

## Progress towards laser spectroscopy of atomic and molecular positronium

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# Progress Towards Laser Spectroscopy of Atomic and Molecular Positronium

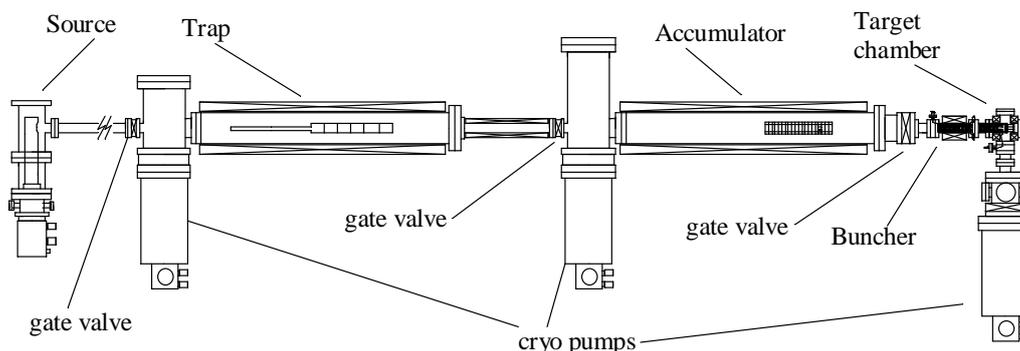
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**Abstract.** The production of molecular positronium and the existence of excited states of this molecule make it natural to consider performing laser spectroscopy on this four-particle matter-antimatter system. We report here progress towards this goal, including the production of a more intense positron beam and the development and testing of laser systems suitable for spectroscopy of atomic and molecular positronium.

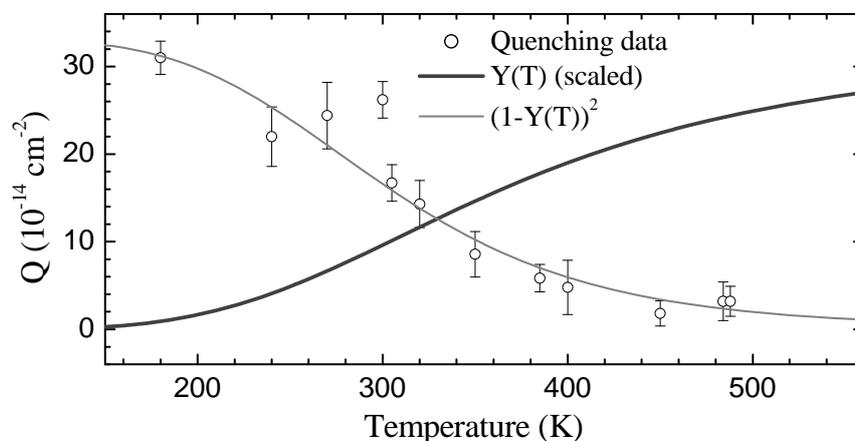
## 1. Dense Positronium Experiments

Recent experiments using positron pulses of up to  $5 \times 10^{19} \text{ cm}^{-2}\text{s}^{-1}$  led to the first observations of interacting positronium (Ps) atoms [1]. This work has allowed us to obtain evidence for the production of molecular positronium (Ps<sub>2</sub>) [2, 3] and the observation of spin exchange quenching of the long o-Ps lifetime following Ps-Ps collisions [4]. The dense positron pulses needed for this work were generated using a Surko-type trap [5] and pulsed electric and magnetic fields to provide spatio-temporal compression [6] (see figure 1). We have also developed new detectors [7] and the technique of “single shot positron annihilation lifetime spectroscopy” (SSPALS) [8] which made it possible to observe short-lived interactions between positronium atoms.



**Figure 1.** The first generation UCR positron accumulator. A rare gas moderator and source based beam supplies a gas filled trap which in turn delivers positrons to a UHV accumulator. After collecting the desired number of positrons in the accumulator they are compressed spatially using a “rotating wall” electric field and then ejected with a 200V pulse which compresses them temporally. A second time-bunching voltage is applied to the travelling positron plasma that compresses the pulse to a sub-nanosecond width, and a 1.5 Tesla magnet around the target further compresses the beam spatially.

