Positronium Physics

D. J. Murtagh

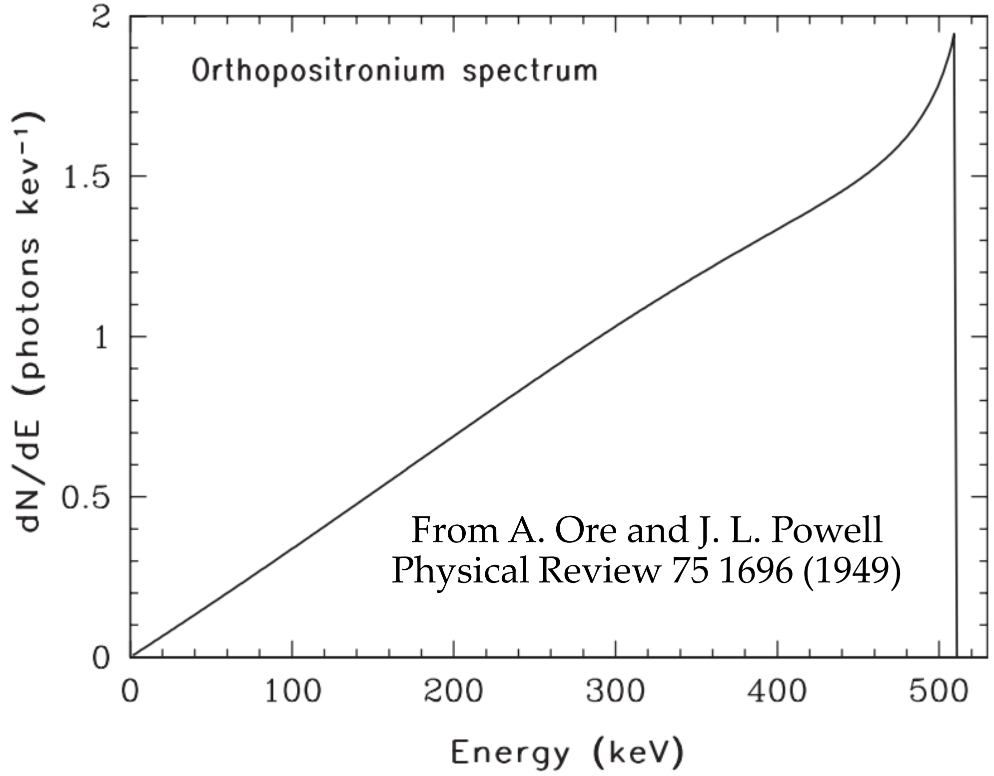
For the AVA School on Precision Studies
24th March 2020

Overview

- In this lecture, I will introduce the field of Positronium physics from a historical perspective
- I will cover what you need to gain an understanding of positronium enabling you to go forward and explore this field for yourself
- At the end I will link to reviews which you can use as resources going forward

Start with the end

- Positronium is the end point of the life of most positrons in vacuua, the direct annihilation cross-section is generally considered to be negligible [1]
- Observations of gamma rays around positron annihilation peak (511 keV) in the interstellar medium find a positronium fraction of ~0.90 [2]
- For some perspective approximately 10 billion kg of positrons annihilate in the Milky Way every second 90% of which is via Ps formation [3]



From A. Ore and J. L. Powell
Physical Review 75 1696 (1949)

1. Van Reeth, P. Laricchia, G. and Humberston., J. W *Physica Scripta*, **71** C9 (2005).
2. T. Siegert, R. Diehl, G. Khachatryan, M. G. H. Krause, F. Guglielmetti, J. Greiner, A. W. Strong and X. Zhang *Astronomy and Astrophysics* **586** A84 (2016)
3. Prantzos, N., Boehm, C., Bykov, A.M., Diehl, R., Ferrière, K., Guessoum, N., Jean, P., Knoedlseder, J., Marcowith, A., Moskalenko, I.V., Strong, A., Weidenspointner, G., *Rev. Mod. Phys.* **83**, 1001 (2011)

Prediction

- Dirac (1930) and Weyl (1931) predict that a particle with the same mass as the electron but with a positive charge should exist
- Anderson (1932a,b 1933)observes said positive electron in a cloud chamber
- Blackett and Occhialini (1933) confirm the charge to mass ratio
- Mohorovičić (1934) predicted the existence of a bound state of the positive and negative electron

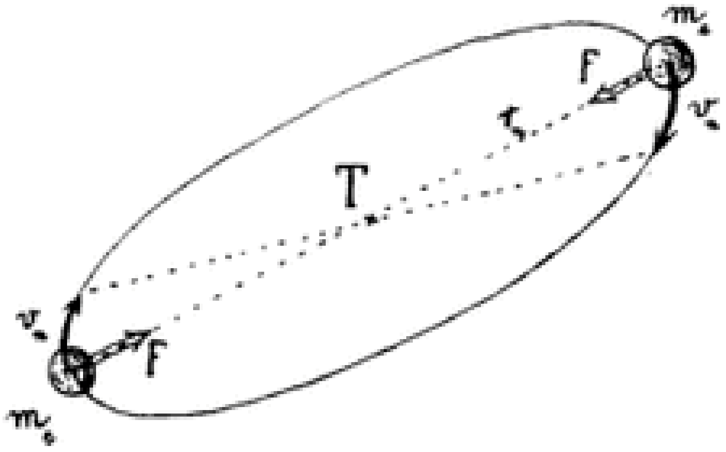


Fig. 1.

1. Dirac P. A. M. *Proc. Roy. Soc* **126** **360** (1930a)
2. Weyl R. N. *Grouppentheorie and Quantenmechanil* **2nd ed.** P234 (1931)
3. Anderson C. D. *Phys Rev* **41** 405 (1932a)
4. Anderson C. D. *Science* **76** 238 (1932b)
5. Anderson C. D. *Phys Rev* **43** 491 (1933)
6. Blackett P. M. S. and Occhilialini G. P. S *Proc. Roy. Soc A* **139** 699 (1933)
7. Mohorovicic S. *Astrn. Nachr.* **235** 94 (1934)

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Electrum
~~Positronium~~ Physics

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Properties of Electrum

- Rydberg ‘constant’ $\frac{M}{M+m}R_\infty = \frac{1}{2}R_\infty$
- Broad structure should follow $\frac{1}{\lambda} = \frac{1}{2}R_\infty\left(\frac{1}{n^2} - \frac{1}{k^2}\right)$
- Ground state binding energy or 6.8 eV
    - Half that of hydrogen which follows directly from the reduced mass

Prediction

- The spectroscopic structure was calculated by Ruark (1945)
- The binding energy and lifetime by Wheeler (1946) also considered were the positive and negative ions of Ps and Ps_2 - P^{+-} P^{++-} P^{--+} and P^{++--} in his own notation

Positronium

ARTHUR E. RUARK*
*Naval Research Laboratory, Office of Research and Inventions, Anacostia, D. C.***
November 13, 1945

POLYELECTRONS*

BY

JOHN ARCHIBALD WHEELER†

1. Ruark A. E. *Phys. Rev* **68** 278 (1945)
2. Wheeler J. A. *Ann. N. Y. A ad. Sci.* **48 291** (1946)

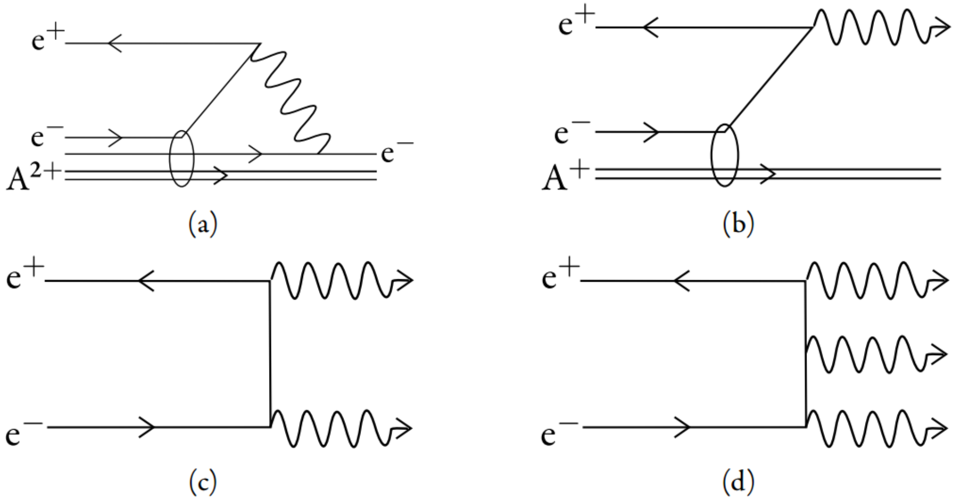
Properties of Positronium

- Lyman – α of 243 nm
- Blamer
  - α (3→2) of 1312.6 nm
  - β (4→2) of 972.2 nm
  - δ (5→2) of 868.0 nm
  - Limit of 729 nm
- Bohr radius is twice that of hydrogen
- Due to the larger magnetic moment of the positron than the proton the structure has a triplet and singlet state
- It has a finite lifetime against annihilation which should be calculated
- The existence of annihilation channels will cause broadening of states

1. Ruark A. E. *Phys. Rev* **68** 278 (1945)

Properties of Positronium

- a[1] and b[2] show photonless and one photon annihilation, these require the presence of a third body in this case an atom
- c[3] and d[4] show two and three gamma annihilation
- Annihilation probability proportional to α^m where α is the fine structure constant and m the number of photons



1. Palathingal J. C., Asoka-Kumar P., Lynn K. G., Posada and Wu X. Y., Phys Rev. Lett. **67** 3491 (1991)
2. Klemperer O., Proc. Camb. *Philos. Soc.* **30** 347 (1934)
3. Chang T., Li Q., Wang Y and Li Y. in "Positron Annihilation" eds. P. G. Coleman, S. C. Sharman and L. M. Diana (North Holland: Amsterdam) p32 (1982)
4. Adachi S., Chiba M., Hirose T., Hagayama S., Nakamitsi Y., Sato T. and Yamada T. Phys. Rev. Lett **65** 2634 (1990)

Properties of Positronium

- Ground state lifetime
  - Singlet state : 125 ps
  - Triplet state 142 ns
- The number of annihilation quanta is determined by charge parity conservation
  - Photon $P_c = (-1)^n$
  - Ps system $P_c = (-1)^{L+S}$
  - $(-1)^n = (-1)^{L+S}$

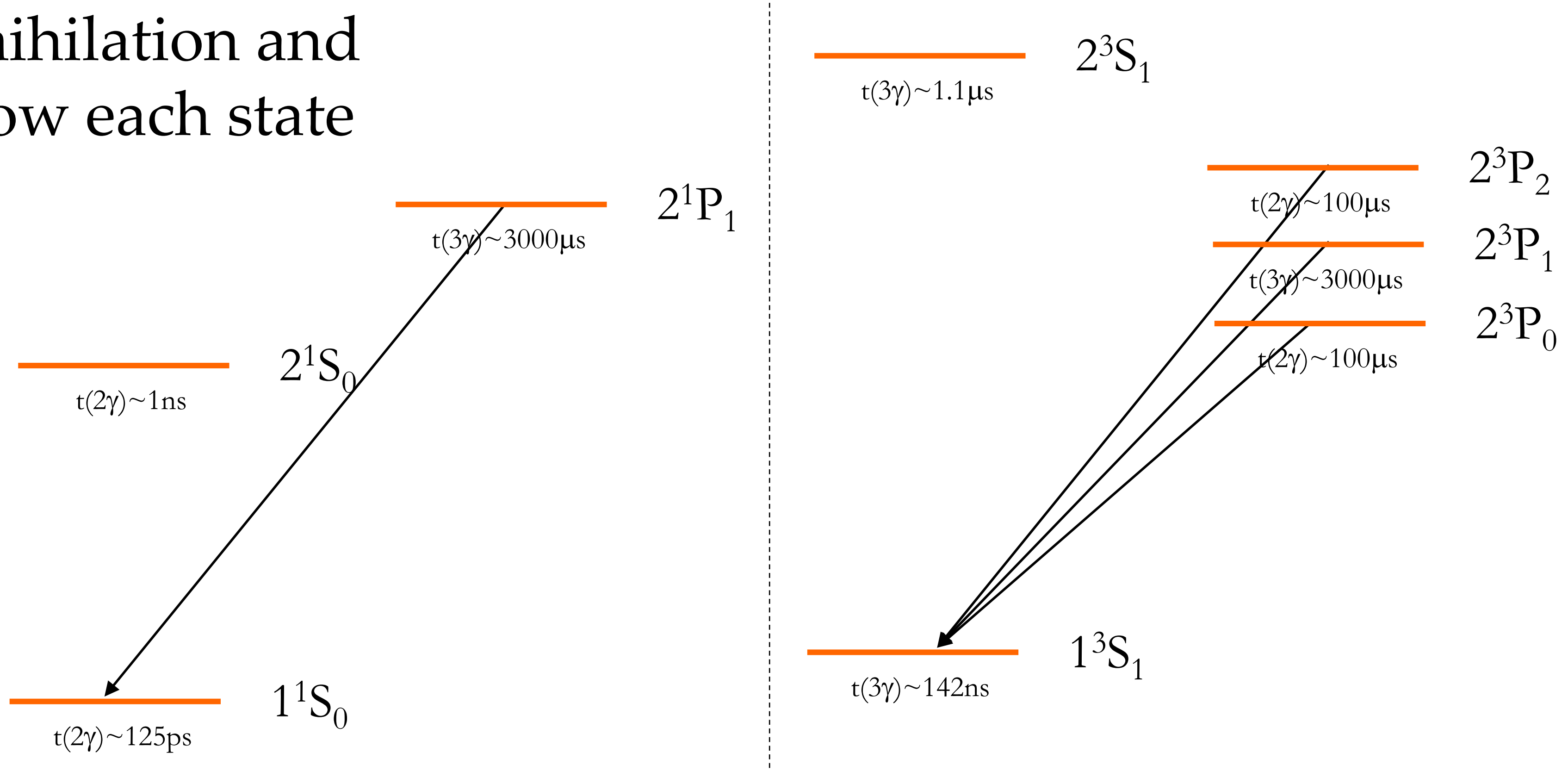
$$\Gamma_{3\gamma}(n_{Ps},{}^3S_1) = \frac{2}{9\pi}(\pi^2-9)\frac{mc^2}{\hbar}\frac{\alpha^6}{n_{Ps}^3}$$

$$\Gamma_{2\gamma}(n_{Ps},{}^1S_0) = \frac{1}{2}\frac{mc^2}{\hbar}\frac{\alpha^5}{n_{Ps}^3}$$

