001; Csonka, 2003) or from gamma rays (Hugenschmidt et al., 2004). Even if a positron source is not intended for studying many-positron interactions, one will inevitably wish to study small samples, which will require some form of phase-space compression to avoid a large loss of beam intensity caused by narrowing a beam with an aperture. The metric for judging the effectiveness of a slow positron source for most experiments is not the intensity in slow positrons per second. Neither is it simply the intensity divided by the transverse phase-space volume occupied by the beam because this quantity can be increased by some form of phase-space compression accomplished either via remoderation or the rotating wall effect. A figure of merit for a slow positron source could be defined (Canter, 1985) as the intensity it would have after being focused to the smallest possible diameter at a standard beam energy (say, 10 keV) and apertured by a standard diameter hole (say, 10 μm) placed at the focal point. A crude estimate for the merit of a positron source would be a quantity  $\Sigma$ , defined as the initial slow positron source intensity divided by its diameter, since one may reduce the beam diameter by multiple stages of remoderation, acceleration and focusing, in each of which both the beam diameter and intensity decrease by about an order of magnitude. By this metric a 50 kW deuteron accelerator

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(5 MeV, 10 mA) yielding a 30 Ci  $^{13}\text{N}$  source 0.1 mm diameter with a 1% efficient solid Ne moderator producing  $10^{10}$  slow positrons per second would have  $\Sigma = 10^{12}$  slow  $e^+$ /cm, whereas a 50 kW electron LINAC or a gamma ray pair production source producing the same number of slow positrons per second from a 1 cm diameter moderator would have a 100 times smaller value of  $\Sigma$ .

## 2. Positronium–positronium (Ps–Ps) interactions

At the time of the 8th International Conference on positron and positronium chemistry (PPC-8) at Coimbra, Portugal for which this paper was prepared, Cassidy et al. (2005) had just obtained evidence for spin exchange quenching of Ps atoms at high densities. Fig. 1 shows a set of three lifetime curves, each obtained using a single pulse of  $1.4 \times 10^7$  positrons with a full-width at half-maximum time-spread of 20 ns. The positrons are either compressed using the rotating wall effect or allowed to expand for a few seconds before being released from the accumulator. The positron pulse is further compressed in a pulsed 1 T magnetic field to a beam of Gaussian cross-section having a minimum (“compressed”) full-width at half-maximum diameter of 0.2 mm corresponding to a central peak areal density of  $5 \times 10^{10}$  positrons per  $\text{cm}^2$ . The positrons were implanted into an ordered porous silica sample 250 nm thick on a silicon substrate in ultra-high vacuum. The  $|m| = 1$  ortho-positronium (*o*-Ps) lifetime measured using the expanded beam is fitted by a single component with decay time  $36.4 \pm 0.1$  ns with an intensity of  $24.3 \pm 0.2\%$ . The lifetime data obtained using a compressed pulse cannot be fitted using a single lifetime decay. The best fit is obtained keeping the *o*-Ps lifetime

fixed at 36.4 ns and assuming the positron polarization is 35% and that there are spin exchange collisions occurring at a rate that is proportional to the Ps density.

At low densities, the  $|m| = 1$  *o*-Ps decays at a rate  $\gamma$  that is the sum of the self-annihilation rate into three photons and the pick-off annihilation rate (predominately into two photons) due to collisions of the *o*-Ps with electrons of the surrounding medium. At high Ps densities, there will be spin-exchanging collisions between pairs of oppositely polarized  $|m| = 1$  *o*-Ps atoms. So long as the Ps kinetic energy is small compared to the binding energy  $E_M = 0.435445$  eV of the  $\text{Ps}_2$  molecule (Hylleraas and Ore, 1947; Ho, 1986; Frolov and Smith, 1997), spin exchange will be determined by the singlet channel (total spin 0) Ps–Ps scattering length  $a_0 = 0.395$  nm (Adhikari, 2002), with total cross-section  $\sigma_0 = 4\pi a_0^2 \approx 1.96 \times 10^{-14}$   $\text{cm}^2$ . Let  $n_{\pm}(\vec{x}, t)$  be the number per unit volume at position  $\vec{x}$  and time  $t$  of *o*-Ps atoms with magnetic quantum number  $m = \pm 1$ . The rate of disappearance of  $m = \pm 1$  atoms due to collisions with  $m = \mp 1$  atoms will be  $d \ln n_{\pm}(\vec{x}, t)/dt = n_{\mp}(\vec{x}, t)\sigma_x \bar{v}$ , where  $\sigma_x = \sigma_0/3 = 0.654 \times 10^{-14}$   $\text{cm}^2$  is the cross-section at low velocity for collisions of a triplet  $m = \pm 1$  with a  $m = \mp 1$  atom, in which the result is either a *p*-Ps or *o*-Ps atom with  $m = 0$ . Here,  $\bar{v} \approx 7 \times 10^6$  cm/s is the mean velocity of a Ps atom at room temperature. Defining  $u_{\pm}(\vec{x}, t) = n_{\pm}(\vec{x}, t)/n_0(\vec{x})$ , where  $n_0(\vec{x}) = n(\vec{x}, 0)$ , the rate equations for  $|m| = 1$  *o*-Ps decay become  $du_{\pm}(\vec{x}, t)/dt = \gamma u_{\pm}(\vec{x}, t) + \beta(\vec{x})u_{\pm}(\vec{x}, t)u_{\mp}(\vec{x}, t)$ , where  $\beta(\vec{x}) = n_0(\vec{x})\sigma_x \bar{v}$ . A finite-time-step solution correct to order  $(\Delta t)^2$  may be written as