Molecular positronium was created and observed by implanting intense positron bursts into a porous film and measuring a change in the Ps lifetime (the quenching signal) as a function of the beam density and sample temperature. Since Ps<sub>2</sub> formation requires a third body only Ps atoms in a surface state are able to form the molecule. Thus by desorbing atoms from this state by heating we observed simultaneously an increase in the Ps fraction ( $f_d$ ) [9] and a concurrent decrease in the quenching signal  $Q$ , as shown in figure 2. The quenching signal, defined as  $Q = df_d/dn_{2D}$  is a measure of the extent to which the amount of long lived Ps present depends on the beam density, and is therefore interpreted as a measure of the amount of Ps<sub>2</sub> formation. The positronium yield as a function of temperature  $Y(t)$  is related to the delayed fraction,  $f_d$  but is obtained from a more careful analysis of the desorption data (see [2] for details). Since free Ps in the voids lives longer than Ps trapped on an internal surface state, thermal desorption leads to an increase in the total amount of long lived Ps, and hence  $Y(t)$ .



**Figure 2.** Temperature dependence of the quenching signal  $Q$  (see text). The inverse correlation of this signal with the square of the thermal desorption data,  $Y(t)$ , is strong evidence for the formation of Ps<sub>2</sub> formation on the internal surfaces of the porous silica by two surface state Ps atoms (after ref [4].)

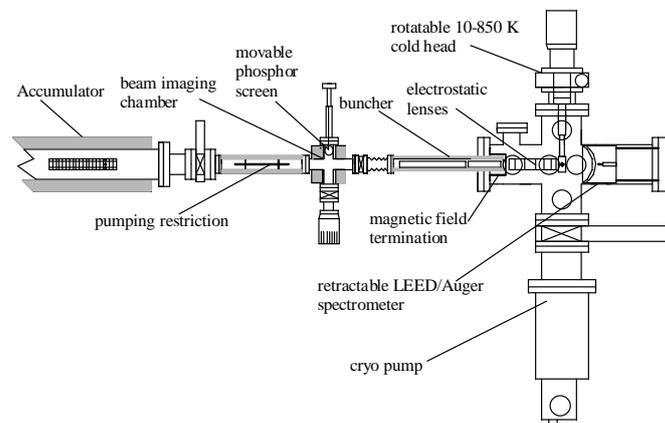
Until our results there had been no previous experimental studies of many Ps systems at all [10]. Even though the positron was first observed in 1933 [11], Ps in 1951 [12] and the positronium negative ion in 1981 [13], the Ps<sub>2</sub> molecule, which is the next member of Wheeler's polyelectron series [14], was only recently observed, largely because the required high positron densities were not available.

While convincing, the data shown in figure 2 constitute an *indirect* observation of Ps<sub>2</sub>; in order to make an unambiguous observation, and indeed to study the molecule and measure some of its properties, such as its lifetime and energy levels, it will be necessary to substantially increase the available signal and perform spectroscopy. This means increasing the positron beam density and developing the required laser systems. This is especially important inasmuch as laser spectroscopy of Ps<sub>2</sub> is most easily performed on free particles in vacuum, and it is therefore desirable to produce the molecules on a well characterised surface prepared such that the small binding energy (0.44 eV [15]) is sufficient to cause molecules to leave the surface after formation. A good candidate for this is Al(111) [10], for which we have obtained some evidence for Ps<sub>2</sub> formation [3]. Previous experiments using Al(111) suffered from poor reproducibility due to the low signal and concurrently long run times needed to obtain data, which meant that the sample surface became contaminated over the course of a single run. This problem will be mitigated in future work since additional differential pumping has vastly improved the vacuum conditions at the target, and of course the increased beam density will reduce data collection times.