1. A. Ore and J. L. Powell *Physical Review* **75** 1696 (1949)

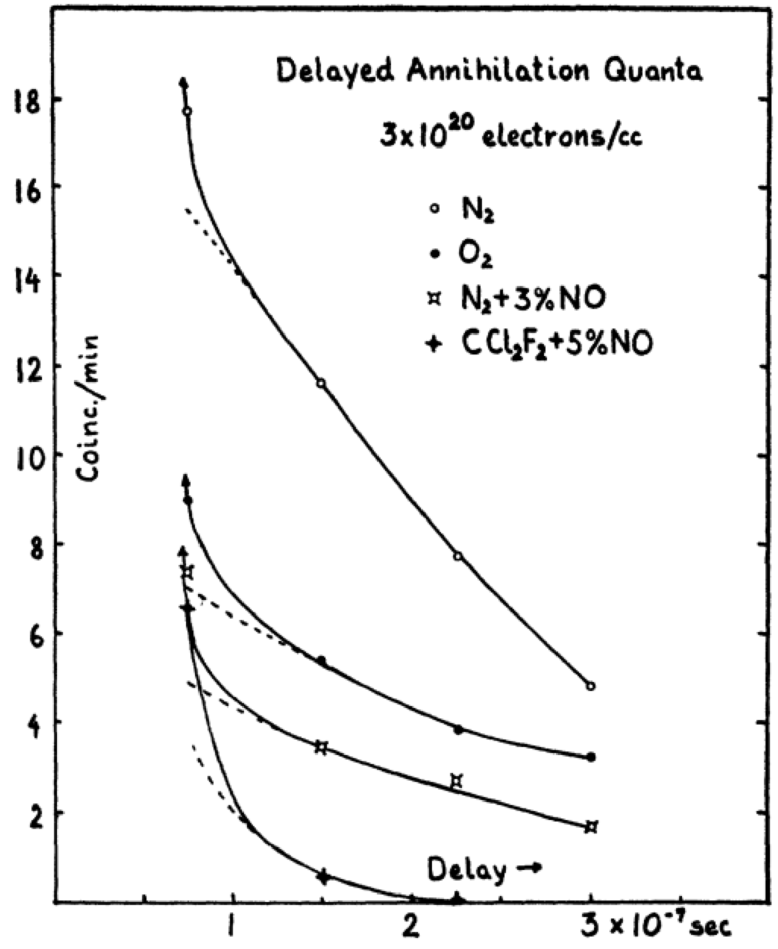
Properties of Positronium

- Energy level diagram for the n<3 manifold of Ps
  - Lifetimes against annihilation and transition shown below each state



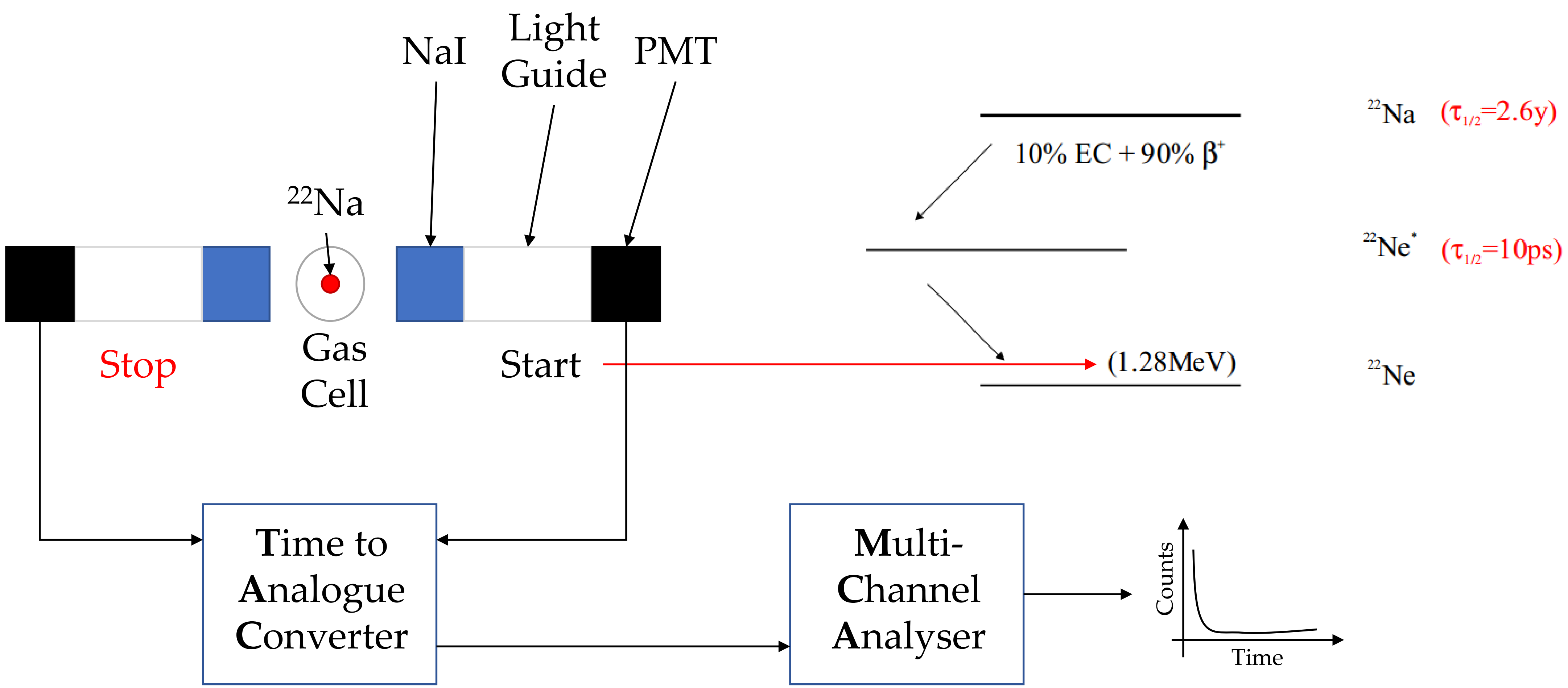
Observation

- The first experimental evidence for the existence of Ps came in 1951 in a paper titled : “Evidence for the formation of Positronium in Gases” by Deutsch



1. Deutsch M. *Phys. Rev.* **82** 455 (1951)

Positron annihilation lifetime

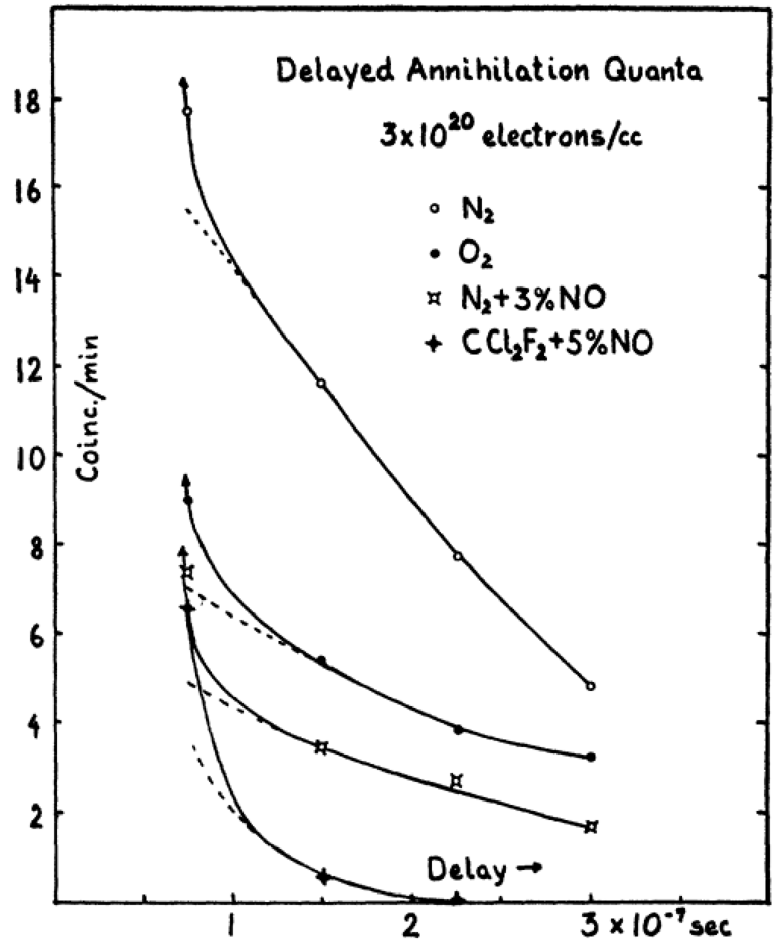


Positron annihilation lifetime

- Positron interactions with gas
  - Annihilation: $e^{+} + A \rightarrow A^{+} + 2\gamma$
  - Ps formation: $e^{+} + A \rightarrow A^{+} + Ps$
  - Direct ionization: $e^{+} + A \rightarrow A^{+} + e^{+} + e^{-}$
- Positron interactions with a metal
  - The positron will lose energy, find a defect and annihilate.

Properties of Ps

- What we see is the time difference between the 1.28 MeV photon from the β^+ decay of ^{22}Na and the annihilation quanta from ortho-Ps
- Mixing NO with the gas being studied enhances the probability of spin flipping the Ps



Summary: Properties of Ps

- Positronium is a hydrogenic quasi-stable atom
- Due to the larger magnetic moment of the positron than the proton the fine structure has a triplet and singlet state
- Ground state lifetime
  - Singlet state : 125 ps
  - Triplet state 142 ns
- The number of annihilation quanta is determined by charge parity conservation
  - Photon $P_c = (-1)^n$
  - Ps system $P_c = (-1)^{L+S}$

- Bohr radius is twice that of hydrogen
- Lyman – α of 234 nm
- Blamer
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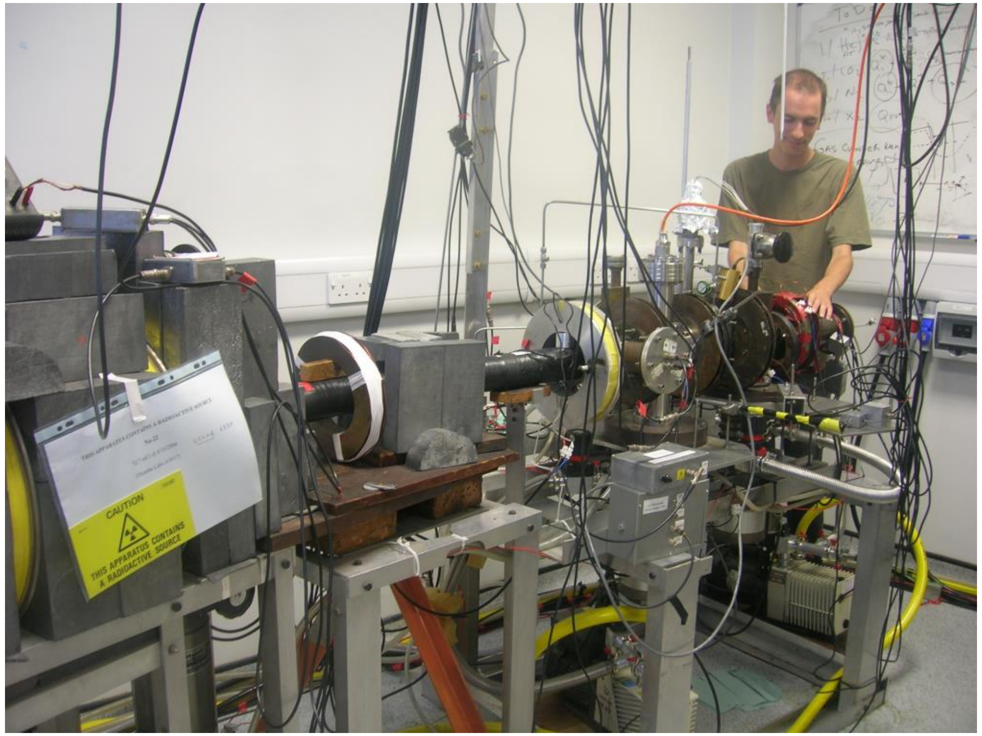
  - Limit of 729 nm

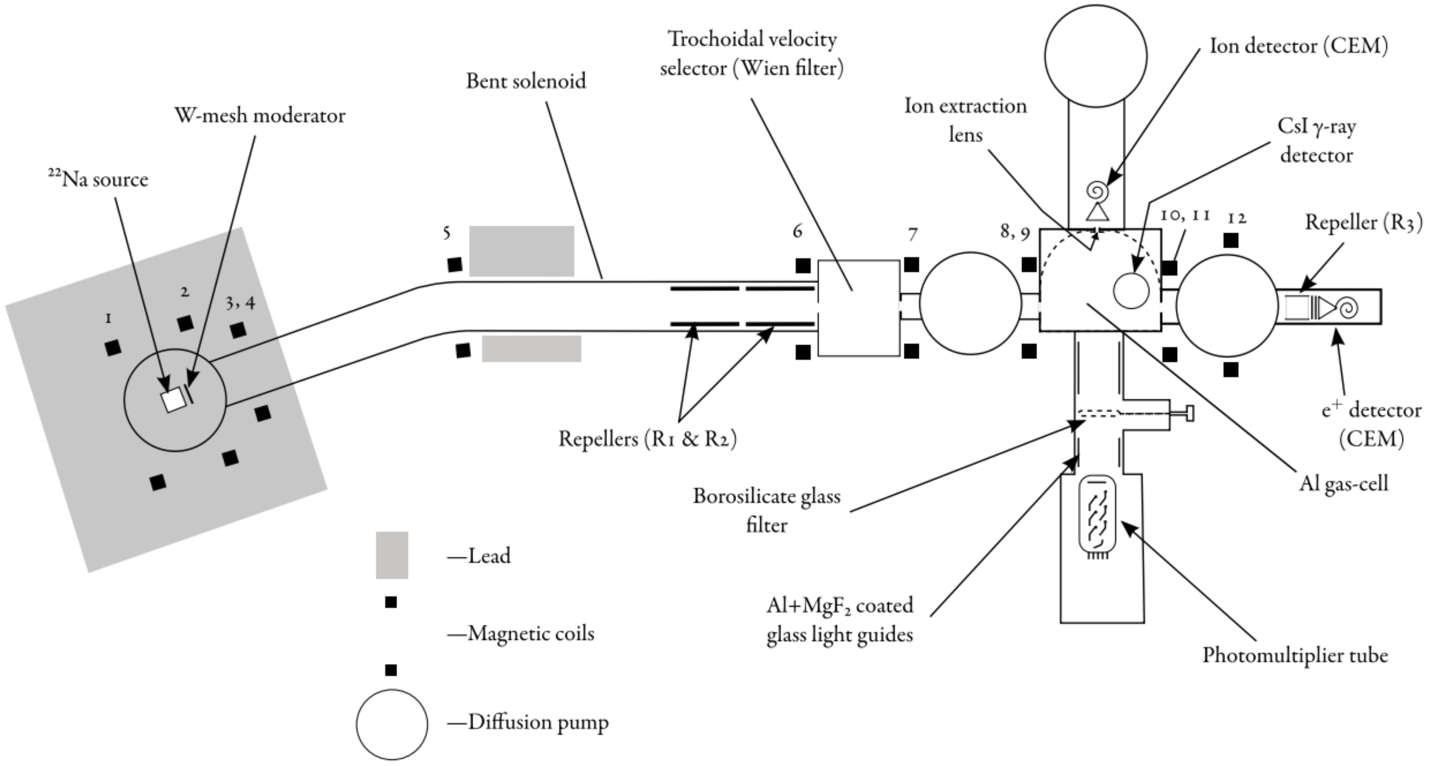
Production of Ps

- The lifetime method used by Deutsch continues to be used to this day in the form of **P**ositron **A**nnihilation **L**ifetime **S**pectroscopy (**PALS**[1])
- To discuss the production of Ps we have to briefly discuss the production of positron beams

