

Optical spectroscopy of atomic and molecular positronium

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Abstract. Positronium (Ps) is a purely leptonic hydrogen-like atom formed from an electron and a positron. Since the interactions of electrons and positrons are thought to be almost entirely electromagnetic, precision measurements of the Ps energy levels should constitute a good test of QED theory. The ultimate precision is limited by the rapid annihilation of the various Ps states and the number of Ps atoms available. Much progress in making better Ps sources has been made since the 1950's when Ps was discovered and its principle characteristics measured in by the pioneering experiments of Martin Deutsch. The most notable milestones were the first reproducible schemes for making slow positrons and Ps in vacuum by Canter and his co-workers in the 1970's and the discovery of the enabling technology for accumulating slow positrons by Surko and co-workers in 1989. These techniques have made it possible to generate high density bursts of slow Ps atoms that has led to the production of di-positronium molecules, Ps₂, and the observation of the Lyman-alpha-like transition in Ps₂ at a wavelength of 251 nm predicted by Varga and co-workers. The possibilities for 1S-2S spectroscopy of triplet and singlet Ps with precisions relevant to the proton charge radius problem and efficient production of slow Rydberg Ps atoms useful for measuring Ps free fall are discussed.

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We met at Lanzhou in a setting fit for kings
To learn more about how the world works,
Mindful of those who could not join us there.

1. Introduction.

These are exciting times for physics, when the fact that 95% of the world is dark matter and dark energy suggests new particles and forces must exist. Some evidence that new physics is already being detected is (1) There is a significant difference in the charge radius of the proton as found from measurements using electrons vs negative muons; and (2) There is a near-earth and cosmic excess of positrons that suggests positron accumulation from dark matter annihilation. One might ask if perhaps measurements on the purely QED atom positronium (Ps) could shed light on the problem. The narrow linewidth of its triplet 1S-2S interval, 1.27 MHz compared to the full triplet 1S-2S interval 1 233 607 216 MHz, suggests that precisions of parts in 10¹² are possible. Such a measurement is not so easy as it might at first appear because Ps spectroscopy is not your ordinary type of atomic physics. This is

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because the positrons for the experiments must be created out of the vacuum with necessarily relativistic energies, the positrons have to be slowed to eV energies and compressed in space and time, the Ps must be formed in vacuum to avoid perturbations, the Ps atoms decay in a few nsec, and they must be detected one at a time.

Positrons are produced by pair production at electron accelerator targets and in nuclear reactors and from the beta decay of certain radioactive isotopes. The many applications for positrons include atomic physics experiments, positron emission tomography (PET), measuring defect densities in materials and electron momentum densities in solids.

Positronium is a purely leptonic hydrogen-like atom formed from an electron and its antiparticle, the positron. The structure and interactions of positronium are thought to be governed purely by electromagnetic interactions. The latter implies: (a) precision measurements of the Ps energy levels should constitute a good test of QED theory [1]; and (b) Ps rapidly annihilates [2], making it difficult to accumulate and greatly restricting the ultimate measurement precisions. Ps was discovered and its principle characteristics measured in the 1950's by Martin Deutsch [3]. The possibilities for atomic physics experimentation opened up with the discovery of slow positron emission from a surface [4] and the first reproducible schemes for making slow positrons and Ps in vacuum by Canter and coworkers [5, 6]. In particular, this work led directly to precision measurements of the Ps 1^3S_1 - 2^3S_1 interval [7]. The development of the enabling technology for accumulating slow positrons [8] is now making it possible to generate high density bursts of slow Ps and to observe their interaction with each other [9] and the production of di-positronium molecules, Ps₂ [10]. Short pulses of Ps atoms suited to laser spectroscopy have been used to study the Lyman-alpha-like transition in the Ps₂ molecule at a uv wavelength of 251 nm [11, 12], as well as the Ps formation and dynamics in various materials [13] and the efficient production of Rydberg Ps atoms [14] that are needed for measuring Ps free fall. Increasing the number density of a positronium gas could enable Ps Bose-Einstein condensation at room temperature [15] and perhaps stimulated amplification of annihilation radiation [16]. The 8 milestones along the way to the present state of experimentation with positronium are presented in the next section.