## 2. Positron Beam Modifications

The experiments described above were all conducted using a pulsed magnet to compress the beam pulse to  $\sim 5 \times 10^{19} \text{ cm}^{-2}\text{s}^{-1}$ , which was sufficient to produce a signal but, at least in the case of Al(111), only barely so. Thus, we should like to increase the positron beam density, and hence the  $\text{Ps}_2$  production rate so as to provide a more intense signal. Furthermore, the production of a Ps BEC (which is one of our long term experimental goals) requires a density increase of at least two orders of magnitude over our previous work. For this reason our recent efforts have been focused on upgrading the positron beam to increase the density by remoderating [16] the positrons in a field free region.

Although positron beam remoderation has been demonstrated [17] this type of brightness enhancement has never been applied to intense pulses obtained from an accumulator. In most respects the methodology is expected to be the same as that used for DC beams. The main differences come from the fact that positrons must be extracted from the magnetic field of the accumulator, and that the use of a buncher leads to a large energy spread, which adversely affects the electrostatic focusing. A modified beam system has been constructed and is shown schematically in figure 3. The source, trap and accumulator sections are identical to those shown in figure 1. The magnetic field is maintained up to the end of the buncher where it is abruptly terminated by a mu-metal enclosure with a 3 mm diameter hole. As the beam exits this hole the radial expansion is countered by an electrostatic lens that focuses the positrons onto a thin (150 nm) Ni foil [18]. This process dramatically increases the transverse energy of the beam, but this energy is dissipated in the remoderating foil, and the  $\sim 10\%$  of the particles that emerge from the other side may then be transported in a field free region and refocused onto the target. The time width of the pulse is generally unaffected by this procedure, although the energy spread caused by the buncher operation does affect the focusing. Nevertheless, in this way we expect to be able to produce a beam spot containing around 3 million positrons in a 25 micron FWHM spot, corresponding to a pulse of  $\sim 4 \times 10^{20} \text{ cm}^{-2}\text{s}^{-1}$  which is an order of magnitude higher than we have obtained previously, and from which we would expect a  $\sim 10\%$   $\text{Ps}_2$  formation fraction.



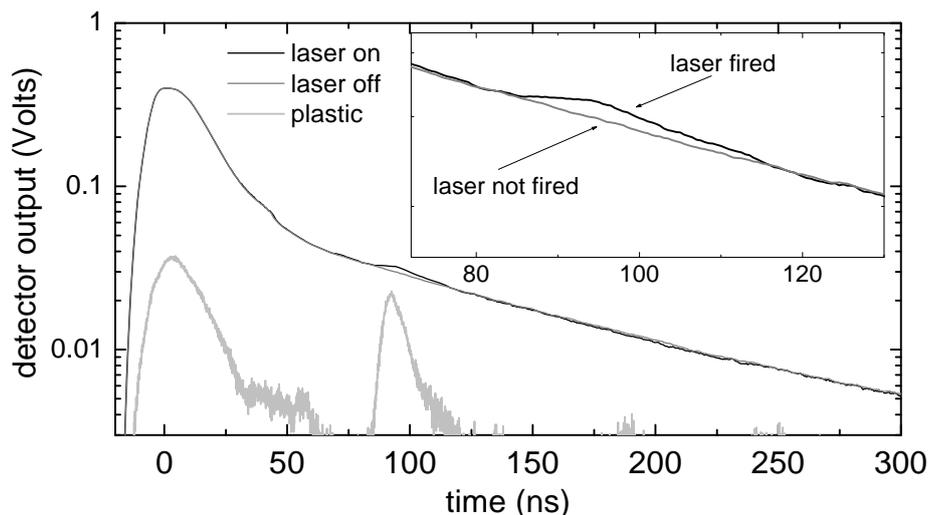
**Figure 3.** The second generation UCR system allows us to extract the positron beam from the magnetic field and remoderate. Then the beam is electrostatically transported and focused onto the target. Although this involves a loss of around 90% of our positrons, the beam density is expected to be substantially increased.

