Excited states of positronium in electric fields

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Funding from NSF, EPSRC, Leverhulme Trust, ERC
Positronium: easy to make, available in many labs, but relatively few experiments involving excited states produced by laser excitation: WHY??

There are some difficulties when it comes to experimentation with Ps atoms:

• There are never enough of them (efficiency)
• They don’t live very long (lifetime)
• They tend to go all over the place (divergence)
• They are low mass and hence generally fast (Doppler)
1989: Surko Leventhal & Passner demonstrate positron accumulation. Now it is possible to obtain high intensity pulsed beams without large facility based devices. This changed opened the door to new experiments (see plenary on Monday)

Cliff Surko and Al Passner with the first buffer gas positron trap at Bell Labs (in Allen Mills’ lab)
Two-stage Surko-type buffer gas trap:
10^5 positrons/pulse
1 Hz
Pulse width 3 ns FWHM
Spot size 4 mm FWHM
Very well suited to Ps excitation with standard pulsed lasers

1s-2p excitation requires about 5 nJ/cm^2 in a 5 ns pulse *per natural linewidth*:

This would be great, except that we have a huge amount of Doppler broadening so we actually need to cover 500 GHz instead of 50 MHz, and thus we need 10,000 times more laser intensity

<table>
<thead>
<tr>
<th>Transition</th>
<th>$e^+$ source</th>
<th>Laser</th>
<th>Comments</th>
<th>year</th>
<th>Ref.</th>
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</thead>
<tbody>
<tr>
<td>$1S_1 \rightarrow 2P_x$</td>
<td>RI/gas</td>
<td>L</td>
<td>No signal observed</td>
<td>1954</td>
<td>[94]</td>
</tr>
<tr>
<td>$1S_1 \rightarrow 2P_y$</td>
<td>RI/gas</td>
<td>L</td>
<td>Signal not statistically significant</td>
<td>1974</td>
<td>[95]</td>
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<tr>
<td>$1S_1 \rightarrow 2S_1$</td>
<td>MB</td>
<td>P</td>
<td>First unambiguous Ps excitation signal</td>
<td>1982</td>
<td>[96]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>MB</td>
<td>P</td>
<td>Precision measurement (12 ppb)</td>
<td>1984</td>
<td>[97]</td>
</tr>
<tr>
<td>$1S_1 \rightarrow 2P_0$</td>
<td>L</td>
<td>P</td>
<td>Ps Lyman-$\alpha$</td>
<td>1990</td>
<td>[98]</td>
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<tr>
<td>$1S \rightarrow 2P \rightarrow n^2S/n^2D$</td>
<td>L</td>
<td>P</td>
<td>First Rydberg Ps</td>
<td>1990</td>
<td>[99]</td>
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<tr>
<td>$1S_1 \rightarrow 2S_1$</td>
<td>M</td>
<td>P</td>
<td>Photoionization by 532 nm laser</td>
<td>1991</td>
<td>[100]</td>
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<tr>
<td>$1S_1 \rightarrow 2S_1$</td>
<td>M</td>
<td>CW</td>
<td>Precision measurement (2.6 ppb)</td>
<td>1993</td>
<td>[101,102]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_0$</td>
<td>ST</td>
<td>P</td>
<td>Doppler spectroscopy of Ps</td>
<td>2010</td>
<td>[103,104]</td>
</tr>
<tr>
<td>$1S_1 \rightarrow 2P_0$</td>
<td>ST</td>
<td>P</td>
<td>PsX formation on semiconductors</td>
<td>2011</td>
<td>[105-107]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>ST</td>
<td>P</td>
<td>Ps cavity shift and narrowing</td>
<td>2011</td>
<td>[108]</td>
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<tr>
<td>$Ps^- \rightarrow Ps + e^-$</td>
<td>L</td>
<td>P</td>
<td>Photodetachment of Ps$^-$ ions</td>
<td>2011</td>
<td>[109]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>ST</td>
<td>P</td>
<td>Excited Ps in Paschen back regime</td>
<td>2011</td>
<td>[110]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>ST</td>
<td>P</td>
<td>Efficient Rydberg Ps production</td>