$$u(\vec{x}, t + \Delta t) = u(\vec{x}, t) + \Delta u,$$

$$p(\vec{x}, t + \Delta t) = p(\vec{x}, t) + \Delta p,$$

with

$$\Delta u = u(1 - \exp\{-\gamma\Delta t\})$$

$$+ \frac{1}{2}(1 - p^2)u^2\beta(\vec{x})\Delta t[1 - \gamma\Delta t - \frac{1}{4}u\beta(\vec{x})\Delta t],$$

$$\Delta p = p(1 - \exp\{-\gamma\Delta t\})$$

$$+ \frac{1}{8}p(1 - p^2)u^2\beta^2(\vec{x})\Delta t^2.$$

These equations are used including a Gaussian beam profile to obtain the fitted curve for the compressed beam in Fig. 1. The fitted value for  $\beta$  implies  $\sigma_x/(1 - P) = \beta\delta/n_{2D}^0 I^2^{1/2} v = 1.7 \times 10^{-14}$   $\text{cm}^2$ , which agrees with the expected value for  $\sigma_x$  if the effective sample porosity is 61%. This is higher than the porosity determined by gas adsorption, and implies that the Ps does not fill the porous material uniformly, either because of the existence of voids or because the Ps diffusion coefficient is not sufficient to make a uniform distribution throughout the thickness of the sample. Alternatively,  $\text{Ps}_2$  molecule formation at the surface of the pores may be a significant factor.

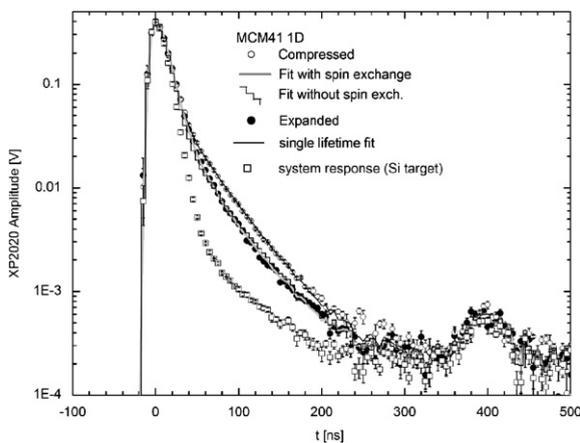


Fig. 1. Single-shot lifetime spectra obtained using 20 ns pulses of  $1.4 \times 10^7$  3 keV positron impinging on an MCM41 1D porous silica sample in vacuum at 300 K.