2. How can we do spectroscopy on transient Ps atoms? 8 milestones 1951-2005.

2.1 Milestone 1: Discovery of Positronium.

The field of experimental positronium physics was born with Martin Deutsch's report [17] that long lived (142 ns mean lifetime) triplet ground state (3S_1) Ps atoms are formed when positrons from a radioactive β^+ source (^{22}Na) stop in N₂ gas. The proof of the discovery was that the 3S_1 atoms which ordinarily decay into three photons, could be made to annihilate into two photons (2γ) from the 1S_0 singlet state by adding a little nitric oxide (NO) to the N₂ to provide nearly free electrons for spin exchanging collisions that convert triplets into singlets, for which the lifetime is 125 ps. The proof of the effect is evident in Fig. 1 which shows an increase in the amplitude of the 2γ annihilation 511 keV photopeak for 5% NO. Three months later Deutsch had measured the decay rate of 3S_1 Ps, with the value $\gamma_3 = (6.9 \pm 0.4) \mu\text{s}^{-1}$ from my fit to the 6 Freon measurements at the highest pressures in Fig. 2 [18], in agreement with the precise current theoretical value $\gamma_3 = (7.04007 \pm 0.00002) \mu\text{s}^{-1}$ [1]. It is proper to ignore the datum at the lowest pressure because many of the positrons will then be annihilating in the walls of the pressure vessel.

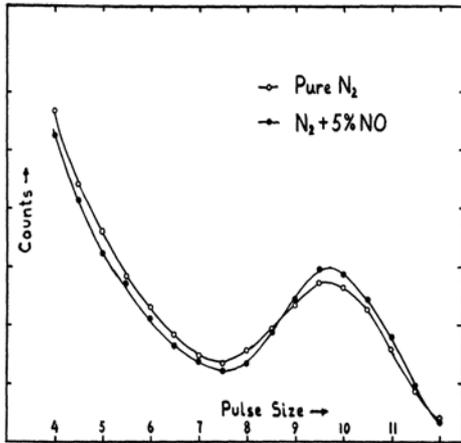


Figure 1. Martin Deutsch's evidence for the formation of triplet positronium in N_2 gas [17], and its absence when quenched by a small amount of nitric oxide.

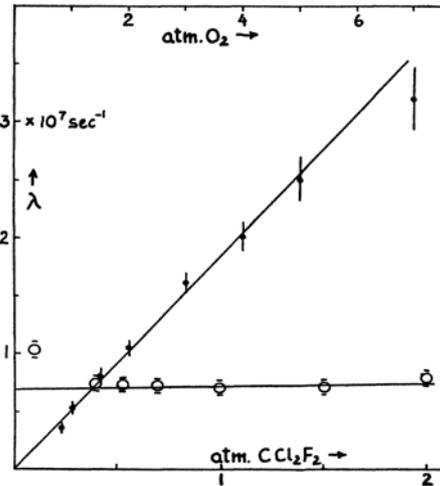


Figure 2. The positronium decay rate in Freon extrapolated to zero pressure, ignoring the lowest pressure datum, yields $\gamma_3 = (6.9 \pm 0.4) \mu s^{-1}$ [18].

Deutsch and his student Dulit next observed [see Fig. 3] that Ps triplet decays are quenched by a magnetic field due to the Zeeman mixing of the triplet $m=0$ singlet and triplet sublevels, which allowed them to deduce that the hyperfine splitting of the ground state is 0.94 meV [19]. This result was quickly followed by Deutsch and Brown's observation of the Zeeman resonance between the triplet $m=0$ and $|m|=1$ levels in a magnetic field [Fig. 4] which yielded a value for the hyperfine splitting (203.2 ± 0.3) GHz [20] in agreement with the current theoretical value (203.3917 ± 0.0005) [1]. These four amazing experiments were all performed within the space of one year. Deutsch then hoped to be able to measure the equivalent of the Lamb shift in the first excited state of positronium. His student Kendall's experiment to produce excited states [21] using an intense Sn lamp producing 242.95 nm radiation yielded a result of the expected sign and magnitude, but only at a 2.5 standard deviation confidence level.