<td>2012</td>
<td>[111]</td>
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<tr>
<td>$Ps \rightarrow Ps + e^- + e^+$</td>
<td>ST</td>
<td>P</td>
<td>Molecular Ps spectroscopy</td>
<td>2012</td>
<td>[112]</td>
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<tr>
<td>$Ps^- \rightarrow Ps + e^-$</td>
<td>L</td>
<td>P</td>
<td>Energy tunable Ps beam</td>
<td>2011</td>
<td>[113]</td>
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<td>$1S_1 \rightarrow 2P_y \rightarrow 1S_0$</td>
<td>ST</td>
<td>P</td>
<td>First optical Ps hyperfine measurement</td>
<td>2012</td>
<td>[114]</td>
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<tr>
<td>$1S_0 \rightarrow 2P \rightarrow n^2S/n^2D$</td>
<td>ST</td>
<td>P</td>
<td>Doppler corrected Balmer spectroscopy</td>
<td>2014</td>
<td>[115]</td>
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<tr>
<td>$1S_1 \rightarrow 2S_1$</td>
<td>RI</td>
<td>CW</td>
<td>Annihilation of $2S_1$ states</td>
<td>2015</td>
<td>[116]</td>
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<tr>
<td>$1S \rightarrow 2P \rightarrow n^2S/n^2D$</td>
<td>ST</td>
<td>P</td>
<td>High-precision Rydberg TOF</td>
<td>2015</td>
<td>[117]</td>
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<tr>
<td>$1S \rightarrow 2P \rightarrow n^2S/n^2D$</td>
<td>ST</td>
<td>P</td>
<td>Selective production of Ps Stark states</td>
<td>2015</td>
<td>[118]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>ST</td>
<td>P</td>
<td>Laser enhanced Ps TOF</td>
<td>2015</td>
<td>[119]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>ST</td>
<td>P</td>
<td>Ps Lyman-$\alpha$</td>
<td>2015</td>
<td>[120]</td>
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<tr>
<td>$1S_1 \rightarrow 2P_2$</td>
<td>ST</td>
<td>P</td>
<td>Ps cooling in transmission targets</td>
<td>2015</td>
<td>[121]</td>
</tr>
<tr>
<td>$1S \rightarrow 2P \rightarrow 1S_0$</td>
<td>ST</td>
<td>P</td>
<td>Stark and Zeeman mixing of n = 2 Ps</td>
<td>2015</td>
<td>[122,123]</td>
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<tr>
<td>$1S \rightarrow 2P \rightarrow 30^5S/30^5D$</td>
<td>ST</td>
<td>P</td>
<td>Ps Stark-states</td>
<td>2016</td>
<td>[124]</td>
</tr>
<tr>
<td>$Ps^- \rightarrow Ps + e^-$</td>
<td>L</td>
<td>P</td>
<td>Ps$^-$ ion shape resonance</td>
<td>2016</td>
<td>[125]</td>
</tr>
<tr>
<td>$1S \rightarrow 2P \rightarrow n^2S/n^2D$</td>
<td>ST</td>
<td>P</td>
<td>Measurement of fluorescence lifetimes</td>
<td>2016</td>
<td>[126]</td>
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<tr>
<td>$1S \rightarrow 3P \rightarrow n^2S/n^2D$</td>
<td>ST</td>
<td>P</td>
<td>$n = 3$/Rydberg excitation</td>
<td>2016</td>
<td>[127]</td>
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<tr>
<td>$1S \rightarrow 2P \rightarrow 10^5S/10^5D$</td>
<td>ST</td>
<td>P</td>
<td>Electrostatic guiding of Ps</td>
<td>2016</td>
<td>[128]</td>
</tr>
<tr>
<td>$1S \rightarrow 2P \rightarrow 30^5S/30^5D$</td>
<td>ST</td>
<td>P</td>
<td>Angle resolved Ps spectroscopy</td>
<td>2016</td>
<td>[129]</td>
</tr>
<tr>
<td>$1S \rightarrow 2P \rightarrow n^2S/n^2D$</td>
<td>ST</td>
<td>P</td>
<td>Rydberg Ps MCP detector</td>
<td>2016</td>
<td>[130]</td>
</tr>
<tr>
<td>$1S \rightarrow 2S_1 \rightarrow 2S_1$</td>
<td>ST</td>
<td>P</td>
<td>1-photon production of $2S_1$ atoms</td>
<td>2017</td>
<td>[131]</td>
</tr>
<tr>
<td>$1S \rightarrow 2P \rightarrow 14^5S/14^5D$</td>
<td>ST</td>
<td>P</td>
<td>Electrostatic velocity selection of Ps</td>
<td>2017</td>
<td></td>
</tr>
<tr>
<td>$1S \rightarrow 2P \rightarrow 32^5S/32^5D$</td>
<td>ST</td>
<td>P</td>
<td>Electrostatic Ps beam focusing</td>
<td>2017</td>
<td></td>
</tr>
</tbody>
</table>