### 3. Ps molecules

Measurements of the spin exchange quenching effect exhibit a linear dependence on the positron density as expected. A measurement of the quenching effect versus implantation energy shows a peak at energies corresponding to the positrons being implanted into the center of the sample, but with a curious increase in apparent spin exchange quenching rate for implantation at very low energies (see Cassidy et al., 2005). It is possible that we are observing the production of Ps<sub>2</sub> molecules from Ps atoms that have taken up residence on the sample surface and leave the surface spontaneously because of the exothermic reaction. Suppose we have a Ps surface density  $n_{2D}$ , and that the Ps atoms form Ps<sub>2</sub> molecules if they get within a radius  $r$ . The Ps<sub>2</sub> formation rate is then  $\Gamma = \frac{1}{2}n_{2D}4r\sqrt{2\bar{v}}$ , where the  $\frac{1}{2}$  is because only half the collisions are  $m = +1$  scattering from  $m = -1$  and the converse. With  $r = 0.1$  nm and assuming the Ps surface state (Sferlazzo et al., 1985) is formed from 5% of the implanted positrons, the Ps<sub>2</sub> formation rate will be  $10^9/s$ , sufficient to turn most of the surface Ps into Ps<sub>2</sub>. Since most of the surface Ps probably annihilates with electrons of the solid in about 1 ns anyway, we cannot say that this effect is evidence for the production of Ps<sub>2</sub> molecules. However, the high density of Ps that is being formed in our samples will be sufficient to obtain conclusive evidence for Ps<sub>2</sub> production using a 1 ns beam pulse and measurements as a function of temperature using an Al target surface. We expect that with an oxygen-treated Al surface (Lynn, 1980; Mills et al., 1991) the Ps<sub>2</sub> molecules will roll off the surface at close to the kinematic rate, resulting in the production about 1 Ps<sub>2</sub> molecule for every 10 incident positrons, sufficient for an accurate measurement of its 1S1S–1S2P interval (Varga et al., 1998) at 235.60 nm.

### 4. Ps<sub>2</sub> emission rate

The Ps apparently residing at the surface of a single-crystal quartz surface (Sferlazzo et al., 1985) has a thermal activation energy 0.15 eV for Ps emission from the surface into the vacuum that was interpreted as the binding energy of the Ps to the quartz surface. At high surface Ps densities, there will be significant interactions of surface Ps atoms, as suggested by Cassidy et al. (2005). Here I show that Ps<sub>2</sub> emission rather than spin exchanging collisions would be the dominant effect in this situation.

The fugacity is defined as  $z = \exp\{\beta\mu\}$ , where  $\mu$  is the chemical potential or Gibbs free energy per particle (Huang, 1987). For a gas of  $N$  particles in a  $d$  dimensional cubical box with sides of length  $L$  obeying Bose–Einstein statistics, the density  $n^{(d)} = N/L^d$  is related to the fugacity by

$$n^{(d)} = \lambda^{-d} g_{d/2}(z) + L^{-d} z/(1-z), \quad (1)$$

where  $g_k(z) = \sum_{l=1}^{\infty} z^l/l^k$  and  $\lambda = \sqrt{2\pi\hbar^2/mkT}$  is the thermal wavelength, which is  $\sqrt{2\pi}$  times the de Broglie wavelength of a particle of mass  $m$  and kinetic energy  $kT$ . For  $z \ll 1$  and  $\lambda \ll L$ , Eq. (1) becomes

$$n^{(d)} = \lambda^{-d} z. \quad (2)$$

Now suppose we have a box of volume  $V$  and surface area  $A$  containing in thermal equilibrium  $N_{Ps}$  Ps atoms and  $N_{Ps_2}$  Ps<sub>2</sub> molecules in the vacuum and  $N_S$  Ps atoms on the surface. Assume that annihilation is turned off and that the total number of positrons is  $N = N_{Ps} + N_S + 2N_{Ps_2}$ . The densities of Ps atoms and Ps<sub>2</sub> molecules in the 3D volume are  $n^{Ps} = N_{Ps}/V$  and  $n^{Ps_2} = N_{Ps_2}/V$ , respectively. The density of Ps atoms on the surface is  $n^S = N_S/A$ . Ps atoms are bound to the surface by an energy  $E_S$  and the binding energy of the Ps<sub>2</sub> molecule is  $E_M = 0.435$  eV (Frolov and Smith, 1997). At equilibrium, the chemical potentials of the three components are  $\mu_{Ps}$ ,  $\mu_{Ps_2} = 2\mu_{Ps} + E_M$ , and  $\mu_S = \mu_{Ps} + E_S$ . We now find from Eq. (2) that

$$n^{Ps_2} = \lambda_{Ps_2}^{-3} \lambda_{Ps}^6 (n^{Ps})^2 \exp\{\beta E_M\} = 2^{3/2} \lambda_{Ps}^3 (n^{Ps})^2 \exp\{\beta E_M\} \quad (3)$$

and

$$n^S = \lambda_S^{-2} \lambda_{Ps}^3 n^{Ps} \exp\{\beta E_S\} = (m_S/m_{Ps}) \lambda_{Ps} n^{Ps} \exp\{\beta E_S\}. \quad (4)$$