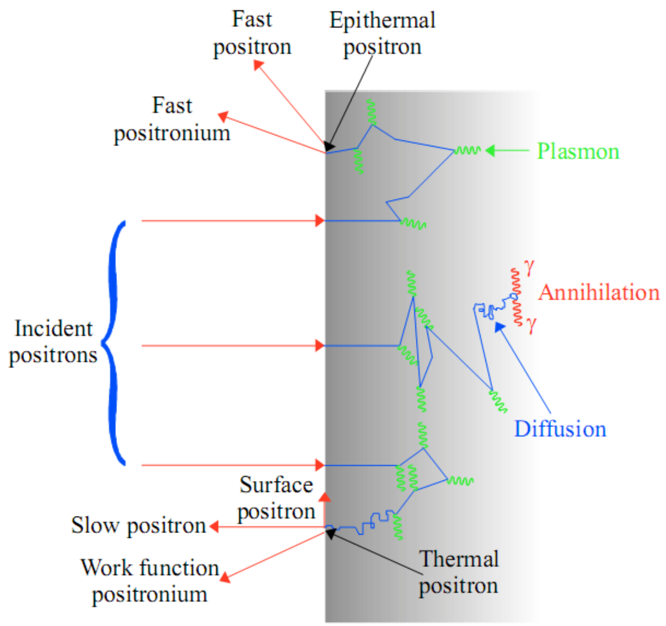


1. Dlubek, G. (2008). Positron Annihilation Spectroscopy. In Encyclopedia of Polymer Science and Technology, (Ed.).

Positron beam production

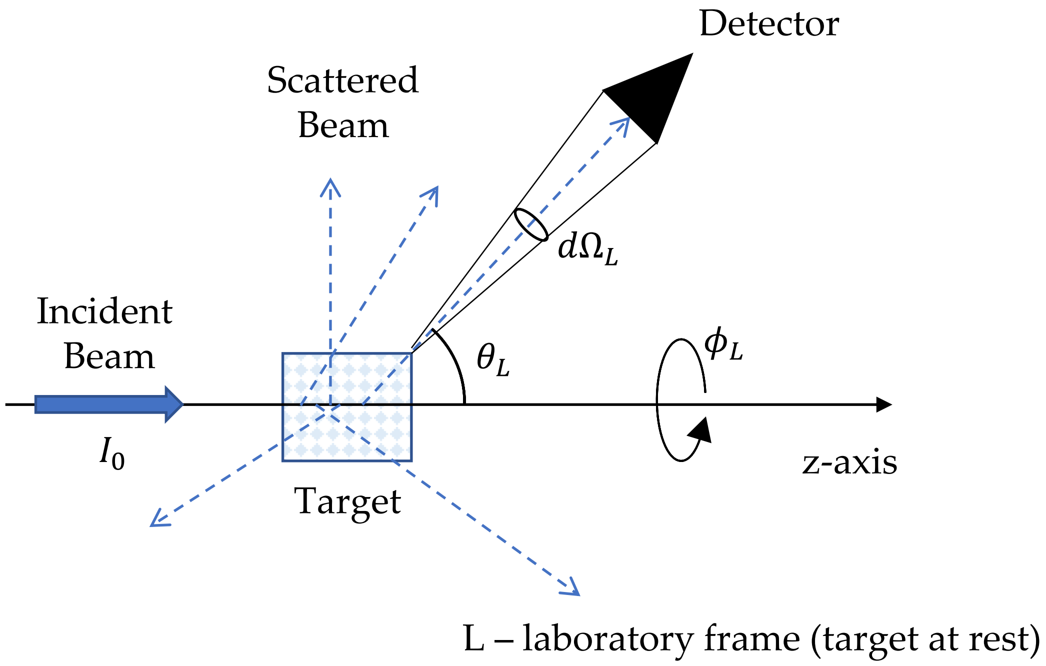


Moderation



Positronium Formation Cross-Section

- Once you can produce a beam of positrons and interact them with a gas of known pressure and cell length you can begin to measure cross-sections



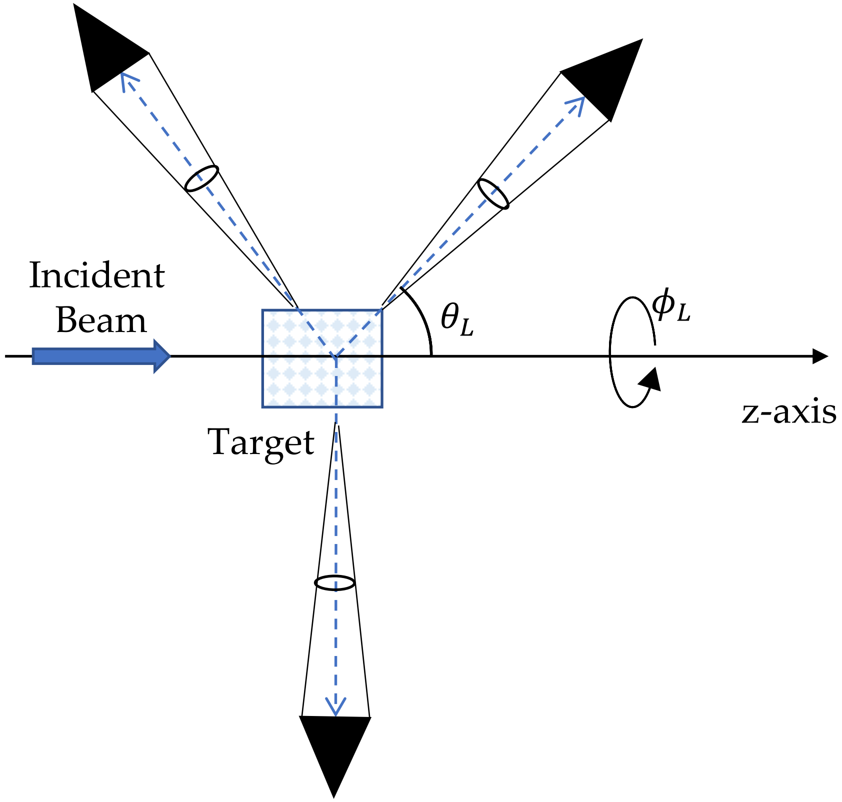
$$dN_{el} = I_0 \sigma_L^{el}(d\Omega_L) d\Omega_L$$

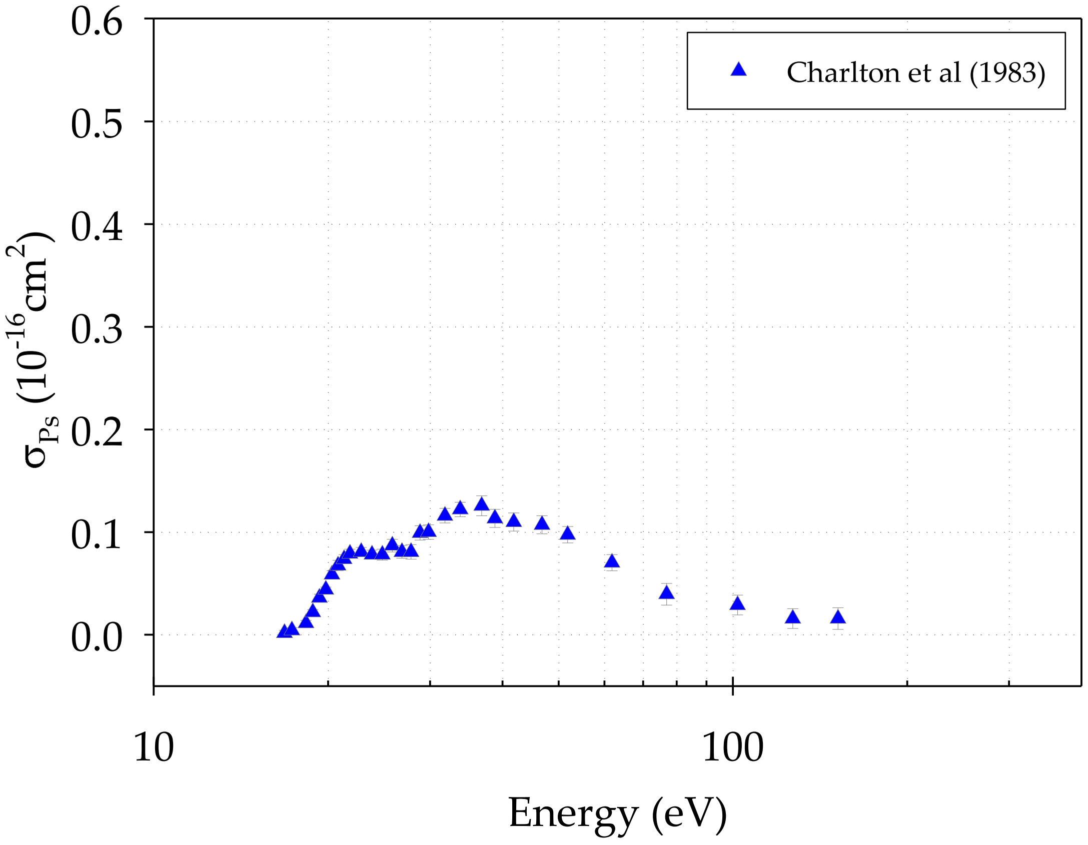
$$I_0 \sigma_L^{el}(d\Omega_L) d\Omega_L = I_0 \sigma_L^{el}(\theta_L, \phi_L) d\Omega_L$$

$$N_{el}^T = I_0 \sigma_L^{el}$$

Positronium Formation

- First measurements of Ps formation cross-sections relied on triple coincidences from o-Ps annihilation

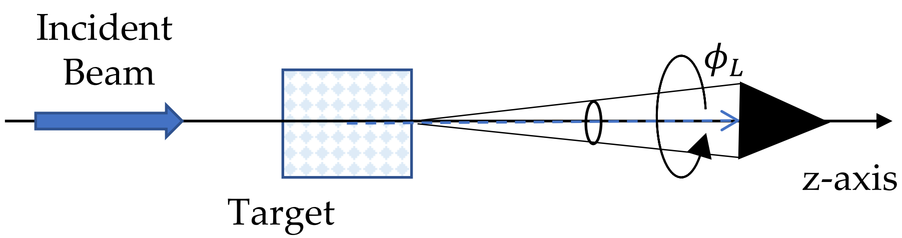


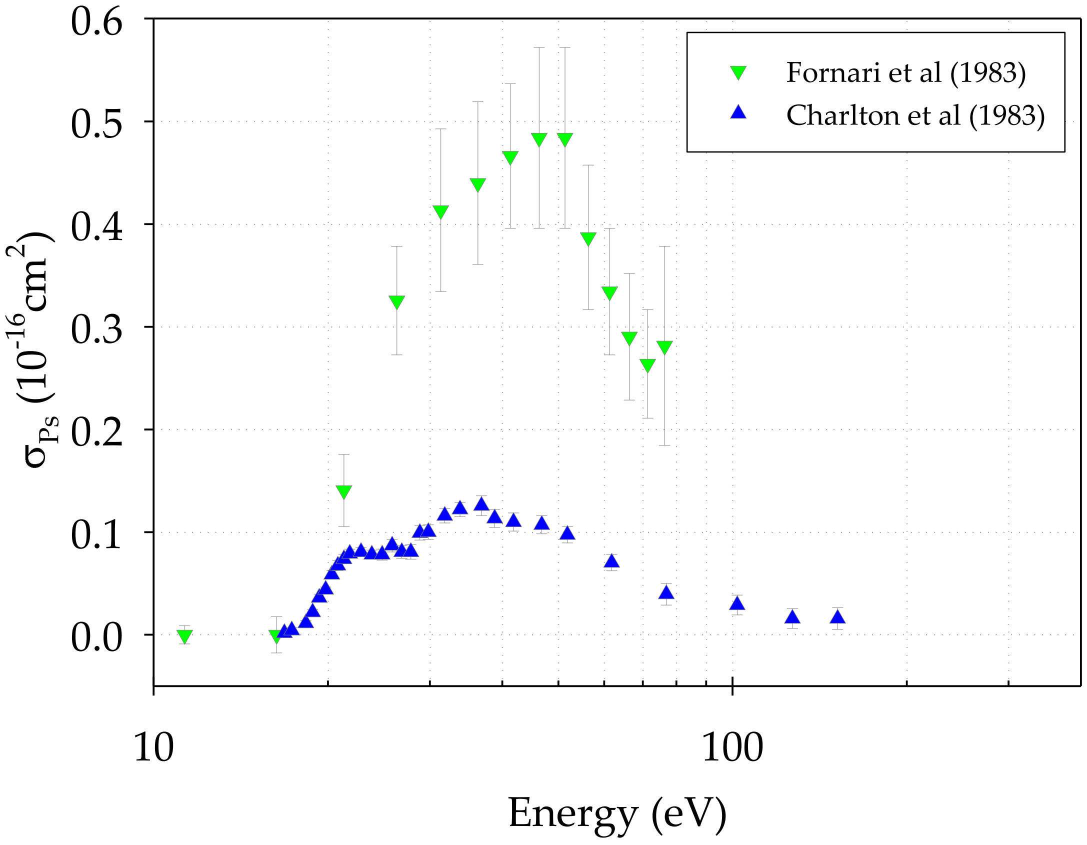


1. See D J Murtagh, M Szłuińska, J Moxom1, P Van Reeth and G Laricchia *J. Phys. B.* **38** 3857 (2005) and references there in

Positronium Formation

- The next method used was the loss of the positron in the final state
- The cross-sections are very different in magnitude - Charlton *et al* didn't know the Ps would exit the gas cell or hit the walls!