Table 1. Time line of optical excitation experiments involving positronium atoms, ions and molecules. This list is not abridged and includes all instances of Ps excitation with laser light to date. We have not indicated the J values for some transitions to Rydberg levels as they are fully Stark mixed. The positron sources used are Radioactive isotopes and gas moderation and Ps production (RI/gas), mono-energetic beams based on radioactive isotopes (RI), magnetic bottle (MB) traps, linear (L) or microtron (M) accelerator based beams, or source-based Surko traps (ST). The light sources used are lasers (L), or pulsed (P) or continuous wave (CW) lasers.
Excitation to $n = 2$ requires UV light ($\sim 243$ nm) that can be produced quite easily. However, it also requires a large bandwidth because of the Doppler broadening. This means that in general one does not excite specific ($n = 2$) states, since the laser bandwidth covers the entire 2P manifold.

Need to use Doppler free methods (e.g., saturated absorption spectroscopy) and a narrower laser bandwidth to see individual states: another example of why we really need colder Ps.
Laser induced changes in the amount of Positronium present may be observed via annihilation radiation using single-shot lifetime spectra:

![Diagram of experimental setup](image1.png)

![Graph showing lifetime spectra](image2.png)

![Graph showing ionisation signal vs. laser wavelength](image3.png)
Lutetium yttrium oxyorthosilicate (LYSO) based detectors are much better for single shot lifetime measurements than PWO

\( n = 45 \) Ps is field ionized immediately after production

\( n = 12 \) Ps hits the chamber wall around 500 ns after production

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Zeeman mixing between singlet and triplet states can give rise to large changes in lifetime. This effect can be enhanced significantly by Stark mixing, so that with suitable laser polarization MQ can occur even in a relatively weak magnetic field (Stark-Enhanced Magnetic quenching).
Laser polarization matters as it affects which states are populated.

(a) $\Delta M_J = 0$

\[
\begin{array}{cccc}
2^3P_2 & -2 & -1 & 0 & +1 & +2 \\
2^3P_1 & -1 & 0 & +1 & & \\
2^3P_0 & 0 & & & \\
1^3S_1 & M_J = -1 & 0 & +1 & &
\end{array}
\]

(b) $|\Delta M_J| = 1$

\[
\begin{array}{cccc}
2^3P_2 & -2 & -1 & 0 & +1 & +2 \\
2^3P_1 & -1 & 0 & +1 & & \\
2^3P_0 & 0 & & & \\
1^3S_1 & M_J = -1 & 0 & +1 & &
\end{array}
\]
Stark enhanced magnetic quenching:

The peaks at 585 V/cm correspond to the magnetic mixing at the (field-free) avoided crossings, where the singlet and triplet character of the Stark and Zeeman mixed eigenstate is maximal.
Ps in the Paschen-Back regime

Ps energy levels scale with $1/n^3$ so it is roughly 8 times easier to mix singlets and triplets after excitation to $n = 2$.