Rewrite these equations to obtain

$$n^{Ps_2} = 2^{3/2} \lambda_{Ps} (m_{Ps}/m_S)^2 \exp\{\beta(E_M - 2E_S)\} (n^S)^2 \quad (5)$$

and

$$n^{Ps} = (m_{Ps}/m_S) \lambda_{Ps}^{-1} \exp\{-\beta E_S\} n^S. \quad (6)$$

Note that the Ps<sub>2</sub> may also have a surface state with a binding energy that is probably greater than  $2E_S$  because the mass is twice as great, and the polarizability is also about twice as great, as that of Ps. Thus, at thermal equilibrium, there will also be a population of Ps<sub>2</sub> molecules on the surface. I am ignoring this because I assume the rate of populating the Ps<sub>2</sub> surface state starting from surface Ps is very small, since in the process of forming a molecule from two Ps atoms on a smooth surface, Ps<sub>2</sub> receives a large-momentum kick perpendicular to the surface that will make it unlikely to enter the surface state. The rates of Ps and Ps<sub>2</sub> hitting a unit area of surface are  $\Gamma^{Ps} = \frac{1}{4} n^{Ps} \langle v_{Ps} \rangle = \frac{1}{4} n^{Ps} \sqrt{8kT/\pi m_{Ps}}$  and  $\Gamma^{Ps_2} = \frac{1}{4} n^{Ps_2} \langle v_{Ps_2} \rangle = \frac{1}{4} n^{Ps_2} \sqrt{8kT/\pi m_{Ps_2}}$ . At equilibrium, the rate of particles leaving the surface is the same as the rate of particles hitting the surface (Richardson, 1924; Pendry, 1980; Chu et al., 1981). If the sticking coefficients are  $S_{Ps}$  and  $S_{Ps_2}$ , the emission rates per unit area for Ps

and  $\text{Ps}_2$  are thus

$$\Gamma^{\text{Ps}} = S_{\text{Ps}}(kT/h)(m_{\text{Ps}}/m_{\text{S}}) \exp\{-\beta E_{\text{S}}\}n^{\text{S}} \quad (7)$$

and

$$\Gamma^{\text{Ps}_2} = S_{\text{Ps}_2}(\hbar/m_{\text{c}})(m_{\text{Ps}}/m_{\text{S}})^2 \exp\{\beta(E_{\text{M}} - 2E_{\text{S}})\}(n^{\text{S}})^2. \quad (8)$$

We are contemplating the situation where there is only surface Ps initially with no Ps or  $\text{Ps}_2$  in the vacuum. The emission rates will then be the same as in Eqs. (7) and (8) except that the  $\text{Ps}_2$  will not be emitted with a distribution of velocities that is in thermal equilibrium because of the low probability of phonon creation during the rapid emission of a newly formed  $\text{Ps}_2$  molecule. Essentially, no  $\text{Ps}_2$  molecules will be emitted with less energy than  $E_{\Delta} = E_{\text{M}} - 2E_{\text{S}}$ , if  $E_{\Delta}$  is greater than zero. (A presumably small portion of the surface Ps binding energy is caused by the Ps being dressed in virtual phonons, i.e. the Ps is in a polaron-like state and sits in a lattice distortion in the form of a surface dimple. The dressing will be largely left behind in the form of an occasionally emitted phonon when  $\text{Ps}_2$  is formed and leaves the surface in a time shorter than a lattice vibration period.) Thus, only the upper portion of the Boltzmann distribution of possible emission energies is effective and the Boltzmann factor  $\exp\{\beta(E_{\text{M}} - 2E_{\text{S}})\}$  is to be replaced by 1, if  $E_{\Delta} > 0$ . The  $\text{Ps}_2$  emission rate per surface-Ps atom therefore becomes

$$\gamma^{\text{Ps}_2} = S_{\text{Ps}_2}(\hbar/m_{\text{c}})(m_{\text{Ps}}/m_{\text{S}})^2[\vartheta(E_{\Delta}) + \exp\{\beta(E_{\Delta})\}\vartheta(-E_{\Delta})]n^{\text{S}}, \quad (9)$$