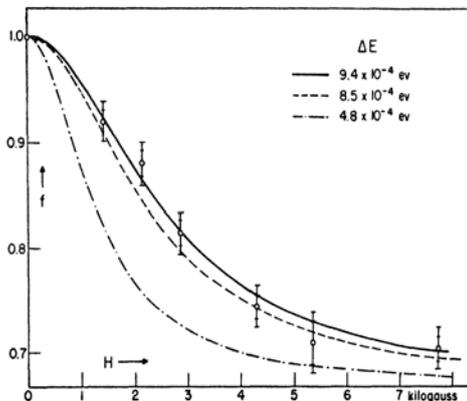


Figure 3. Magnetic quenching of three photon annihilations of positronium [19].

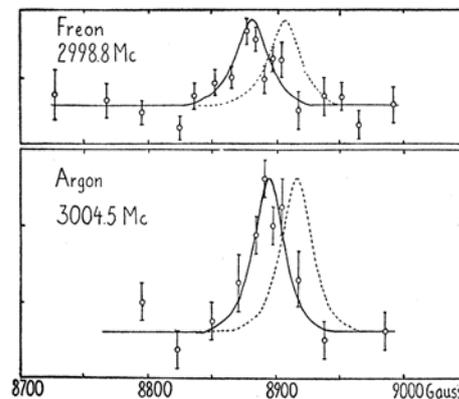


Figure 4. Zeeman resonance between the $m=0$ and $|m|=1$ states of positronium [20].

2.2 Milestone 2: The first stable slow positron moderator.

At this point progress in experimental positronium physics, so wonderfully initiated in the wonderful work of Deutsch, ceased until the field was revitalized by the successful implementation of the slow positron method over the years 1972 through 1975 by Karl Canter working with his colleagues Coleman, Griffiths, and Heyland at University College London and Mills and Berko at Brandeis University. Madansky and Rasetti's unsuccessful attempt at making a beam of low energy positrons [22] was followed by several working low intensity slow positron beams that were difficult to reproduce [23, 24, 25, 26] and finally by the serendipitous discovery of a reproducible smoked MgO positron moderator that yielded positron beams with a fast positron to slow positron conversion efficiency of about 3×10^{-5} [5], more than an order of magnitude higher than before. (One version of the story of the discovery of the MgO moderator is given in [27].) It was later discovered that solid Ne makes a 300 times better moderator than MgO [28]. A Ne moderator works by the same principle as MgO, namely that a wide band gap insulator allows few eV positrons to diffuse through the solid without energy losses due to electronic excitations, resulting in a high moderation efficiency, since this is approximately equal to the ratio of the few eV positron diffusion length to the fast positron implantation depth. Solid Ne makes a superior moderator because it has a much longer positron diffusion length, since the positron mean free path for elastic scattering in solid Ne is very large due to the presence of a deep Ramsauer-Townsend minimum in the positron-Ne scattering cross section at about 0.6 eV [29, 30].

2.3 Milestones 3 and 4: $n=1$ and $n=2$ Ps production in vacuum.

The new moderator made it possible for Canter, Mills, and Berko at Brandeis University to efficiently make ground state positronium by colliding slow positrons with a solid target in vacuum [6], as illustrated in Fig. 5. Because the target could be located far from the large background of gamma rays from the ^{22}Na source they were also able to detect a tiny amount of first excited state positronium atoms that were formed in vacuum and to measure the Lamb-shift-like $2^3\text{S}_1-2^3\text{P}_2$ interval of positronium [31, 32].

2.4 Milestone 5: Laser excitation of Ps.

Adding a positron trap and buncher to a slow positron beam permitted Chu and Mills [33] to match a continuous slow positron source to a pulsed laser to make the first laser spectroscopy on Ps. The stochastic trap shown in Fig. 6 captured positrons that were given a large angular momentum after they were inside the trap so they could not escape past a magnetic mirror. After being accumulated for about 0.1 ms the positrons were formed into a 10 ns bunch using a harmonic potential well that was suddenly switched on. The positrons then formed Ps at a Cu target and some of them were driven to the 2S state by two-photon 1st order Doppler-free excitation using 486 nm pulsed laser light and photoionized by a third photon. The photoemitted positrons were detected with high efficiency and low background using a microchannel plate detector and an E×B velocity selector. The resonance signal, the laser off background rate, and the frequency marker derived from a deuterium Balmer- β lamp and a Fabry-Pérot interferometer are shown in Fig 7.