Work is in progress to produce the electrostatic pulsed beam. The estimate of a 25 micron spot size is a conservative one based on numerous simulations. So long as space charge effects are not significant it may be possible to do much better, and 5 microns is not out of the question (corresponding to a pulse approaching  $1 \times 10^{22} \text{ cm}^{-2}\text{s}^{-1}$ ). This, however, may require a new lens design to optimize filling and minimize aberrations for future Ps BEC experiments, but for the  $\text{Ps}_2$  spectroscopy a spot size smaller than 50 microns will probably be sufficient.

So far we have successfully extracted the positron beam from the magnetic field and focused it with the first lens into a screen placed at the remoderator position. We estimate that this process leads to a loss of no more than 20% of the beam, although it is difficult to be precise when measuring this via annihilation gamma rays because the field termination position, being only  $\sim 1$  cm away from the remoderator, leads to a large background signal. Characterization of the beam will be more thorough when it is remoderated and transported to the target region.

### 3. Laser spectroscopy of Ps and Ps<sub>2</sub>

Prior experiments concerning the formation of Ps<sub>2</sub> in porous silica demonstrated the expected existence of the bound state of two positrons and two electrons, but provided no other information about the molecule. Several excited states of Ps<sub>2</sub>, however, have been predicted [19, 20, 21, 22]; laser spectroscopy of these states may be used to measure its energy interval and lifetime, which can be compared directly with theory (the  $\sim 10$  ppm level we expect to obtain from our initial measurements will already be sensitive to relativistic corrections [22]). Moreover, the excited state energy interval is sufficiently different from that of Ps (251 nm instead of 243 nm for Ps) that such spectroscopy would constitute a direct and unambiguous observation of the molecule.



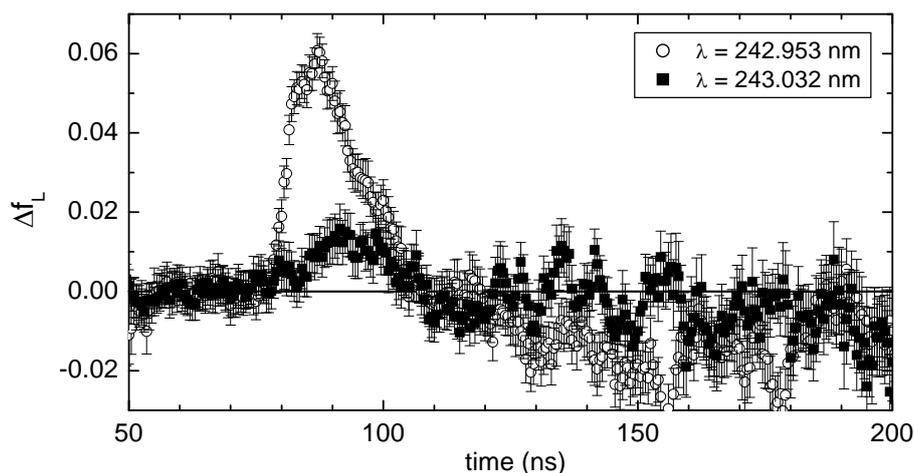
**Figure 4.** Positronium lifetime spectra obtained with and without firing the laser at 242.953 nm (in air) using a lead tungstate scintillator coupled to a photomultiplier. An increase in the Ps annihilation rate caused by resonant excitation to the 2P state and (subsequent magnetic quenching) is highlighted in the inset. Also shown is the output from a plastic scintillator coupled to a photo-multiplier that was able to detect both annihilation gamma rays following the positron pulse implantation as well as a portion of the laser light. The lifetime spectra shown are averages of 10 shots each. See reference [8] for details of the single shot lifetime measurement technique.