where  $\vartheta(x)$  is the Heaviside unit step function. If  $S_{\text{Ps}_2} = 1$ ,  $m_{\text{S}} = m_{\text{Ps}}$ ,  $E_{\Delta} > 0$ , and  $\tau_{\text{S}}$  is the surface Ps lifetime, then  $\gamma^{\text{Ps}_2}\tau_{\text{S}} = 1$  when  $n^{\text{S}} = [1n^{\text{S}}/\tau_{\text{S}}] \times [0.8 \times 10^9/\text{cm}^2]$ . In Cassidy, et al. (2005), we reported achieving positron areal densities of  $3.3 \times 10^{10}/\text{cm}^2$ . If 10% of the positrons populate the Ps surface state, it is possible that a large fraction of the Ps on the surface of an insulator can be turned into  $\text{Ps}_2$  molecules at these densities. In particular for quartz, the activation energy determined by Sferlazzo et al. implies  $E_{\text{S}} = 0.15$  eV and  $E_{\Delta} = 0.13$  eV, so that  $\text{Ps}_2$  should be spontaneously emitted from the surface at the temperature-independent rate

$$\gamma_0^{\text{Ps}_2} = S_{\text{Ps}_2}(m_{\text{Ps}}/m_{\text{S}})(\hbar/m_{\text{c}})n^{\text{S}}. \quad (10)$$

It is instructive to estimate the  $\text{Ps}_2$  emission rate from kinetic theory, and to see what the rate  $\gamma_0^{\text{Ps}_2}$  and the sticking coefficient  $S_{\text{Ps}_2}$  mean in terms of an effective Ps–Ps interaction radius on the surface. Suppose we have a triplet Ps (*o*-Ps) surface density  $n^{\text{S}}$ , and that two surface Ps atoms with total spin  $S = 0$  form  $\text{Ps}_2$  molecules if they get within a radius  $r$ . The  $\text{Ps}_2$  formation rate is then

$$\gamma_0^{\text{Ps}_2} = \frac{1}{3}n^{\text{S}}4r\sqrt{2}\langle v_{\text{S}} \rangle, \quad (11)$$

where the factor 1/3 is the fraction of the collisions that are between Ps atoms with the proper spins for a reaction to take place. Eq. (10) then implies that the interaction radius is approximately 1 de Broglie wavelength of the surface Ps atoms times the sticking coefficient,

$$r = \frac{3\sqrt{2}}{16}S_{\text{Ps}_2}\lambda_{\text{S}}. \quad (12)$$

So long as the Ps kinetic energy is small compared to the binding energy of the  $\text{Ps}_2$  molecule, spin exchange will be determined by the singlet channel (total spin 0) Ps–Ps scattering length  $a_0 = 0.395$  nm (Adhikari, 2002), with total cross-section  $\sigma_0 = 4\pi a_0^2 \approx 1.96 \times 10^{-14}$  cm<sup>2</sup>, and spin exchange quenching cross-section  $\sigma_{\text{X}} = \sigma_0/9$  for unpolarized triplet Ps atoms. We may then write for the spin exchange rate

$$\gamma^{\text{X}} = n^{\text{S}}w^{-1}\sqrt{2}\langle v_{\text{S}} \rangle\sigma_{\text{X}}, \quad (13)$$

where  $w \approx 1$  nm is the effective width of the surface Ps wave function perpendicular to the surface. This implies an effective interaction radius for spin exchange,

$$r^{\text{X}} = \frac{3}{4}\sigma_{\text{X}}/w \approx 0.15 \text{ nm}. \quad (14)$$

Given that  $\lambda_{\text{S}} \approx 3.0$  nm at 300 K, it is evident that for  $S_{\text{Ps}_2} > 0.05$ , the interactions of high-density surface Ps will be dominated by  $\text{Ps}_2$  molecule production. Just as in the case of Ps emission from a metal surface, where the sticking coefficient is close to unity due to the large interactions of Ps with the surface electrons (Mills et al., 1991),  $\text{Ps}_2$  should also have a large sticking coefficient, because it can easily break up into two surface Ps atoms upon impact with an insulator surface. An incoming  $\text{Ps}_2$  molecule is strongly coupled by the surface potential to a pair of Ps atoms moving in opposite directions along the surface, with no need for energy or momentum transfer to the only available solid excitations, the bulk and surface phonons to which Ps couples only weakly. Experiments to measure the  $\text{Ps}_2$  emission rate are needed to reveal the magnitude of  $S_{\text{Ps}_2}$ .