2.5 Milestones 6 and 7: Positron accumulation and rotating wall compression.

In 1989 Surko, Leventhal and Passner accumulated 10^4 times more slow positrons using a buffer gas to trap the positrons between the end cap electrodes of a Penning trap [8]. As seen in Fig. 8, the lifetime of the positrons in the trap was about 1 minute. Greaves and Surko [34] subsequently found that positrons could be compressed to high density by a rotating electric field that was applied for a few seconds [Fig. 9]. These inventions are combined in the three stage positron beam shown in Fig. 10.

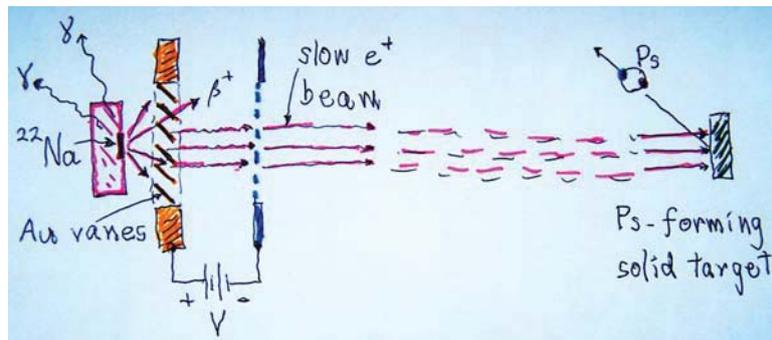


Figure 5. Canter, Coleman, Griffith and Heyland replaced the hollow Au cylinder slow positron moderator of Coleman et al. with an MgO smoked Venetian blind. Replacing the dirty metal with a wide band gap insulator made a long mean free path for the slow e^+ in the solid and thus a large reemission probability and efficient moderator.

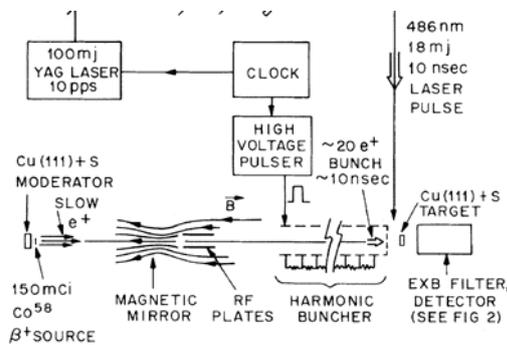


Figure 6. Stochastic positron trap and harmonic buncher of Chu and Mills [7].

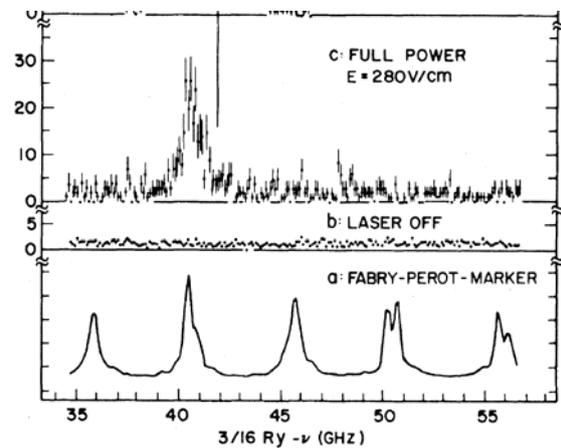


Figure 7. 2 photon resonant 3 photon ionization of triplet Ps [7].

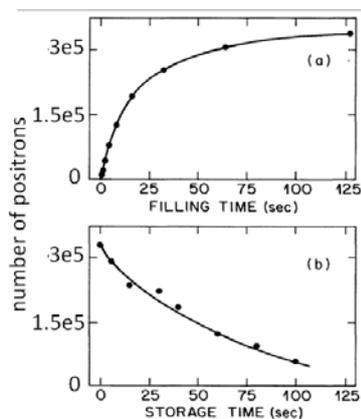


Figure 8. Storage of 0.3 million positrons in a buffer gas trap by Surko et al. [8].