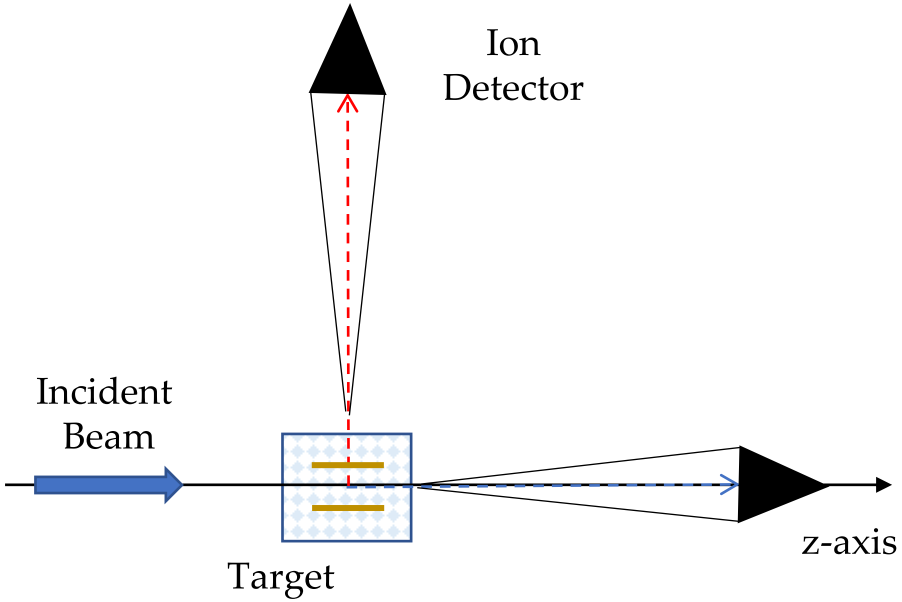


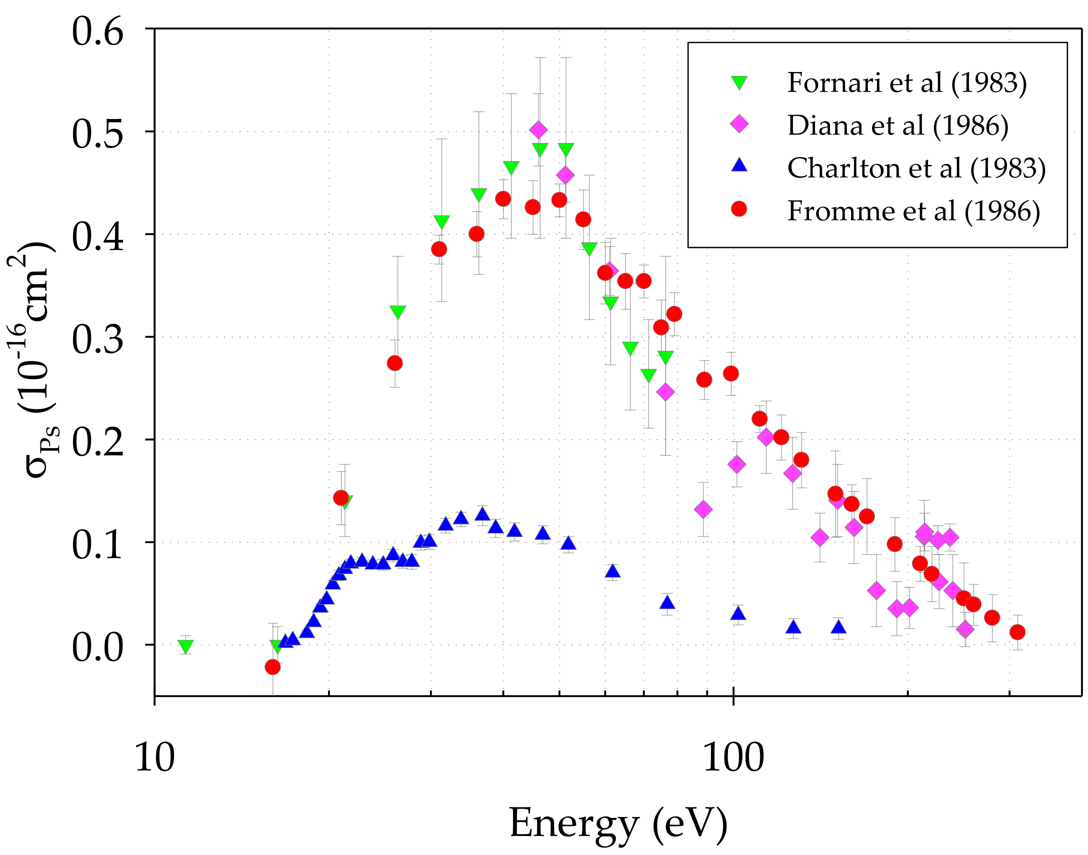


1. See D J Murtagh, M Szłuińska, J Moxom1, P Van Reeth and G Laricchia *J. Phys. B.* **38** 3857 (2005) and references there in

Positronium Formation

- Another idea was to measure the production of ions and positrons in coincidence. In the case of positronium an ion should be formed without an associated positron

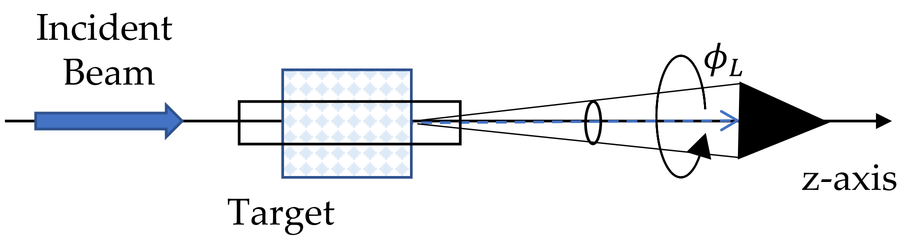


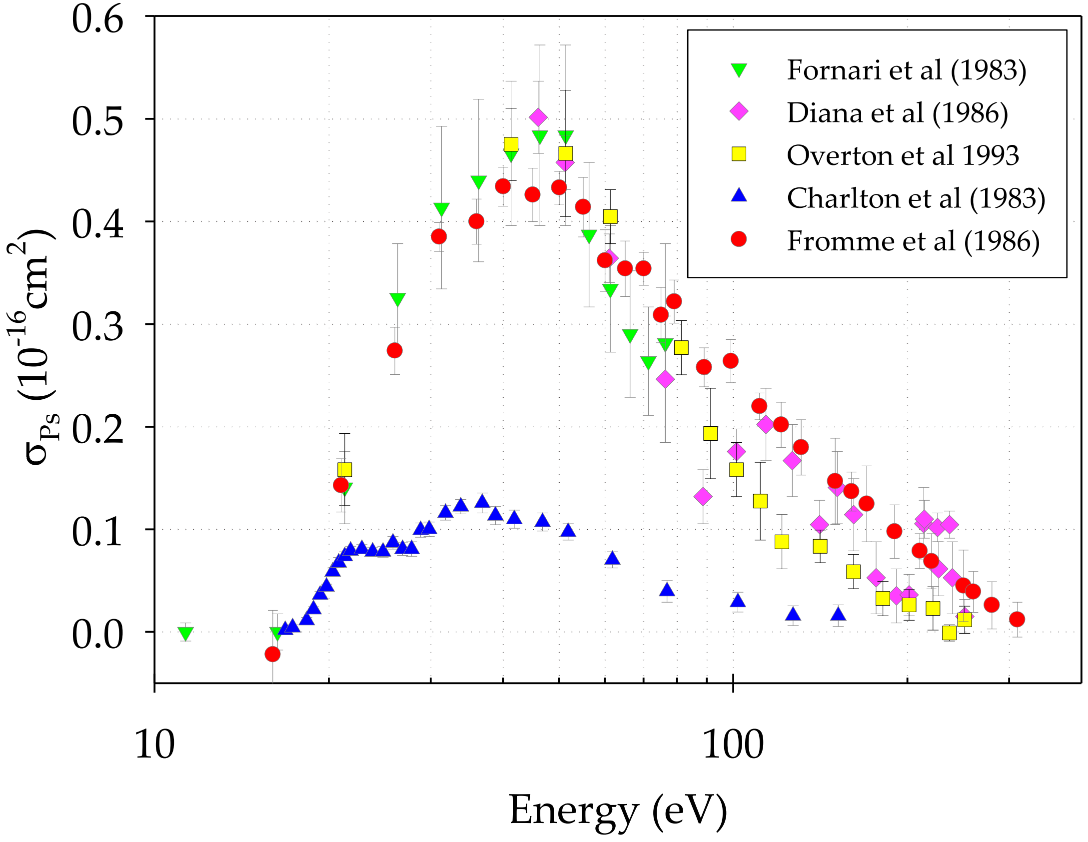


1. See D J Murtagh, M Szłuińska, J Moxom1, P Van Reeth and G Laricchia *J. Phys. B.* **38** 3857 (2005) and references there in

Positronium Formation

- Using the beam attenuation method special care has to be taken to confine scattered particles

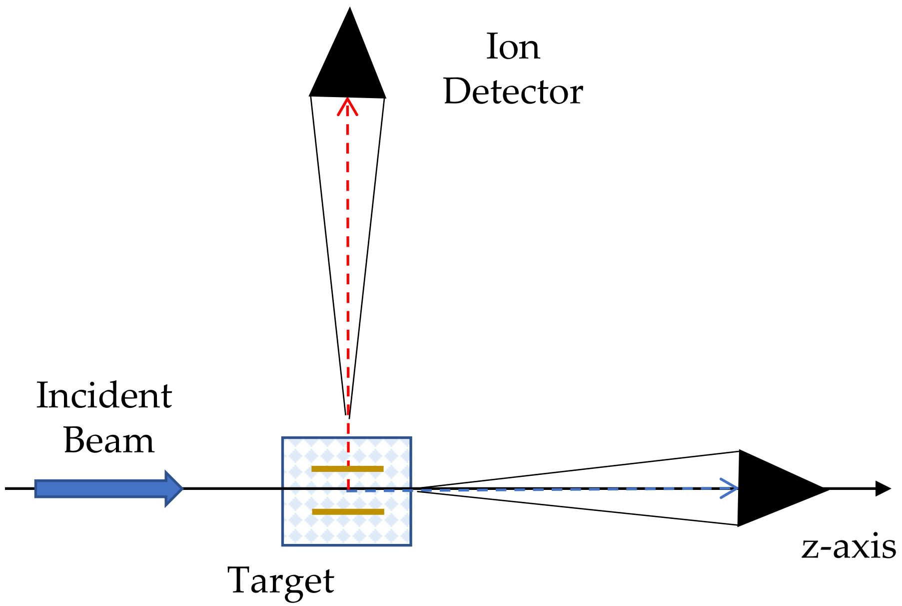


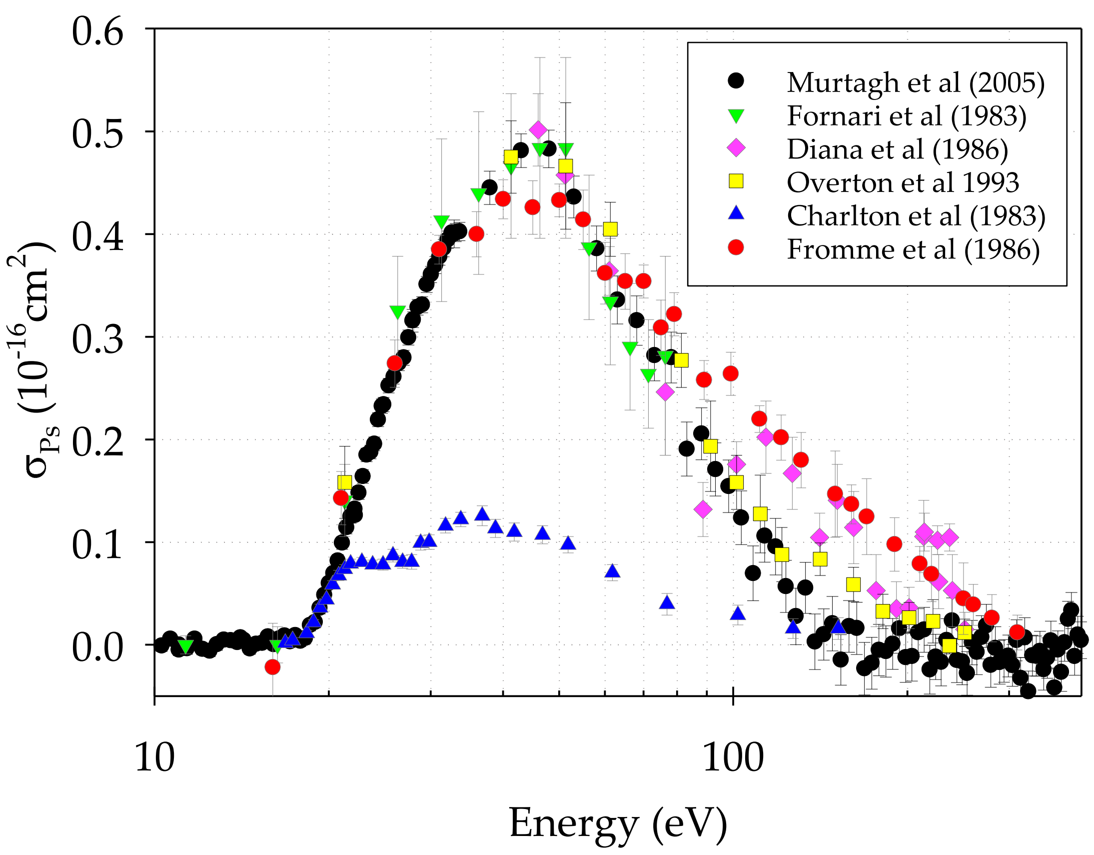


1. See D J Murtagh, M Szłuińska, J Moxom1, P Van Reeth and G Laricchia *J. Phys. B.* **38** 3857 (2005) and references there in

Positronium Formation

- $\sigma_i^t = \sigma_{Ps} + \sigma_i^+ + \Sigma\sigma_{HO}$
- $\sigma_{Ps} = \sigma_i^t - \sigma_i^+$





1. See D J Murtagh, M Szłuińska, J Moxom1, P Van Reeth and G Laricchia *J. Phys. B.* **38** 3857 (2005) and references there in

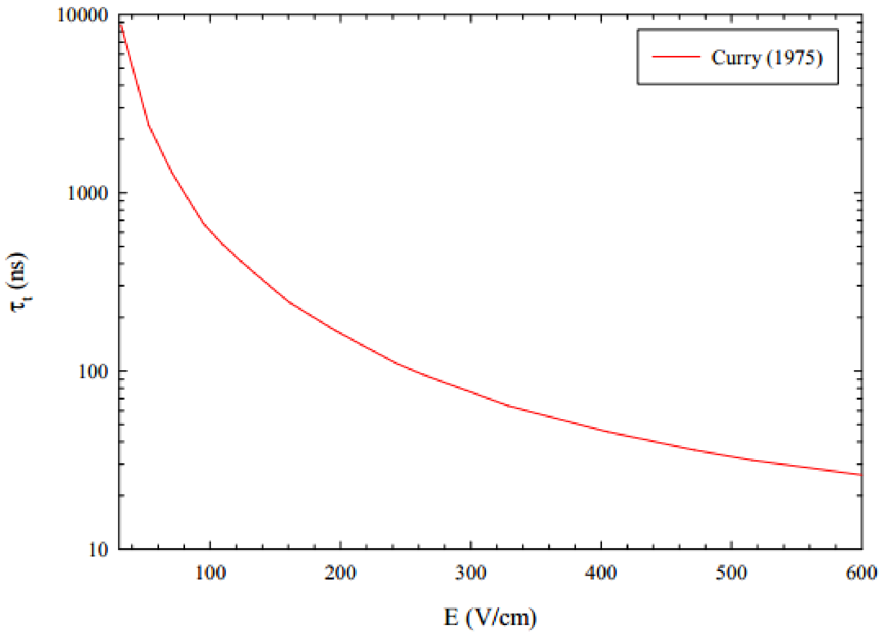
Positronium in external fields

- Stark effect → Mixes *l* states
- Zeeman effect → Mixes triplet and singlet states
- Motional Stark effect → Mixes *l* states however unlike the conventional Stark effect is due to the Ps motion transverse to a **B** field : $\vec{E}_{mse} = \gamma(\vec{v} \times \vec{B})$

$$\gamma = 1/\sqrt{1-\beta^2},\ \beta = v/c$$

$$\lambda_3' = \frac{\lambda_3 + y^2\lambda_0}{1+y^2} \qquad y = \frac{\sqrt{1+x^2}-1}{x} \qquad x = \frac{4\mu_B B}{\Delta W}$$