At Ps surface densities such that the surface Ps undergoes Bose–Einstein condensation, namely  $n_{\text{S}} > \lambda_{\text{S}}^{-2} = 3 \times 10^{11}/\text{cm}^2$  at 10 K, a substantial fraction of the surface Ps would be in the ground state, leading to a high probability of  $\text{Ps}_2$  molecule emission into the vacuum with no thermal energy spread about the energy  $E_{\Delta}$ . The  $\text{Ps}_2$  emission process is recoilless (Mossbauer, 1958), with momentum being conserved by the recoil of the whole crystal. The  $\text{Ps}_2$  would be emitted within a narrow cone of half angle  $\Delta\theta \approx \lambda_{\text{Ps}_2}/D$  in a direction perpendicular to the sample surface, where  $D$  is the Ps surface coherence length or the diameter of the positron beam spot, whichever is least. The threshold for stimulated emission of  $\text{Ps}_2$  will occur when the number of  $\text{Ps}_2$  molecules emitted  $N$  is greater than the number of available  $\text{Ps}_2$  modes  $M$  within the emission cone:  $N > M \approx [(E_{\text{M}} - 2E_{\text{S}})\Delta\theta\tau/\pi\hbar]^2$ ,  $\tau$  being the  $\text{Ps}_2$  lifetime,  $\tau \approx$

0.25 ns (Schrader, 2004). For  $D = 5 \mu\text{m}$ , the threshold for stimulated emission of a  $\text{Ps}_2$  “atom laser” beam (Mewes et al., 1997) will be reached with only a few thousand  $\text{Ps}_2$  molecules. The availability of recoilless  $\text{Ps}_2$  emission would be useful for a precision measurement of the frequency of the optical transition of  $\text{Ps}_2$  to its  $L = 1$  excited state (Varga et al., 1998), and for producing monoenergetic collimated sources of high-energy  $\text{Ps}^+$  and  $\text{Ps}^-$  ions, and fast Ps.

## 5. Importance of positronium Bose–Einstein condensation

While it is intriguing that the light mass of Ps means that it can undergo Bose–Einstein condensation at room temperature (Platzman and Mills, 1994), it is the novel effects associated with the collective properties of antimatter that make the field of Ps BEC worthy of study.

- First, it is important to note that the total spin 2 *o*-Ps–ortho-Ps scattering length (Adhikari, 2002) is positive,  $a_2 = 0.83 \text{ \AA}$ . The small positive scattering length makes a collection of spin-polarized *o*-Ps atoms (all in the  $m = 1$  triplet state) a nearly noninteracting ideal gas that may undergo Bose–Einstein condensation.
- It is interesting that at very high densities, a Bose–Einstein condensed state may not be the appropriate way to describe the ground state of a Ps gas. Rather, there may be a transition to BCS pairing rather than a collection of individual Ps atoms. Although it would be difficult to achieve the needed densities ( $10^{23}/\text{cm}^3$ ), it is not impossible, and the problem of a dense gas of positrons and electrons is an interesting one (Schrader, 2005).
- It should be possible to make a Ps atom laser beam from a Ps BEC that is experiencing stimulated tunneling into the vacuum from a physical cavity via one or more narrow channels in the cavity wall. The Ps atoms will tunnel without gain or loss of energy, thus producing an extremely monoenergetic beam in vacuum, the energy being that of the Ps in the many-particle ground state of the cavity. If the tunneling is occurring across the width of the cavity, the emitted Ps atoms will form a narrow beam with angular divergence equal to the width of the cavity divided by its diameter.
- A positronium atom laser beam would make a perfect source for measurements of the positronium  $1^3\text{S}_1$ – $2^3\text{S}_1$  interval (Mills and Chu, 1990; Karshenboim, 2002) with a precision of order  $10^{-3}$  of the one part in  $10^9$  natural line width. Because the Ps atom is probably fully described at this level of precision in terms of pure quantum electrodynamics theory, a Ps

1S–2S measurement could be directly related to the fundamental constants, allowing an improved interpretation of more precise standards, such as the hydrogen 1S–2S interval.