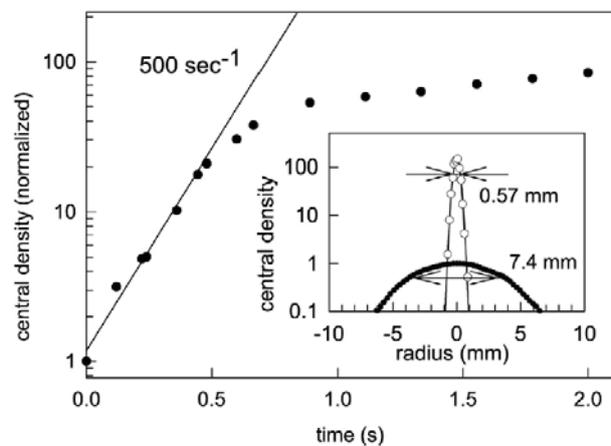


Figure 9. Compression of positrons to a high central density using a rotating wall by Greaves and Surko [34].

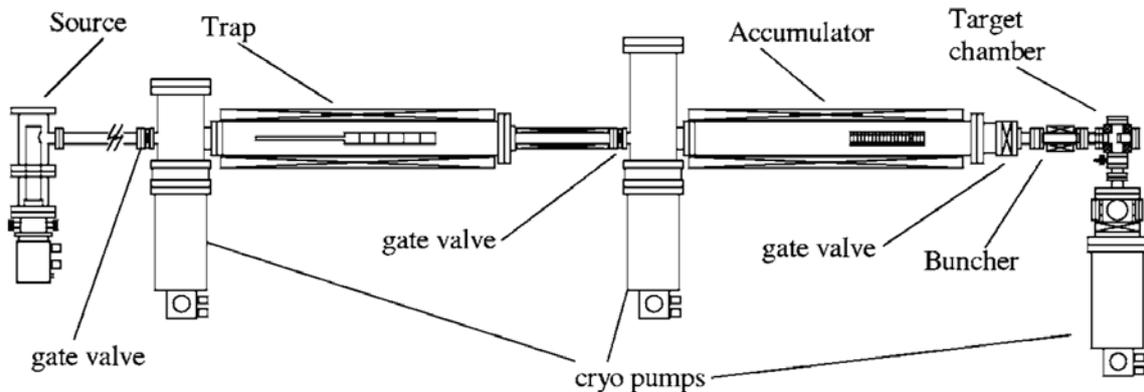


Figure 10. Positron beam consisting of a 50 mCi positron source and solid Ne moderator, a Surko buffer gas trap, and a Greaves-Surko rotating wall positron accumulator. This device produces 15 ns pulses containing $2 \times 10^7 \sim 50$ eV positrons at a rate of 1 per minute [35].

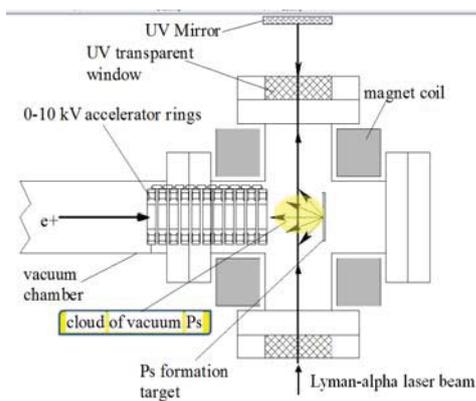


Figure 11. Positronium forming target, showing UV windows and mirror for Ps Lyman-alpha light, positron buncher and accelerator rings, pulsed magnet coils for compressing the positron beam, Ps-forming target and cloud of Ps in vacuum.

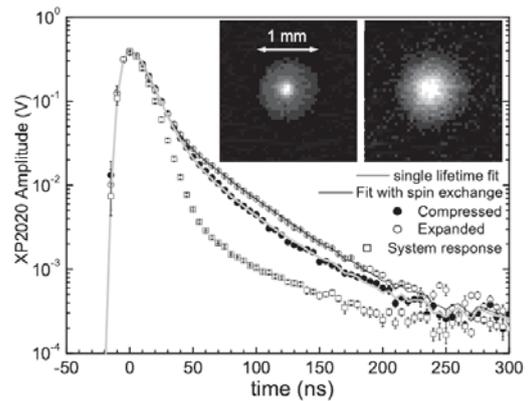


Figure 12. Observation of Ps-Ps spin exchange. When the positron beam is compressed the ~ 100 ns lifetime of triplet Ps is shortened due to spin exchange collisions converting triplet Ps into singlet Ps which has a 125 ps lifetime. The inset shows the cross section of the compressed and expanded positron beam.