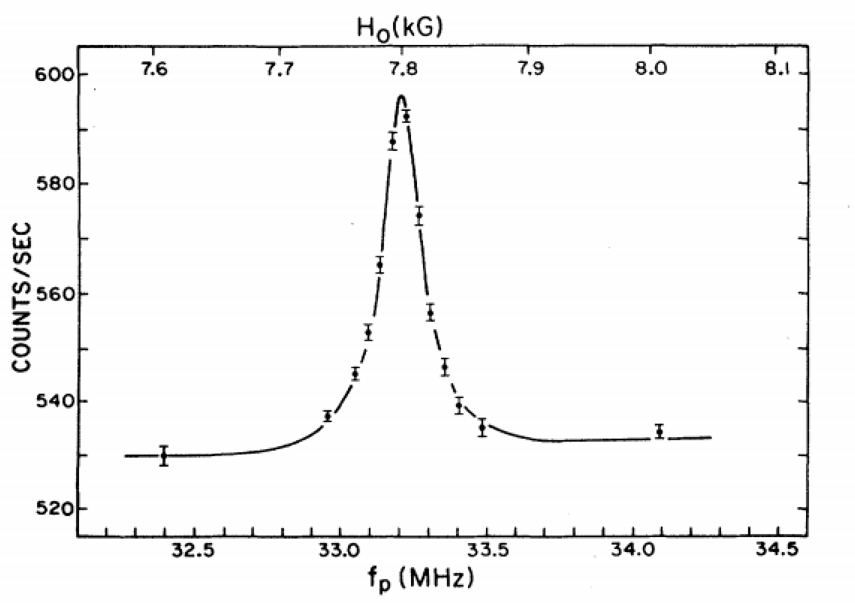
$$\Delta W = 25{,}423\ MHz$$

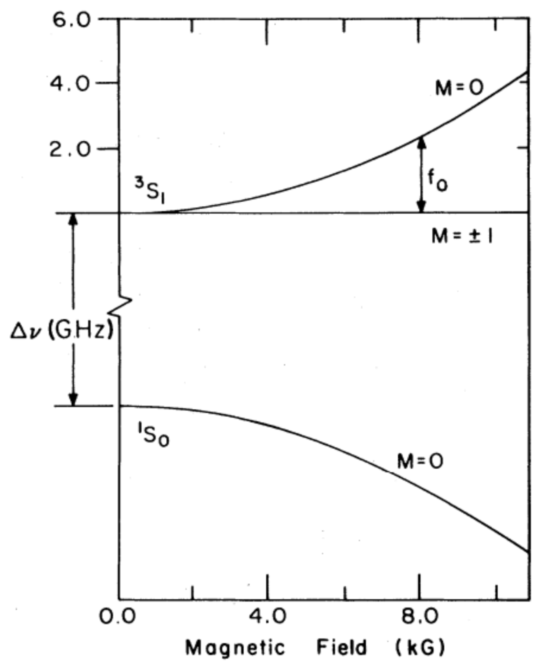


1. Consolati G. J. *Radioanalytical Nuc. Chem. Art.* **210** **273** (1996)
2. Curry S. M. *Phys. Rev. A.* **7** 447 (1975)

Hyperfine structure of Ps

- Triplet and Singlet $M_J = 0$ states are mixed in a magnetic field
- By inducing a transition from the triplet $M_J=\pm 1$ states to $M_J=0$ an increase in the 2γ annihilation rate can be observed



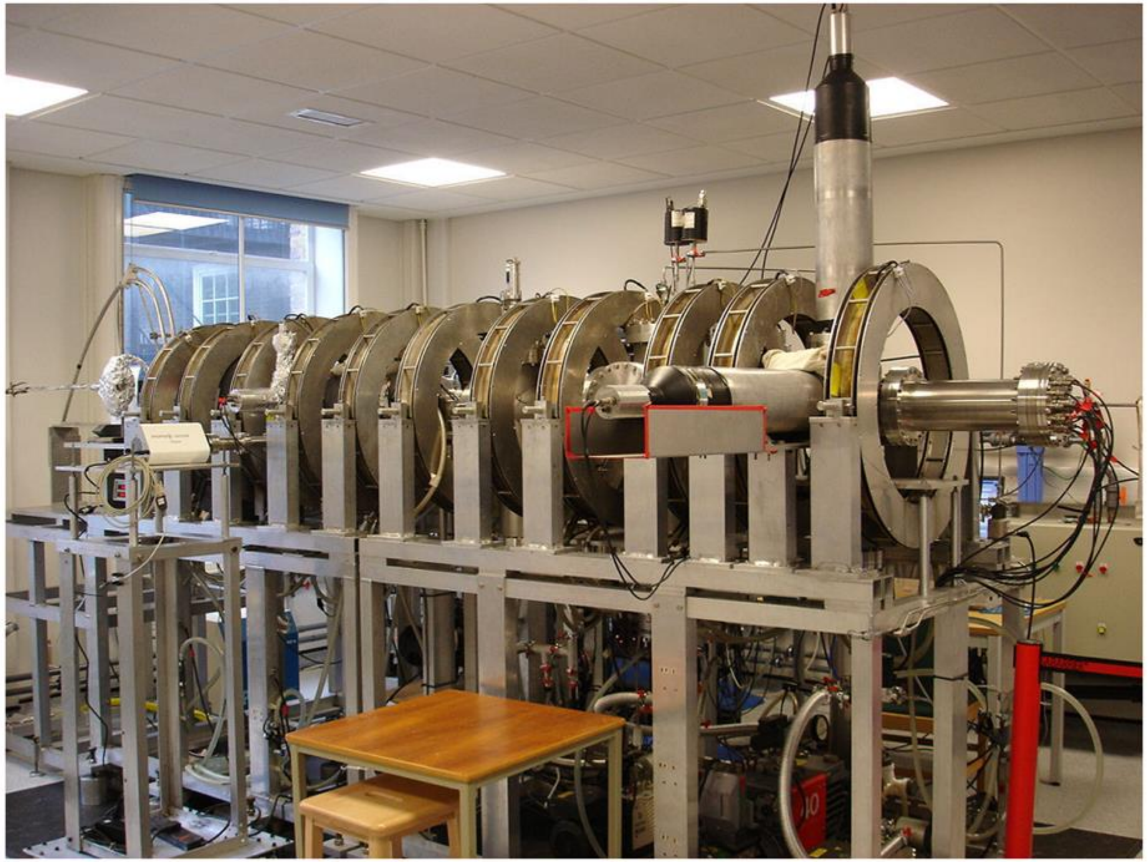


$$\Delta\nu = 203.38910(74)\ \mathrm{GHz}$$

1. M. W. Ritter, P. O. Egan, V. W. Hughes, and K. A. Woodle *Phys. Rev. A* **30**, 1331 (1984)

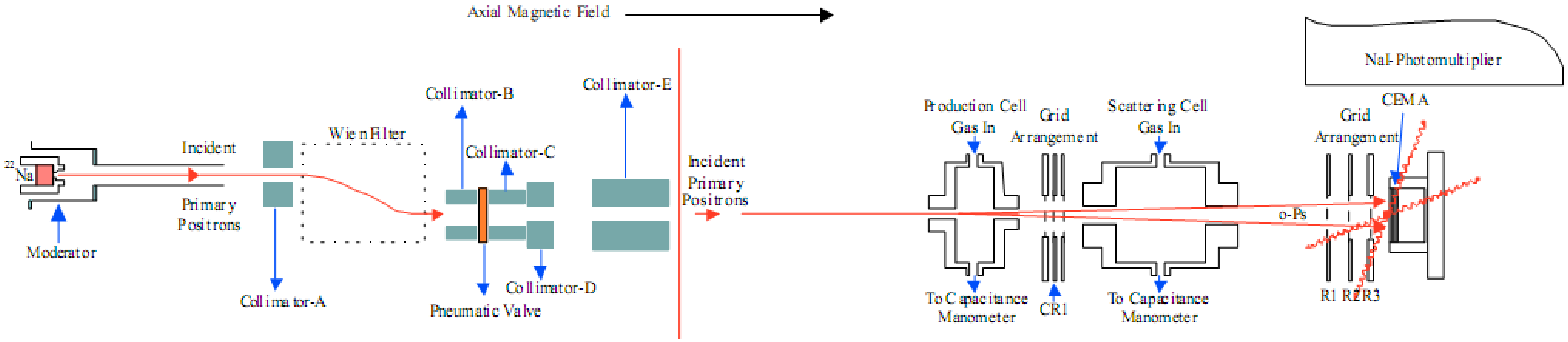
Positronium Beam

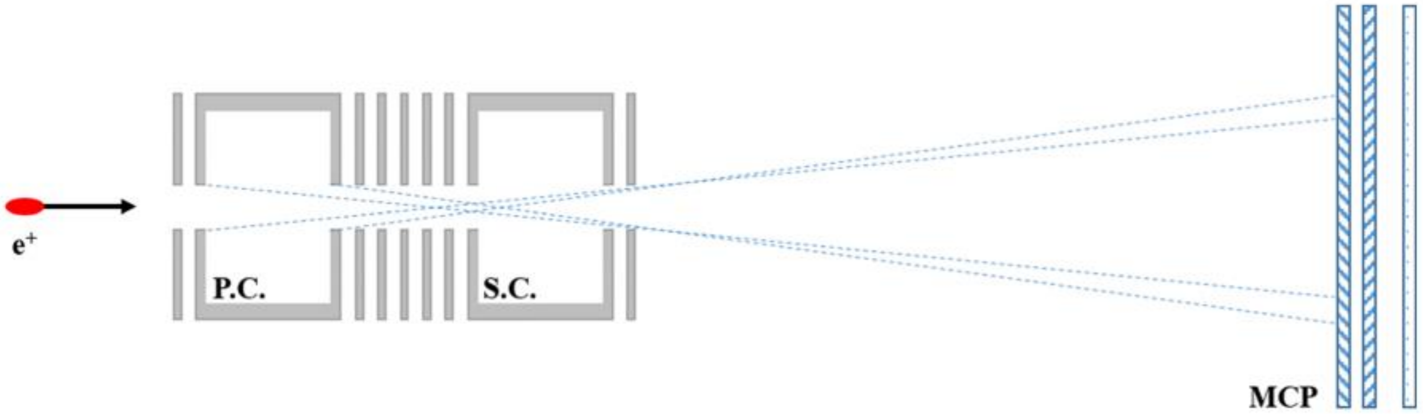
- We know it's possible to collide positrons with a gas target and produce Ps
- We know from the early work of Charlton that the Ps has energy to leave the production region
- The next step was to create a beam of positronium



1. N. Zafar, G. Laricchia & M. Charlton *Hyperfine Interactions* **89** 243 (1994)

Positronium Beam

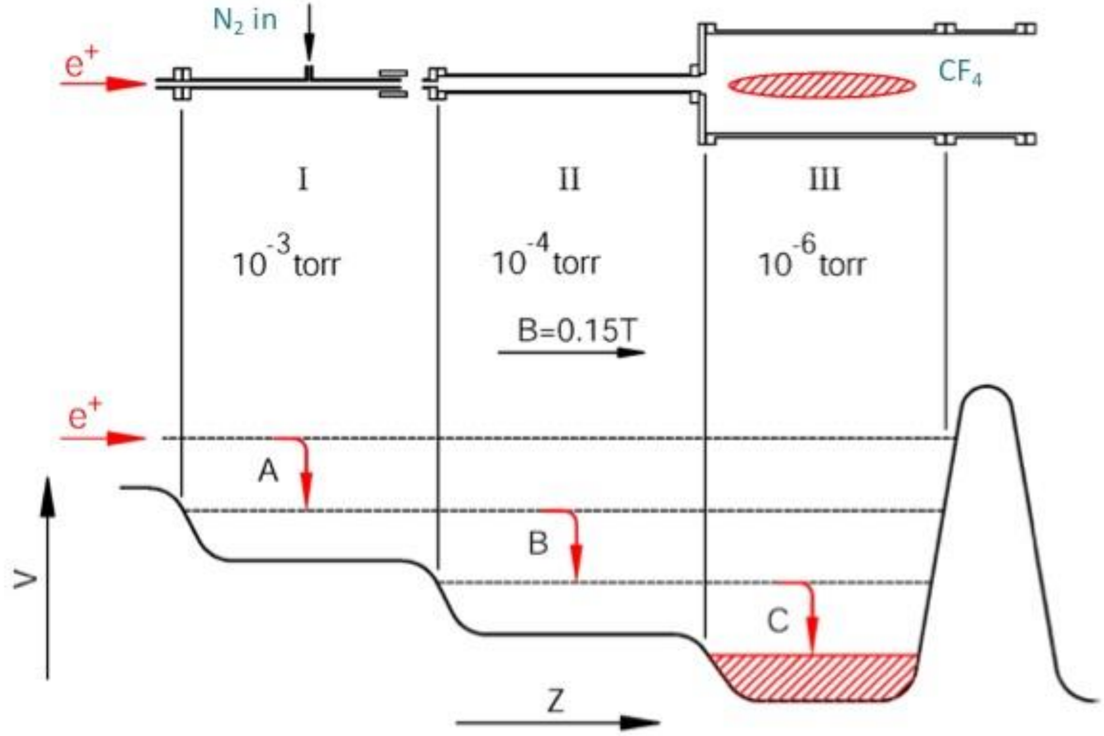




1. N. Zafar, G. Laricchia & M. Charlton *Hyperfine Interactions* **89** 243 (1994)
2. J. R. Machacek, S. J. Buckman, and J. P. Sullivan *Review of Scientific Instruments* **91**, 033311 (2020)