The production of intense bursts of positronium atoms and molecules is well suited to laser spectroscopy, and we have tested the integration of the laser system developed for excited state Ps<sub>2</sub> studies with our positron beam by producing Ps in vacuum and exciting the 1S-2P transition. Tunable 243 nm radiation was generated by sum frequency mixing of the output of a Q-switched Nd:YAG (Surelite 1-20) third harmonic (355nm, 7ns) with a dye laser (Spectra Physics PDL-1 with LDS 765 dye) tunable between 755 nm to 775 nm. The diameter of the output beam was  $\sim 6$  mm at the position of the sample. The 243 nm pulse energy was approximately 100  $\mu$ J. This same laser system will be able to provide 251 nm light required for studies of excited state Ps<sub>2</sub> (with a different dye). The laser pulses were introduced into the beam imaging chamber (see figure 3) just after the accumulator. This meant that the positron pulses used were  $\sim 15$  ns wide as the main buncher was not used. Clearly this

arrangement would not be suitable for  $\text{Ps}_2$  studies due to the short lifetime of this molecule, but it is adequate for Ps studies where the vacuum lifetime is  $\sim 140$  ns.

Ps was formed with  $\sim 40\%$  efficiency implanting positrons at 2.6 keV into a porous silica film provided by Lizskay and co-workers [23]. Figure 4 shows single shot lifetime spectra generated following the implantation of positron pulses into the silica film with and without resonant light; the change in the Ps decay rate caused by the laser is evident at  $\sim 100$  ns. The arrival time of the laser pulse is indicated by a pulse generated in a plastic scintillator that had a pinhole in the light shield to admit a small fraction of the laser light that was split off from the main beam.

Positronium excited to a triplet 2P state in a magnetic field (in this case 900 G) will be mixed with a singlet state so that after radiative decay back to the (singlet) ground state the lifetime will be greatly reduced [24], as was observed by Ziock et al. in an experiment similar to the one described here [25]. The signal generated via the quenching effect would obviously be absent in a field free region, but it is also possible to observe 2P states via photoionization (532 nm light from a doubled YAG laser is suitable for this) [26]. In this case the liberated positrons will interact with the chamber wall or the target material and annihilate, generating a gamma ray pulse that is correlated with the ionization laser pulse. One can also use sub-ionization threshold light to excite Ps atoms from the 2P state to higher so-called Rydberg states, which when done with intense bursts of Ps is of interest to some experimenters wishing to create antihydrogen [27].



**Figure 5.** Fractional difference between laser on and laser off curves shown for two different laser wavelengths. The lifetime spectra were normalised to be equal just before the laser pulse arrives to account for small drifts, and the area of the difference curves were taken to be representative of the number of Ps atoms excited by the laser.

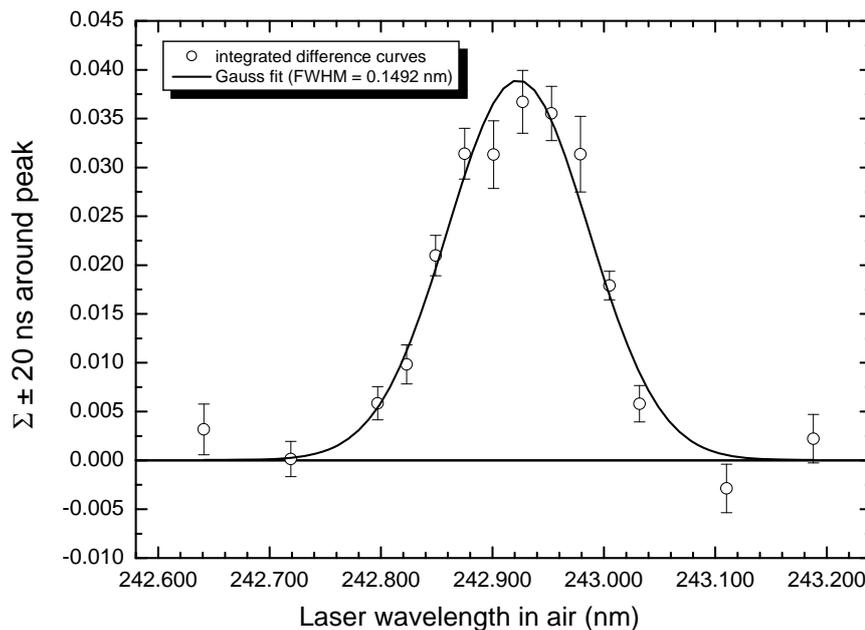
The data were analysed to determine the fractional difference between the laser on and laser off lifetime spectra,  $\Delta f_L = (\text{laser on} - \text{laser off})/\text{laser off}$ . The areas of the residual lifetime spectra  $\pm 20$  ns around the peak were then taken to be the signal, which is proportional to the number of excited Ps atoms created by the laser pulse. Two examples of these difference curves are shown in figure 5. By making such measurements at different laser frequencies we may then measure the linewidth of the transition, which in the present case is dominated by the Doppler width of the near thermal Ps. The lineshape measured in this way is shown in figure 6.