2.6 Milestone 8: Single shot lifetime spectroscopy.

The positron apparatus of Fig. 10 [35] is completed by the addition of a positronium-forming target shown in Fig. 11 [36] and a pulsed magnet that compresses the positron beam to a density of about 10^{10} to 10^{11} positrons per cm^2 . Unlike previous experiments that detected one positron annihilation at a time, it is now possible to obtain a complete positron lifetime distribution using a single 1 ns pulse of positrons. Fig. 12 shows the lifetime distributions from three single shots of the positron beam at low and high density using a porous silica target, and at low density on a stainless steel target which does not form much Ps. The quenching of the long lifetime component when the positron beam is compressed was the first evidence for Ps atoms interacting with each other.

3. Examples of recent spectroscopy on Ps ions, atoms, and molecules.

3.1 Photodissociation of Ps^- ions.

The first example of a recent laser experiment using pulsed positrons is the first observation of the photodissociation of the positronium negative ion. This item can be efficiently produced by spontaneous desorption from a W surface coated with a partial monolayer of an alkali metal to reduce its electron work function [37]. Figure 13 shows the 511 keV photopeak from positron annihilations in W plus Ps singlet annihilations. A photopeak at higher energies is due to the Doppler-shifted annihilation photons from Ps^- ions that are moving towards the gamma ray detector, having been accelerated by the W target being at a negative potential relative to a grounded grid. When light from a pulsed YAG laser at 1064 nm is aimed at the Ps^- ions some of them lose the extra electron before being accelerated, as is evident from the data presented in Fig 14 [38].

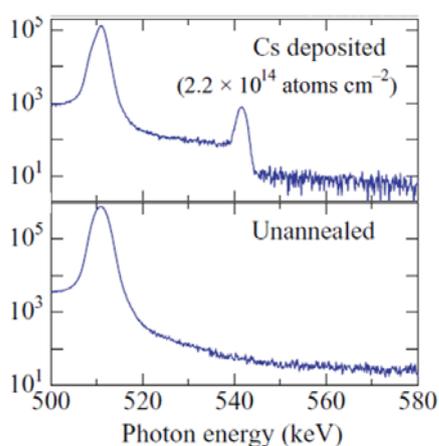


Figure 13. Gamma ray spectra from Ps negative ion annihilation in flight after desorption from a W surface with and without a Cs layer [37].

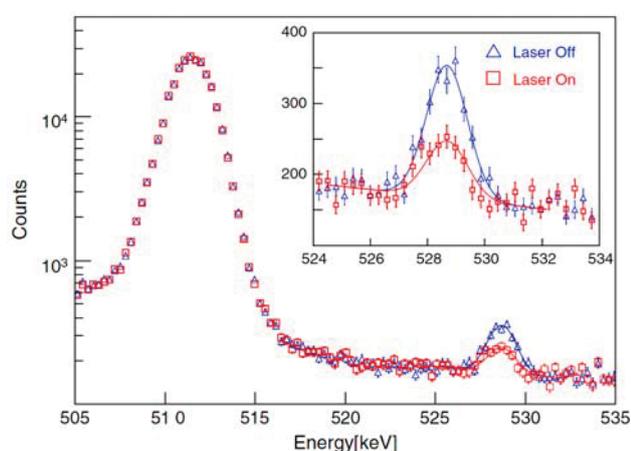


Figure 14. Evidence for photodissociation of Ps negative ions by Nagashima et al. [38].