- A Ps atom laser beam, excited Doppler-free to a high Rydberg state, could be used in a Mach–Zehnder interferometer to measure the Ps gravitational red shift, and by inference revealing the gravitational interaction of antimatter (Philips, 1997).
- Since Ps atoms annihilate, it is possible for significant rates of stimulated annihilation to occur in a sufficiently dense Ps BEC, in which all the atoms are in the para state, providing the basis for an annihilation gamma ray laser (Mills et al., 2004) and a measurement of the Compton wavelength with an uncertainty comparable to its parts in  $10^{11}$  line width.
- A Ps BEC could be used as a refractive medium to allow focusing an annihilation photon laser beam to a small spot to obtain high local intensities.

## 6. Formation of dense collections of positronium in cavity structures

The first experiments will probably make use of the formation and accumulation of Ps in porous targets (Gidley et al., 1999). Porous silica structures containing cavities could be very useful for obtaining a high-density accumulation of Ps, because (1) about 50% of all positrons implanted into such materials form Ps, three-quarters of which will be in the long-lived (142 ns) triplet ground state which is called *o*-Ps; (2) the Ps has a long lifetime (100 ns) and large diffusion length (a few  $\mu\text{m}$ ); and (3) the Ps has a lower energy in larger-size pores, leading to an accumulation of Ps in large pores or cavities. At low temperatures, a macroscopic quantity of the Ps will accumulate in the largest pores because of the large confinement energy of the Ps in the smaller pores. It has recently been determined that the Ps is still able to diffuse in methyl silsesquioxane films (Fischer et al., 2005) at temperatures as low as 20 K. If the porous material contains a low-density collection of macroscopic cavities, the Ps will collect into one or more cavities, and then condense by Bose–Einstein condensation when the density is sufficiently great.

Forming a dense collection of Ps in a small cavity requires having a pulse of many positrons focused to a small diameter spot on a Ps-forming target containing a small cavity. The positron beam pulse for forming the Ps could be produced as follows:

- We are now obtaining 10 ns bursts containing  $N = 6.7 \times 10^7$  positrons from our positron accumulator. In a magnetic field  $B = 500 \text{ G}$ , the beam diameter is currently being compressed to

$w = 2.7$  mm full-width at half-maximum. We will make a longer accumulator with higher confining potentials that will be equivalent to ten such accumulators that will be filled to yield a total  $N = 6.7 \times 10^8$  positrons. The positrons will be accelerated to 10 keV and extracted through an array of small holes in a mumetal shielding plate into a magnetic-field-free region. The array will consist of a 3 mm diameter hexagonal pattern of 49 holes with diameter  $\delta = \frac{1}{4}$  mm. The open area fraction will be about 63% and the thickness of the mumetal plate in the region of the whole array will be  $\frac{1}{4}$  mm, more than sufficient to prevent magnetic saturation of the mumetal. The positrons will be efficiently transmitted through the open area since they will be traveling nearly parallel to the beam axis. The perpendicular energy width of the positrons after leaving the magnetic field will be  $E_{\perp} = 0.55$  eV, using Canter's formula (Canter, 1995)  $E_{\perp} = [35 \mu\text{eV}] \times [\delta/1 \text{ mm}]^2 \times [B/1 \text{ G}]^2$ .

- The positrons will be focused by electrostatic lens elements such that the beam converges with a 0.2 rad half-angle, thus raising the perpendicular energy to 400 eV. The positrons will converge to a 100  $\mu\text{m}$  diameter spot on the back of a single-crystal Ni remoderator film 100 nm thick. The positrons will be re-emitted from the exit side of the film with an efficiency of approximately 15%, thus yielding 10 ns bursts containing  $6 \times 10^7$  positrons with a transverse energy distribution of about 0.05 eV, slightly higher than a room-temperature energy spread. The positrons will be accelerated to 2 keV and brought to a focus on the sample surface containing Ps cavities. Allowing for aberrations, the full-width at half-maximum of the focused positron beam should now be  $w = 5 \mu\text{m}$ .
- The average positron density per unit area, weighed by the density distribution, is  $\langle n \rangle = 2 \ln\{2\} \pi^{-1} (N/w^2) = 0.441 \times (N/w^2) = 1.1 \times 10^{14}$  positrons per  $\text{cm}^2$ . The central density will be twice as great,  $n(0) = 2.2 \times 10^{14}$  positrons per  $\text{cm}^2$ . The positron pulse must now be implanted into an appropriate target that will (a) transmit the positrons into a BEC-forming cavity, (b) dissipate the energy of the implanted positrons, (c) emit Ps into the cavity with a high probability and (d) have a long lifetime for Ps in the cavity.
- A target satisfying all these criteria would be made on a single-crystal quartz substrate covered by a 100 nm thick film of methyl silsesquioxane. The porous film with the holes will be covered by a 10 nm amorphous  $\text{SiO}_2$  film having a regular array of 0.5  $\mu\text{m}$  square bubbles 50 nm high. The single-crystal quartz will quickly dissipate the positron energy as ballistic phonons (Platzman and Mills, 1994), and it will form Ps and emit it into the porous silica with greater than 50% probability (Sferlazzo et al., 1985).