The laser linewidth used to perform these measurements was approximately 0.03 nm which was a compromise between resolution and signal (this corresponds to around 20% of the expected thermal linewidth). Accurately interpreting the observed lineshape will require some modelling since the Ps will have a velocity distribution that depends on the relative time delay between the laser and positron pulses as well as the distance from the sample where the excitations occur. Moreover, the

measurement is made in one dimension that is perpendicular to the beam axis, and so the Ps angular distribution must also be taken into account. As a first approximation the measured lineshape would be the full width at half maximum (FWHM) of a Maxwell-Boltzmann distribution

$$\Delta\lambda \approx \frac{\lambda_0}{c} 2\sqrt{2\ln 2} \sqrt{\frac{kT}{m_{Ps}}} . \quad (1)$$

Here  $\lambda_0$  is the resonant wavelength,  $m_{Ps}$  is the positronium mass,  $T$  is the Ps temperature and other symbols have their usual meanings. A fit to the data yields a FWHM of 0.149 nm which, according to eqn (1) implies a Ps temperature of  $\sim 800$  K, or  $\sim 70$  meV. This is consistent with the expected rate of cooling in the porous medium. Ps emitted from the bulk silica material initially has an energy of  $\sim 1$  eV or more [28, 29] and cooling occurs via collisions with the internal pore surfaces. Lower Ps temperatures would result if the positron implantation energy were increased since more collisions occur before emission into the vacuum. These data demonstrate that we are able to produce clouds of excited state Ps atoms, and that we are able to measure Doppler broadened linewidths to a few % precision. The lifetime spectra were averaged over 10 shots but this was done primarily because the laser power was not constant and could vary by as much as a factor of two from shot to shot. Absent this power fluctuation the counting statistics should be sufficient to measure linewidths to a similar accuracy in a series of single shot measurements.



**Figure 6.** 1S-2P Doppler broadened lineshape. Positronium atoms emitted from the porous silica are cooled via collisions to near thermal temperatures, giving rise to the Doppler broadened width of this line. The exact lineshape will also depend on the details of the Ps thermal distribution, the angular distribution sampled by the probe beam and the relative timing between the positron and laser pulses.

Planned experiments with  $Ps_2$  will be carried out in much the same way as that described above. In this case the fragments resulting from ionization will be detected using a micro-channel plate (mcp) detector [30], greatly reducing the background and increasing the sensitivity of the measurement as compared to simply observing the annihilation radiation. This arrangement will also make it possible to detect either positive or negative Ps ions [13] since ionized  $Ps_2$  will result in one of these states, and the mcp can be biased to detect either of them.

#### 4. Summary and outlook

The production of molecular positronium in porous films was an encouraging start, but to go further it will be necessary to use a positron beam with a higher density and to perform laser spectroscopy. We have redesigned our experimental arrangement to generate an electrostatic beam that may be remoderated, which is expected to increase the beam density by at least an order of magnitude. A laser system has been developed that will provide  $\sim 100$  mJ/pulses of tunable 243 or 251 nm light, suitable for conducting resonant excitation-ionization studies of  $\text{Ps}_2$ . The next round of experiments will be performed using an Al(111) [3] crystal as the target, although a single crystal quartz surface may also be suitable [31].  $\text{Ps}_2$  emitted into vacuum will be probed and ionized using appropriate lasers, and the resulting fragments detected with high efficiency using a mcp detector. This will provide an unambiguous signal of  $\text{Ps}_2$  formation and will allow us to study this unique molecule and obtain data that may then be compared to theory.

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