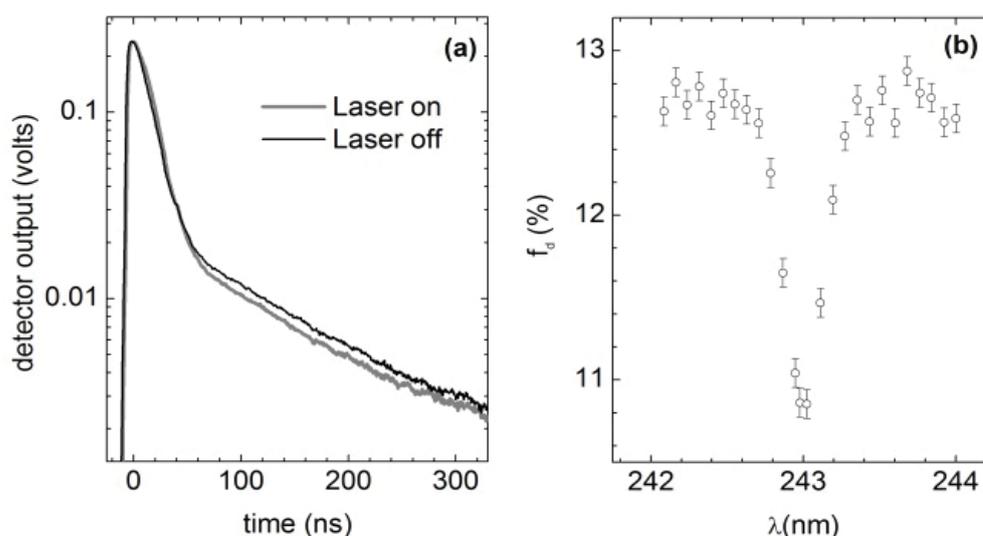


Figure 15. Single shot lifetime spectra with and without a 243 nm laser (a) and lineshape for the 1^3S-2^3P transition (b).

The new source of Ps^- ions that made this experiment possible will be very useful for a number of other applications, including precision spectroscopy of the Ps^- ion Feshbach resonances and the production of fast Ps beams.

3.2 Ps Lyman- α spectroscopy.

The second example concerns exciting the Ps Lyman- α transition using 243 nm light from a frequency-doubled pulsed dye laser at 486 nm. Fig. 15a shows that the amplitude of the time-delayed portion of a single-shot Ps lifetime spectrum is reduced when the laser is resonant with the Lyman- α transition; Fig. 15b shows the resonance in the “delayed fraction, f_d , which is the fractional area of the lifetime curve from 50 to 300 ns. The full width at half maximum (FWHM) of the resonance is about 0.33 nm compared to the 0.09 nm one would observe from a Boltzmann distribution of Ps atoms at 300 K. Ps Lyman- α spectroscopy has revealed several effects that need to be mentioned in order to understand the experiments that observed the excitation of the Ps_2 molecule to its first excited state.

3.2.1 Ps confinement energy in porous silica. First of all, Ps formed in porous silica after shallow implantation of the positrons at 1.1 keV is emitted into vacuum with a Doppler distribution of velocities parallel to the surface such that the FWHM of the Lyman- α resonance is 0.48 nm. As the implantation energy is raised the Ps suffers more and more collisions on its way to the surface and the emission energies fall until a minimum width of 0.16 nm is reached for implantation energies greater than 2.5 keV [36]. The reason this width is greater than the 0.095 nm width characteristic of the 300 K sample temperature is that the zero-point energy of the Ps in the pores is transferred to the center of mass kinetic energy upon emission from the porous sample.

3.2.2 Squeezing Ps in a pore shifts its 1S-2P resonance wavelength. If the laser is tilted so that it is not parallel to the sample surface, but instead enters the porous silica sample we observe a spectrum consisting of a sum of two Lyman- α resonances. One resonance is due to Ps confined in the pores for which the resonance is shifted to a shorter wavelength because the larger diameter 2P states are squeezed more than the 1S states [39]. The other resonance is shifted to longer wavelengths because the vacuum Ps has an average velocity component directed towards the laser.

3.2.3 A reflective sample can give a double peaked resonance. With a tilted surface reflection of the laser light can make double peaked resonance due to the presence of both red and blue shifts [40].