- The Ps in the porous silica will diffuse on the order of  $\lambda = 2 \mu\text{m}$ . About 25% of the positrons implanted into a circle of radius equal to the diffusion length will collect into the bubbles. With the positron pulse centered on a bubble, the initial number of *o*-Ps atoms in a bubble cavity will be  $N = \frac{1}{4} n(0) \pi \lambda^2 = 7 \times 10^6$ .
- To ensure that the cavity Ps lifetime is long, the Ps must be prevented from sticking to the walls. A nanotexture similar to that of a silica gel will be formed on the walls by means of helium ion implantation followed by annealing in oxygen. The quantum size effect will cause the Ps surface binding energy to become negative, thus preventing the Ps from rapidly annihilating on the walls.
- The target cavities will be shaped using a lithographic mask to expose a photoresist on the surface of a silicon wafer. The silicon wafer will then be coated with a 10 nm oxide layer by standard semiconductor technology (Yamane, 1988). After the quartz substrate and silicon wafer have been ion-bombarded to nanotexture their surfaces, they will be wafer-bonded and annealed to remove water layers. The final step in the preparation of the target will be to etch away the silicon, leaving a single-crystal quartz substrate with an array of bubble cavities covered by thin  $\text{SiO}_2$  membranes.

Unlike alkali atoms, Ps has a de Broglie wavelength much longer than its atomic radius at temperatures of order 100 K, and it interacts weakly with the walls of porous silica materials. The long de Broglie wavelength means that a large number of Ps atoms could be collected into a cavity in the porous material without any significant single-particle energy level shift due to the hard-core repulsion of the other atoms. Condensation of a large number of atoms in a single cavity could be detected by observing changes in the triplet Ps lifetime, because spin exchange quenching of the lifetime will be prevalent for a high-density Ps gas due to Ps–Ps collisions as long as the Ps is not 100% polarized (Mills, 1995b; Cassidy et al., 2005).

Note that the positrons from the  $^{22}\text{Na}$  source are spin polarized due to parity non-conservation in  $\beta$ -decay, and that the polarization is conserved in the magnetic field and is not affected significantly by the remoderation process. The average helicity of the  $\beta$  particles is  $v/c$ , averaged over the  $\beta$  spectrum and is about 70% for  $^{22}\text{Na}$ . On the other hand, the average polarization of the positrons emitted into a  $2\pi$  solid angle from a pure  $^{22}\text{Na}$   $\beta$  spectrum is half this amount, or about 35%. This polarization is reduced somewhat in our system, because the source has a Ta backing that is meant to increase the positron intensity by back-reflecting some fractions of the positrons that would otherwise be absorbed by the

source holder. On the other hand, the source is covered by a 5  $\mu\text{m}$  Ti foil that attenuates lower-energy positrons, and thus increases the average polarization in our beam. In addition, the moderator favors forward-emitted positrons and also tends to increase the polarization. A reasonable guess is that these effects approximately cancel, leaving us with a roughly 35% positron beam polarization.

About one-sixth of the *o*-Ps should remain after spin exchange collisions have removed the minority spin positrons in the  $m = -1$  and  $m = 0$  triplet Ps states. Thus, about 20 ns after the positrons have been implanted into the quartz substrate, we should have a gas containing  $N/6 = 1 \times 10^6$  100% polarized  $m = 1$  triplet Ps atoms in the cavity. The Ps density in the cavity will be  $8 \times 10^{19}/\text{cm}^3$ . After cooling for two lifetimes (about 100 ns), the Ps density in the cavity will be  $1 \times 10^{19}/\text{cm}^3$ , for which the BEC transition temperature will be 60 K. The Ps Bose–Einstein condensation dynamics, the macroscopic quantum effects of the condensate, and the properties of ultra-cold Ps emitted from the condensate may then be studied by optical spectroscopy.

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