Buffer gas trap based beams

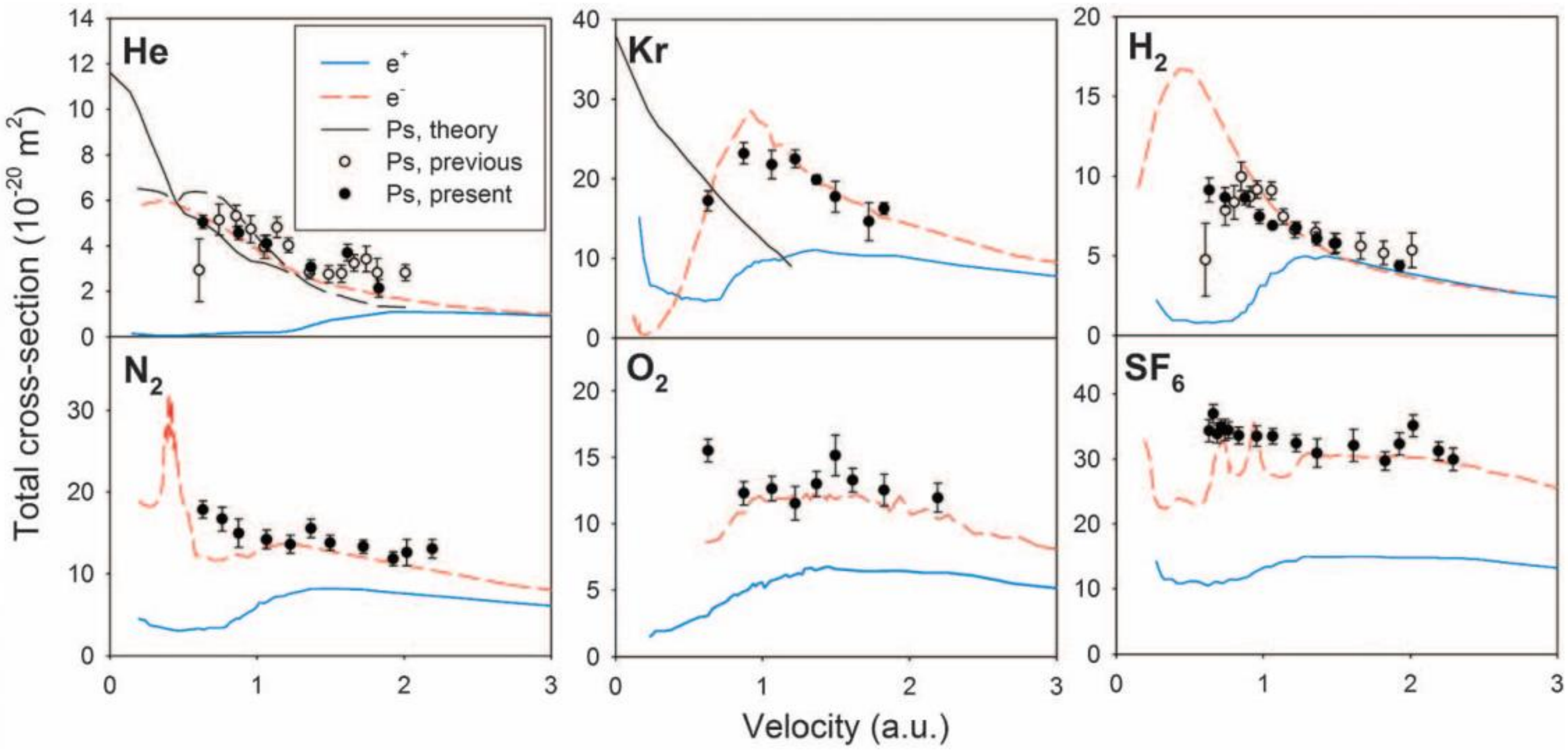
- The combination of a solid Ne moderated beam and N_2 buffer gas trap has enabled the production of high intensity, low energy spread, pulsed positrons beams in laboratories around the world



1. Gullikson E. M. and Mills A. P. *Phys. Rev. Lett.* **57** 376 (1986)
2. e.g. Danielson, J. R.; Dubin, D. H. E.; Greaves, R. G.; Surko, C. M. *Reviews of Modern Physics.* **87** 247 (2015)

Positronium interactions with atoms and molecules

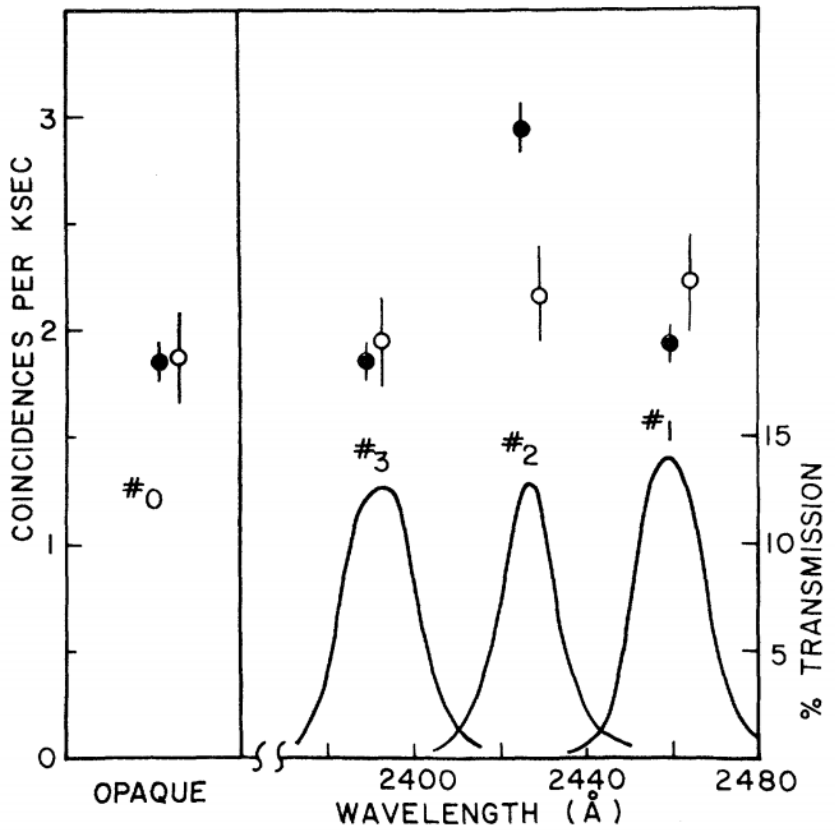
- Positronium scatters like an electron with the same velocity



1. S. J. Brawley, S. Armitage, J. Beale†, D. E. Leslie, A. I. Williams, G. Laricchia *Science* **330** 789 (2010)

Excited states of Positronium

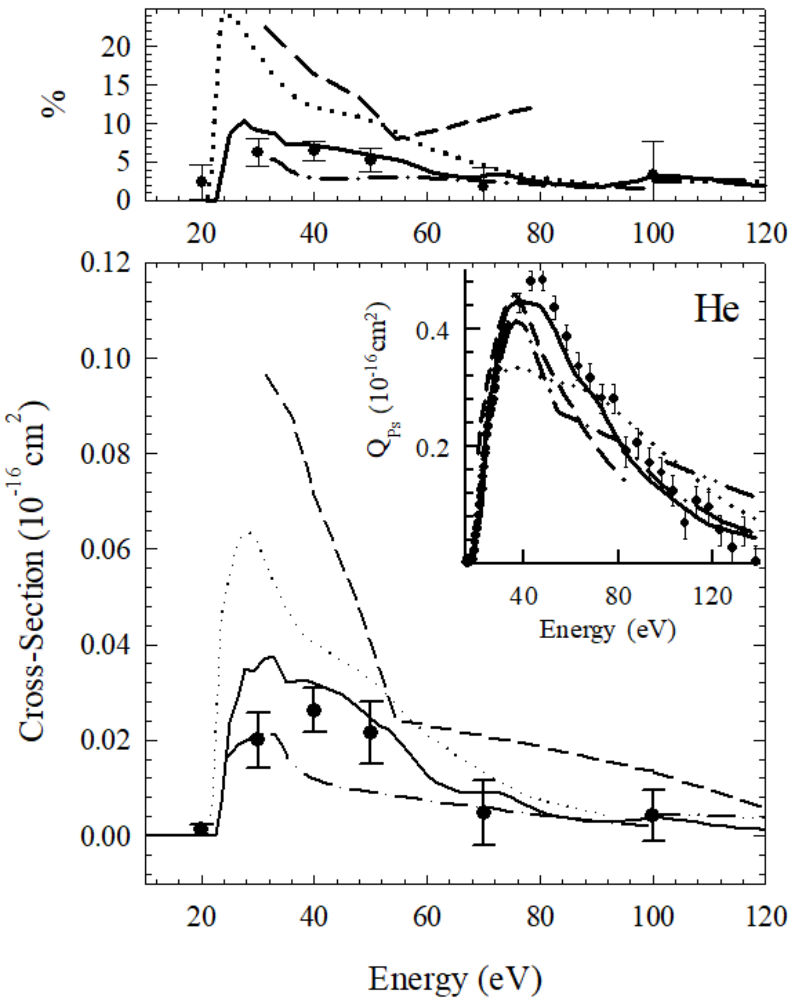
- The first observation of Ps formed with *n>1* was made by Canter et al in 1974
- Positrons were allowed to impinge on a Ge target
- A PMT was used to observe photons
- 3 different filters were placed in front of the PMT to isolate the 243nm light



1. K. F. Canter, A. P. Mills, Jr., and S. Berko Phys. Rev. Lett. **34**, 177 (1974)

Excited states of Positronium

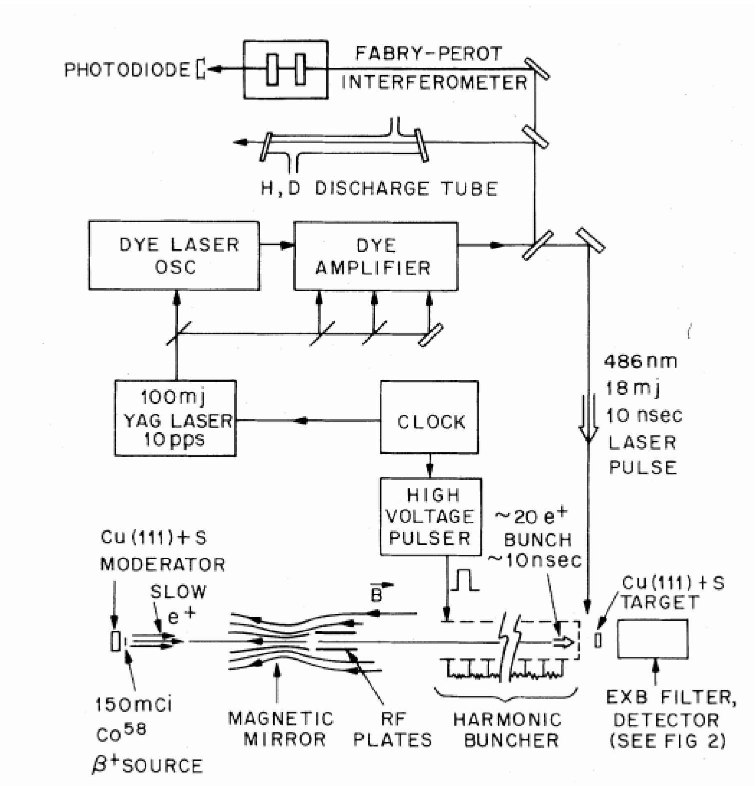
- First observation of Ps(n=2) in a gas target was made in 1985 by Laricchia et al
- The Ps(2P) formation cross-section for He, Ar and Xe have been measured.
- The fractional contribution was found to be
  - He - (6±1)%
  - Ar - (12±4)%
  - Xe - (26±9)%

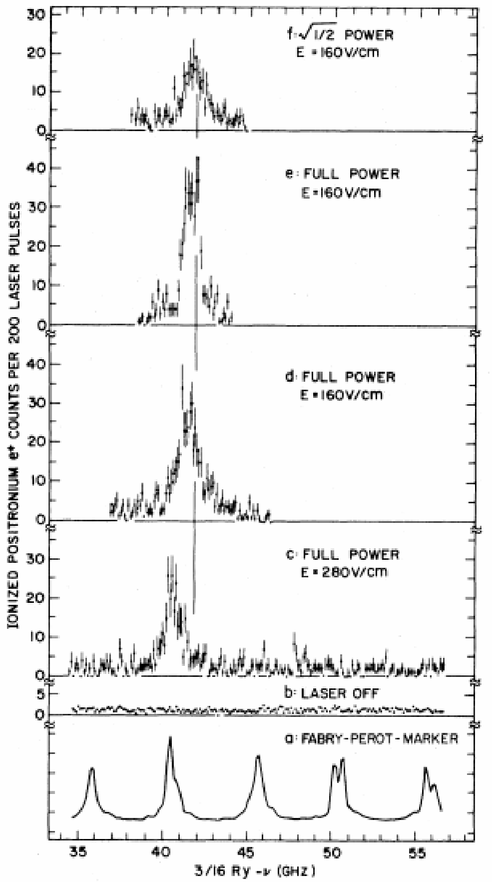


1. Laricchia G., Charlton M., Clark G. and Griffith T.C. *Phys. Lett.* **109A** 97 (1985)
2. D. J. Murtagh, D. A. Cooke, and G. Laricchia *Phys. Rev. Lett.* **102**, 133202 (2009)

Laser Interactions with Ps

- The first observed laser induced transition was by Chu and Mills in 1982
- A resonant ionization process was used to detect positrons from the laser excited Ps atoms

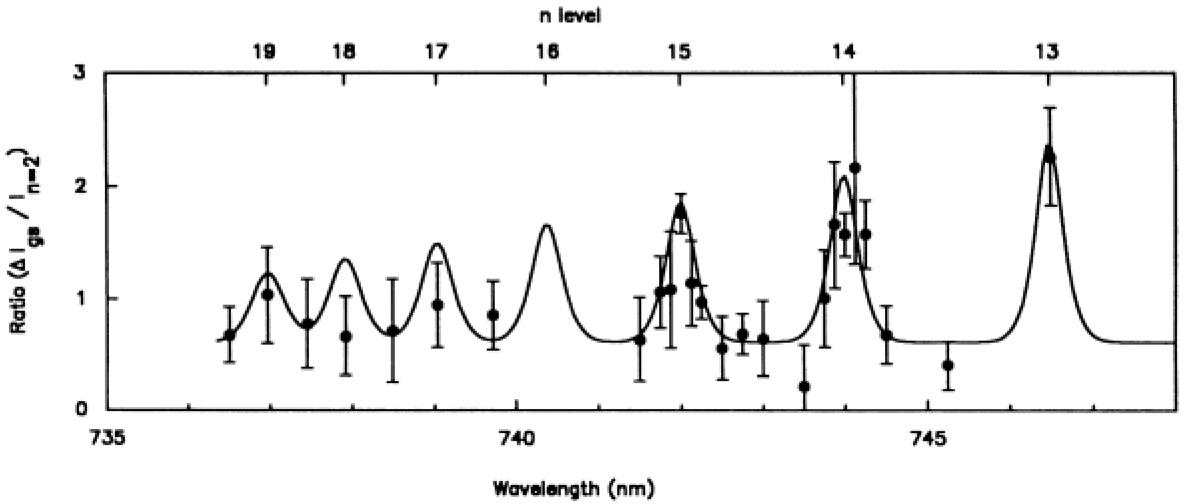




1. Steven Chu and Allen P. Mills, Jr. Phys. Rev. Lett. 48, 1333 (1982)

Highly excited positronium

- Rydberg state Ps was first observed in 1990 by Ziock *et al*
- Pulses of positrons from the Lawrence Livermore 100 MeV linac → 10^5 e^+ in 15 ns bunches
- 1s → 2p → 13*l*,14*l*,15*l* (highly excited states stark mixed)