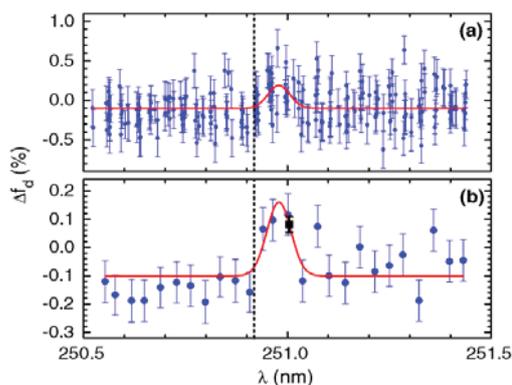


Figure 16. Change in the delayed fraction f_d caused by resonant excitation and 532 nm photoionization of Ps_2 formed in the cavities of a porous silica target. (a) raw data; (b) binned data.

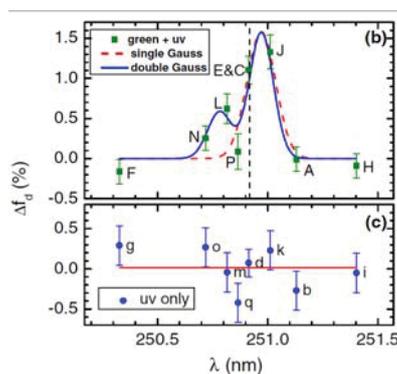


Figure 17. Same as Fig. 16 except the Ps_2 was formed at the surface of an Al(111) crystal. (b) Double resonance caused by reflection of the laser from the crystal surface (c) no 532 nm laser.

3.3 Spectroscopy on the dipositronium molecule.

3.3.1 Formation of Ps_2 . Positrons implanted onto a metal surface at high densities can make both Ps and Ps_2 [41], the dipositronium molecule that is the analog of the hydrogen molecule H_2 . On the other hand, Ps confined at high densities in the cavities of porous silica can also form Ps_2 [10]. It is to be noted that making Ps_2 represents an important milestone towards getting high Ps densities for a Ps BEC [42].

3.3.2 Optical excitation of Ps_2 . The positronium molecule was predicted to have an $L=1$ excited state that would be accessible via an electric dipole transition from the ground state using light at a wavelength of 250.917(1) nm [43]. An experiment reported by Cassidy et al. succeeded in observing optical transitions of Ps_2 [44] that was formed within porous silica (see Fig. 16) and at an Al(111) surface (see Fig. 17). The measurement using the porous target shows a shift to lower energies than the prediction presumably because of a lowering of the $L=1$ energy due to wall interactions. The poor fit of a single Gaussian resonance to the data for Ps_2 formed at an Al(111) surface is explained by the reflection of the laser beam from the surface, the beam having been tilted towards the surface so that it would be sure to interact with the short-lived Ps_2 before it decayed in vacuum after traveling about 20 μm from the surface.

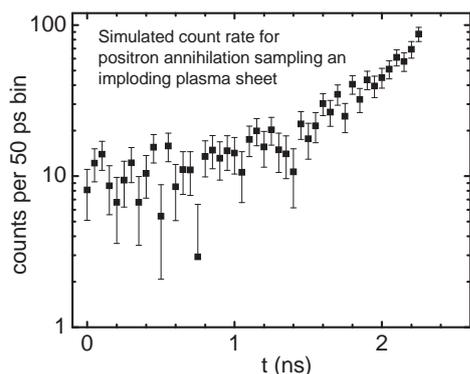


Figure 18. Simulation of the count rate in a planar implosion of $L\text{-}H_2$ assuming 10^9 positrons have been implanted at the beginning. The annihilations are assumed to be detected with 0.1% efficiency and recorded at 50 ps time intervals.

4. What is next?

The possibilities for future work include for example experimentation on the Ps BEC, measurements of the triplet and singlet $1S\text{-}2S$ intervals with parts per trillion precision, measurement of the gravitational acceleration of cold Rydberg Ps atoms, and spectroscopy of ordinary atoms and molecules to which have been added one or more Ps atoms or positrons. There are also numerous experiments to be done using dense positrons, such as the dense positron gas moving on the surface of a W field emission tip. In this case one may speculate that the positrons will collect at the tip to make a 4D (in the sense that the particles are in a 2D harmonic potential) overdamped BCS superconductor and a very bright slow positron source. Finally there are single shot applications that could benefit from having many positrons present for an experiment that cannot be repeated often, such as measuring the positron annihilation rate (simulated in Fig. 18) in a planar imploding plasma [45] to report the electron density and temperature as a function of time.

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