This means it would be in principle be possible to laser cool Ps in a strong magnetic field.
Saturated absorption spectroscopy: measurement of Hyperfine interval

Crossover peak can only be seen as a result of magnetic quenching

Not a very good measurement, partly because of laser width and low number of Ps with the correct speed
2S production by single-photon excitation

(a) Energy/\hbar (GHz)

(b) $3S_1$ Character fraction

(c) $3S_1$ mean lifetime (ns)

Electric field (kV/cm)

(a) $n = 8$, $E_{\text{ex}} = 0$ kV/cm

(b) $n = 2$, $E_{\text{ex}} = 2.23$ kV/cm

(c) $n = 2$, $E_{\text{ex}} = 1.11$ kV/cm
Stark states \((n = 11)\)

Increasing the electric field in the excitation region splits the line (Stark effect). Limited spectral resolution means large fields are needed to see different Stark states. The asymmetry comes mostly from the Stark effect on the intermediate \(n = 2\) states.
We can make any states we want, but we cannot resolve them above $n = 30$ or so because of the large bandwidth of our laser and Doppler broadening effects.
Guiding Rydberg Ps with an electrostatic quadrupole field

The quadrupole guide is at ground potential when the positron pulse passes through. It is turned on (in 10 ns) after the Ps is excited.

Detector A measures single-shot lifetime data.

Detectors B and C measure single events.
Stark-state dependent transport

(a)

ring
-1.6 kV
target
-2 kV
6 mm
UV and IR lasers

(b)

Normalized $S_{\gamma}$ (arb.)

$E = 667 \text{ V cm}^{-1}$
100 GHz gaussian

Guide off
Guide on (0.25 kV)
Guide on (0.5 kV)
Guide on (1.0 kV)
Guide on (2.0 kV)

Background Subtracted Countrate (arb.)

$\lambda$ (nm)
Selecting the low field seeking states by tuning the excitation laser is more efficient (since we don’t excite atoms to unguidable high field seeking states.

This is not always possible: you have to apply a field to split the line enough so that you can access one part of the Stark manifold. That means the width has to be on the order of the laser bandwidth and/or the Doppler width of the transition. For us this means states above \( n = 15 \) or so start to overlap.
Curved guide: useful for getting the Ps beam off the positron trap axis, and also acts as a velocity selector.
microwave spectroscopy of $n = 2$ Ps with Rydberg Helium field characterization

Can use He as a way to map out the magnetic field in the Ps excitation region, and for E field cancellation. We will also perform optical transitions ($n = 25$ -2) to measure Ps Rydberg constant: ultimate goal is to get to part in $10^{12}$
Helium tests: 2-photon excitation scheme:

In order to achieve better state selection and avoid the limited overlap of the laser with the Doppler broadened transitions we will attempt to produce Rydberg Ps via a two-photon transition. Initial test experiments done with metastable Helium:

Circular polarization. No field applied in the excitation region. Approximately 5 GHz wide dye laser, with beam focussed to a spot of size 0.5 mm by 0.1 mm (1/e waist). Pulse energy ~ 10 mJ in 5 ns.

Directions of future work:

• Find improved sources of cold Ps
• Better resolution of Rydberg states (pulse amplification)
• Microwave spectroscopy of $n = 2$ levels (fine structure)
• Improve control of Ps motion with electrostatic fields
• Ps control with time varying fields: deceleration/focusing
• Creation of long-lived circular states (with microwaves)
• Microwave spectroscopy of Rydberg levels (Rydberg constant)
• Optical transition ($n = 25$ to $n = 2$: Better Rydberg constant)
• Ps free-fall/interferometry gravity measurements.....

Thank you for your attention
What might a Ps Rydberg gravity experiment look like?

- What n states will be optimal?
- How will the circular states be produced?
- How cold must the walls be? (BBR)
- Will patch fields deflect the beam?
- How long does the flight tube have to be?
- How does this mirror/lens work?
- Will the “atom-optics” work on circular states?
- Can very slow atoms be detected without affecting their positions on a micron scale?
- What Ps converter will work at low temp?
- Needs a point e+ source
- Zero or low B field
- Ps has to be very cold