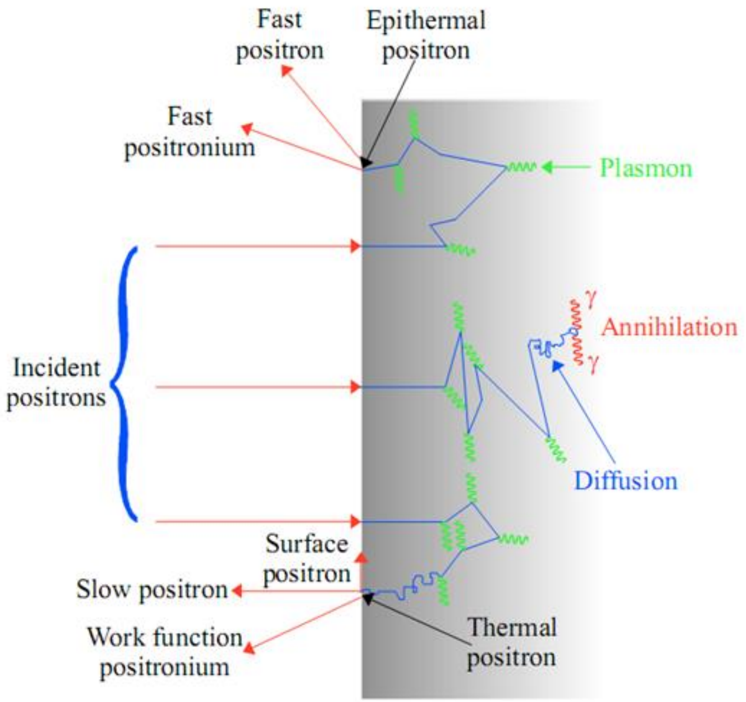


For an excellent review of the topic of Ry Ps see D. B. Cassidy *Eur. Phys. J. D.* **72** 53 (2018)

1. K.P. Ziock, R.H. Howell, F. Magnotta, R.A. Failor, K.M. Jones, *Phys. Rev. Lett.* **64**, 2366 (1990)

Efficient production of Ps

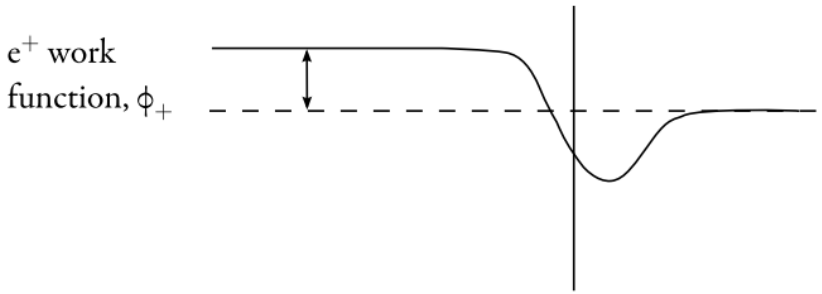
- Ps beam experiments make use of charge exchange collisions with gases
  - High energy Ps → okay for scattering not good for applications like optical excitation or charge exchange
- The previous optical excitation experiments made use of a single crystal Cu foils heated to 1000 K



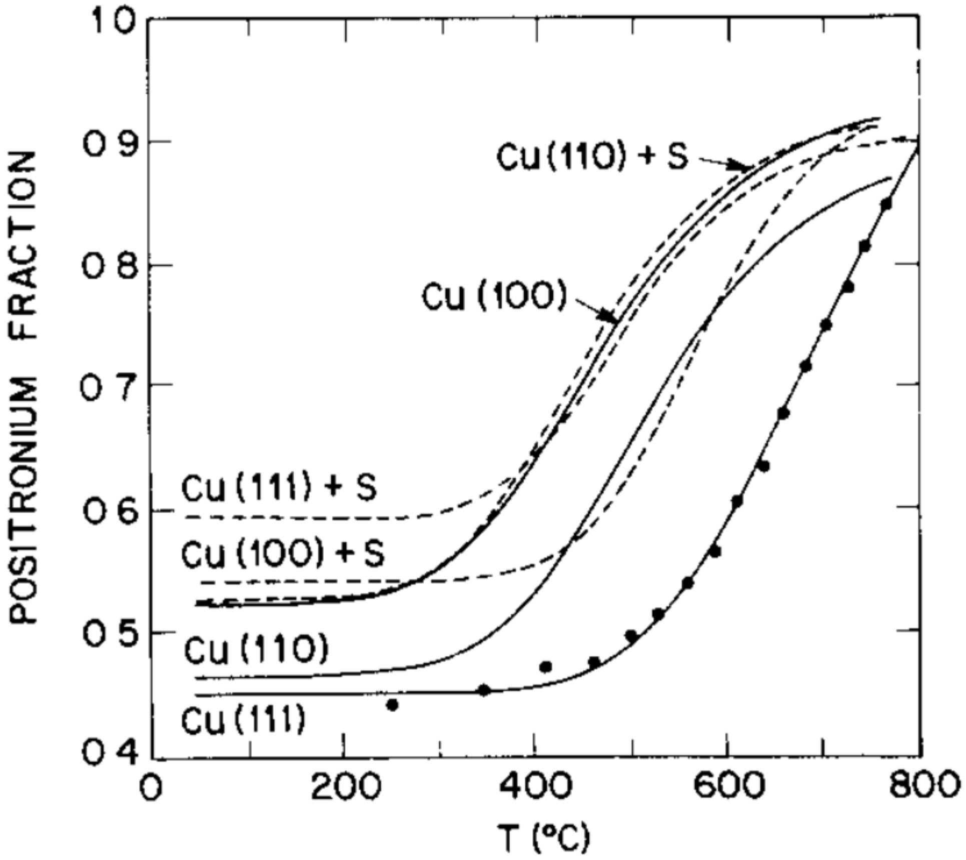
$$\phi_{Ps} = \phi_+ + \phi_- - E_b$$

Efficient production of Ps

- Positrons can become trapped at the surface of metals
- Positronium production fraction
  - $f_{Ps} = f_{dir} + f_t(T)$



$$E_a = E_t + \phi_- - E_B$$

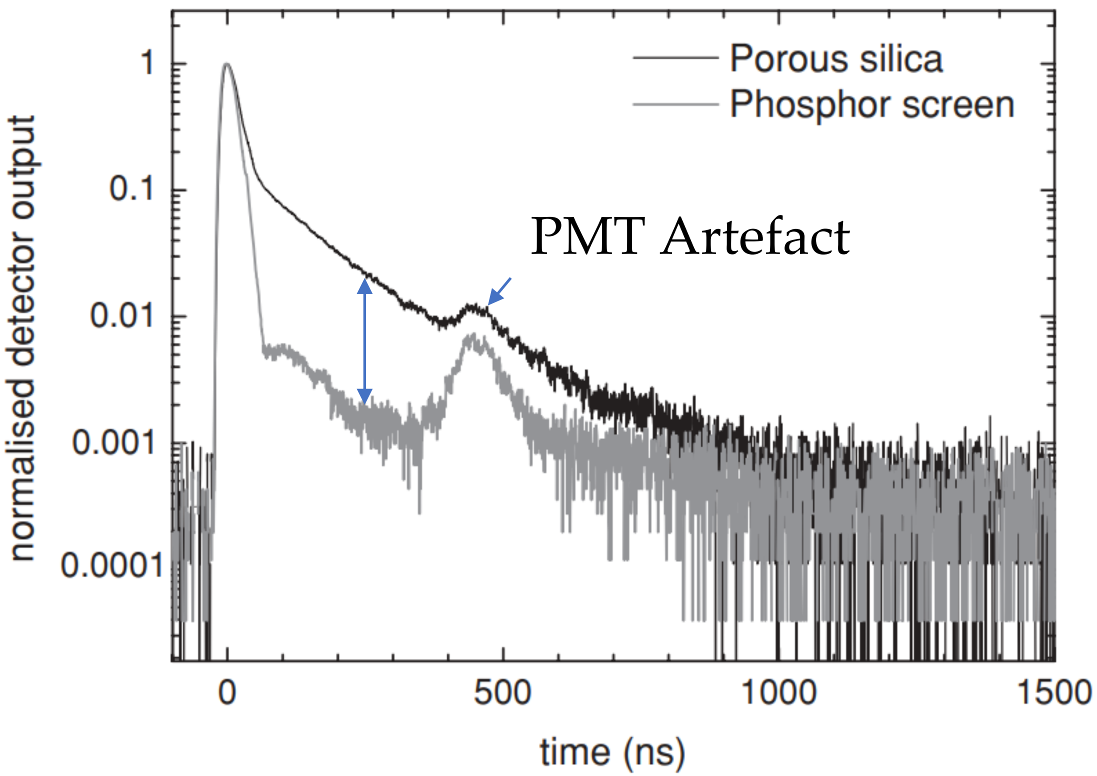


1. Robert Paulin, Roger Ripon, and Werner Brandt *Phys. Rev. Lett.* **31**, 1214 (1973)
2. Allen P. Mills *Solid State Comm*, **31** 623 (1979)

Efficient Ps production

- Thin mesoporous films have been used effectively as efficient Ps converters
- **S**ingle **S**hot **P**ositron **A**nnihilation **L**ifetime **S**pectroscopy (**SSPALS**)

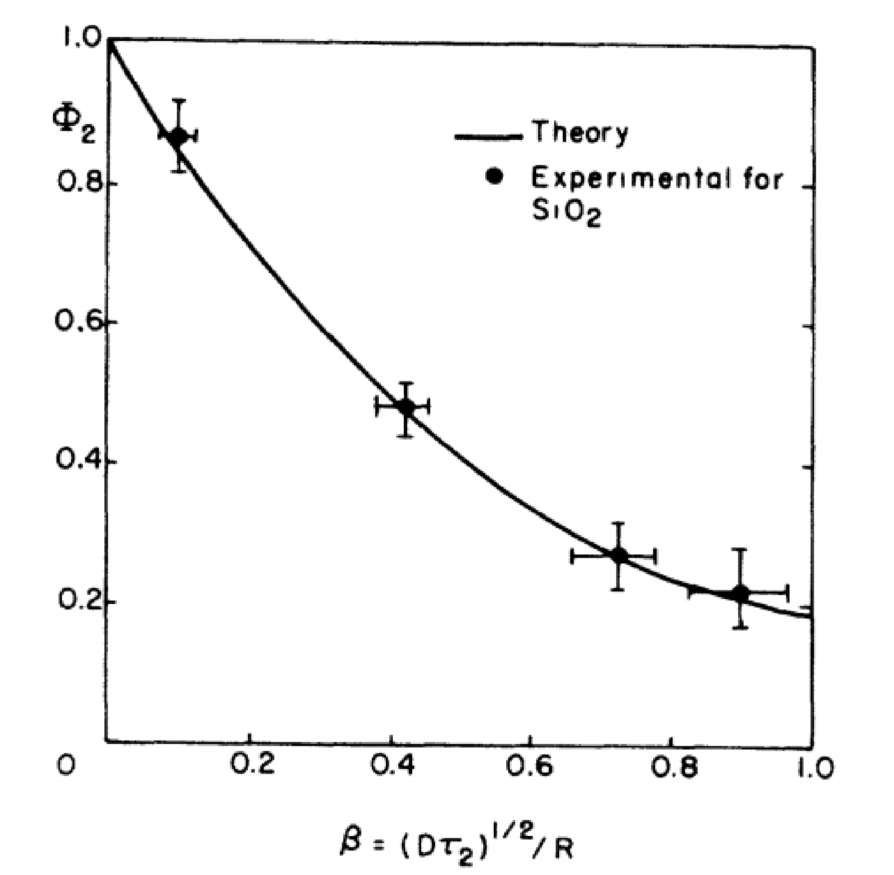
Ps - 1.5 keV
Background - 5 keV



1. E.g. D. B. Cassidy, P. Crivelli, T. H. Hisakado, L. Liszkay, V. E. Meligne, P. Perez, H. W. K. Tom, and A. P. Mills, Jr. *Phys. Rev. A* **81**, 012715 (2010)
2. See references in D. B. Cassidy *Eur. Phys. J. D.* **72** 53 (2018)

Efficient production of Ps

- Powders (e.g. MgO, SiO_2, Al_2O_3) can also efficiently form Ps
- Ps can also diffuse from the bulk in these materials and reach the surface to be emitted into vacuum



Diffusion Coefficient D = (5.8 $\pm$ 1.9) x 10^{-5} cm^2s^{-1}

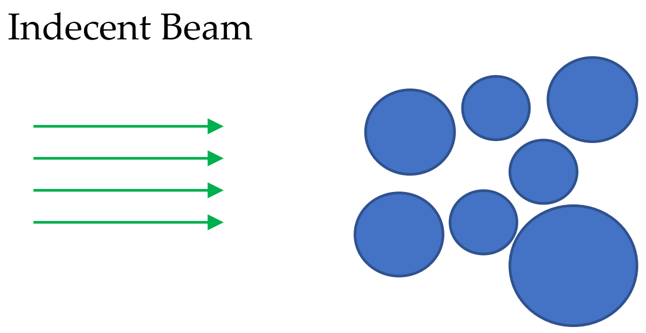
Radius R = 273, 76, 45, 36 A

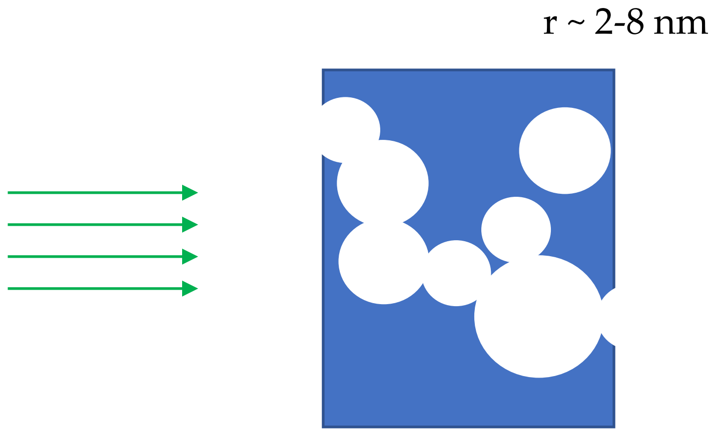
O-Ps in bulk τ_2 = (1.8 $\pm$ 0.2) ns

1. Werner Brandt and Robert Paulin *Phys. Rev. Lett.* **21**, 193 (1968)

Powders and Pours

- Ps can be formed in the bulk and on surfaces
  - $T_{bulk} = -\phi_+ - \phi_- + E_G - E_B + 6.8\ eV$
  - $T_S^v = -\phi_+ - \phi_- + 6.8\ eV$
  - $T_S^c = -\phi_+ - \phi_- + E_G + 6.8\ eV$
- Ps atoms formed on surfaces with thermal conduction electrons can penetrate into the bulk
- Ps formed in the bulk are confined within the voids





1. Y. Nagashima, Y. Morinaka, T. Kurihara, Y. Nagai, T. Hyodo, T. Shidara, and K. Nakahara *Phys. Rev. B* **58**, 12676 (1998)

Pours and trapping

- Positronium trapped in a cavity will cool via multiple collisions with the walls
  - Rate is dependent on pour size
  - $\Delta E = \frac{2m_{Ps}}{M}(\mathrm{E(t)} - \frac{3}{2}K_b T)$
  - Classical Model → okay for large pour size
- Quantum mechanical effects will begin to dominate when the de Broglie wavelength is close to the size of pours
  - $\lambda = h\sqrt{\frac{1}{2E_{Ps}m_{Ps}}}$
- As the wavelength approaches the pour size cooling stops
- Quantum tunnelling then dominates the diffusion of the Ps through the material
- Ps emitted to vacuum will convert this potential energy into KE

1. Saito H., Hyodo T. Cooling and Quenching of Positronium in Porous Material. In: Surko C.M., Gianturco F.A. (eds) *New Directions in Antimatter Chemistry and Physics*. Springer, Dordrecht (2001)
2. D. B. Cassidy, P. Crivelli, T. H. Hisakado, L. Liszkay, V. E. Meligne, P. Perez, H. W. K. Tom, and A. P. Mills, Jr. *Phys. Rev. A* **81**, 012715 (2010)

Dense Positronium Clouds

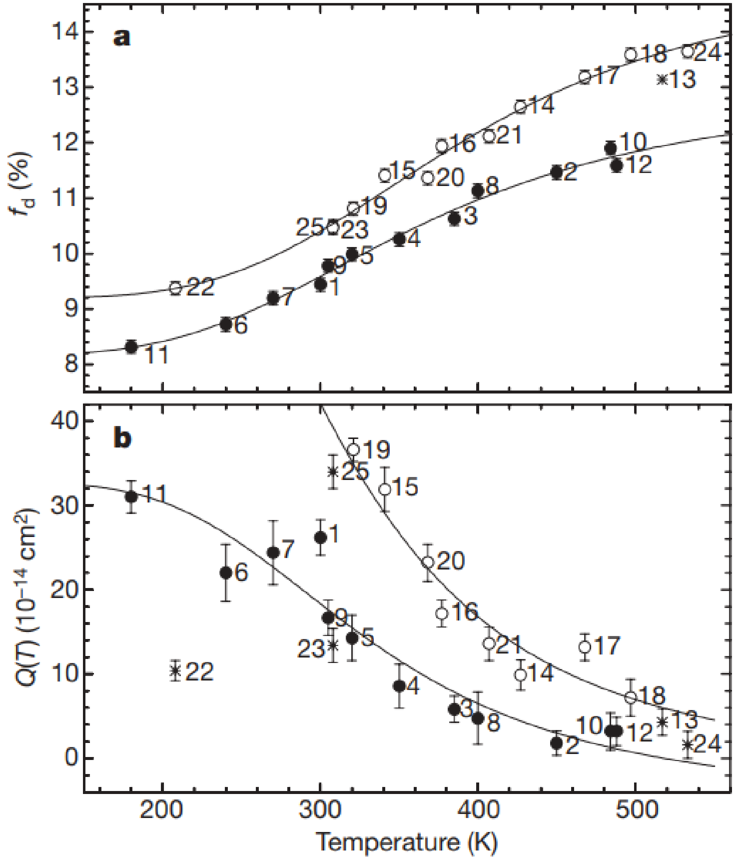
- If you can produce a dense enough positron plasma and have a good enough Ps converter you may be able to interact more than one Ps atom
  - Sub ns bunches
  - Areal density ~1x10^{10} cm^{-2}

- Elastic Scattering
  - $oPs + oPs \rightarrow oPs + oPs$
- Spin Exchange
  - $oPs + oPs \rightarrow 2pPs + 2E_{hfs}$
- Ps molecule formation
  - $X + oPs + oPs \rightarrow X + Ps_2 + E_b$
  - Three body recombination

1. D. B. Cassidy & A. P. Mills Jr Nature volume 449, pages195–197(2007)

Molecular Ps

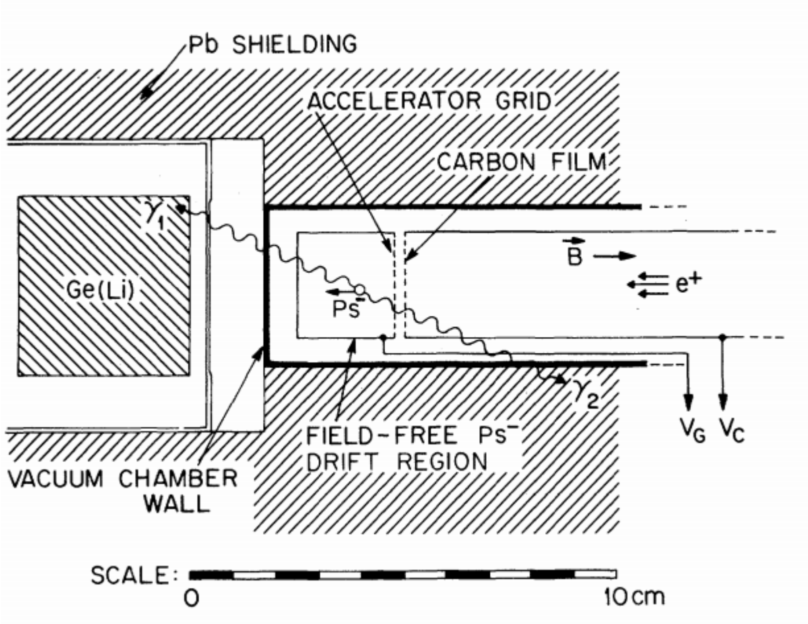
- Using SSPALS Cassidy and Mills looked for a signal from Ps_2 formation
- Both spin exchange and Ps_2 formation will cause oPs to annihilate more quickly
- This reduces the so called ‘delayed fraction’ (f_d)
- By increasing the temperature of the sample Ps atoms cannot bind to surfaces limiting the availability of a third body and hence suppressing Ps_2

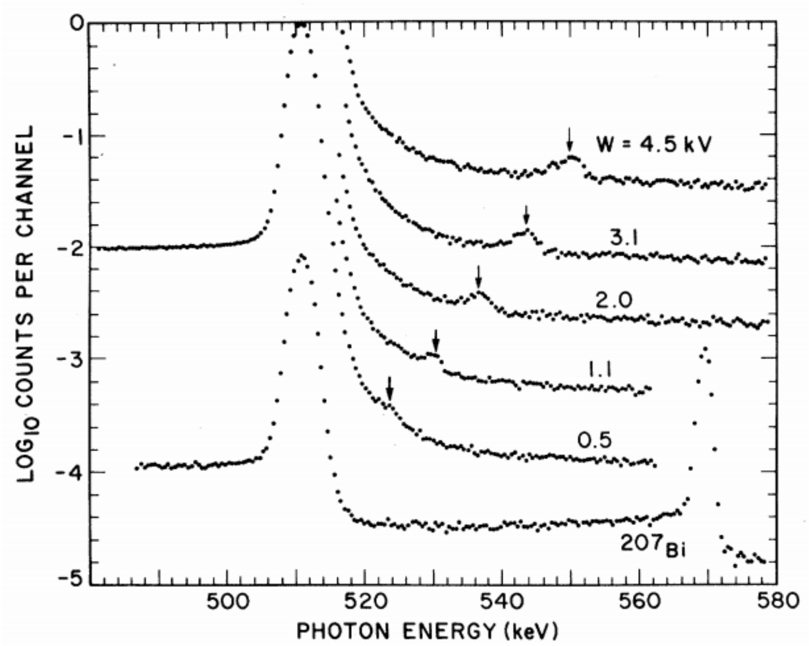


1. D. B. Cassidy & A. P. Mills Jr Nature volume 449, pages195–197(2007)

Observation of the Positronium Ion

- Mills observed a blue shifted annihilation line from Ps^- formed by passing a positron beam though a carbon foil ~50 A

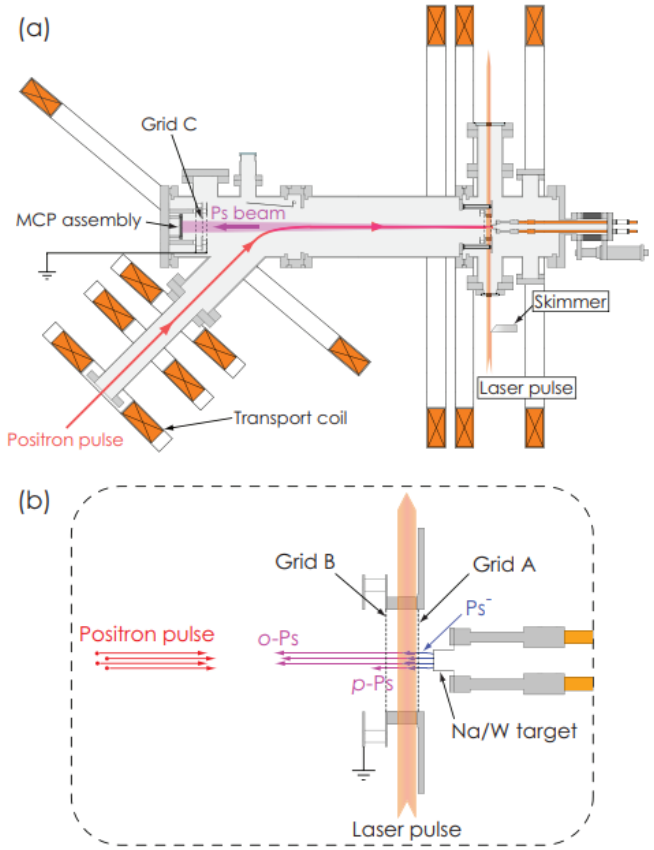




1. Allen P. Mills, Jr. *Phys. Rev. Lett.* **46**, 717 (1981)

Positronium Ion (negative)

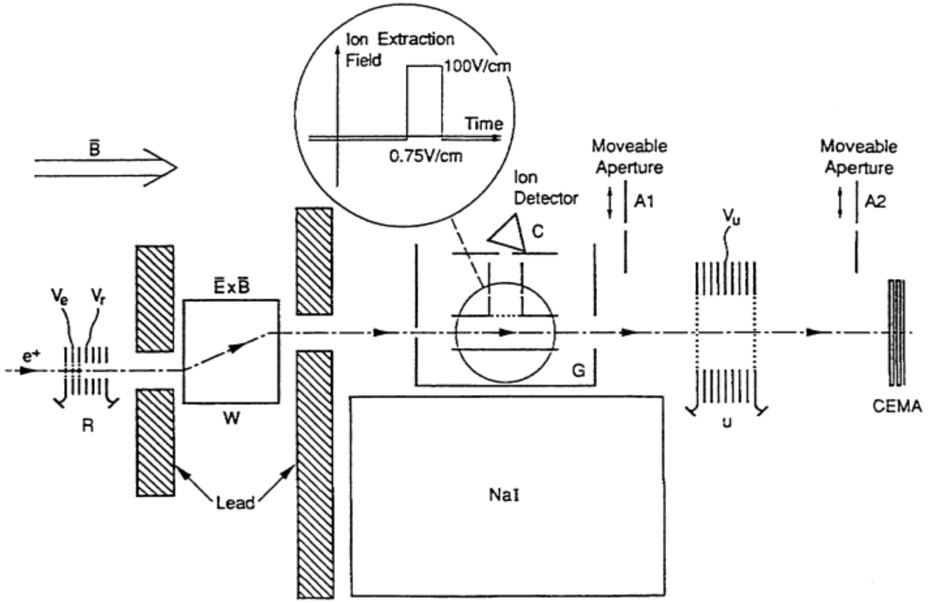
- Properties
  - Analog to the H^- ion
  - Binding energy ($Ps+e^-$) 0.33eV
  - Lifetime 479 ps
  - 2γ annihilation
- Conversion efficiency from C foil 0.028%
- Recently Nagashima and co-workers have produced efficient Ps-converters using Na coated W (1.5%)



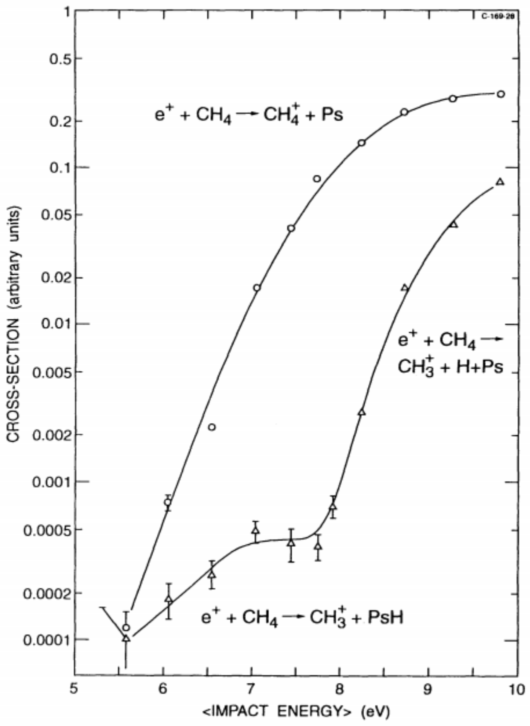
1. K. Michishio, T. Tachibana, R. H. Suzuki, K. Wada, A. Yagishita, T. Hyodo, and Y. Nagashima Appl. Phys. Lett. 100, 254102 (2012)

Positronium Hydride

- We have seen that Ps atoms behave much like H, forming Ps^- and Ps_2 what about binding to other atoms and molecules?



$$e^+ + CH_4 \rightarrow CH_4^+ + Ps, \quad 6.18\ \text{eV},$$
$$CH_3^+ + PsH, \quad 7.55\ \text{eV} - B_{PsH},$$
$$CH_3^+ + H + Ps, \quad 7.55\ \text{eV},$$
$$PsCH_3 + H^+, \quad 11.31\ \text{eV} - B_{PsCH_3},$$
$$Ps + CH_3 + H^+, \quad 11.31\ \text{eV}.$$



BE = (1.1±0.2) eV

1. D. M. Schrader, Finn M. Jacobsen, Niels-Peter Frandsen, and Ulrik Mikkelsen Phys. Rev. Lett. 69, 57 (1992)

Conclusion

- From the first prediction of Mohorovičić in 1934 the field of positronium physics has been an active and interesting.
- Positronium is the most common pathway for positron annihilation in vacuum and some solids.
- Of the so called 'polyelectrons' Wheeler predicted Ps, Ps^- and Ps_2 have been produced (Ps+ is more difficult)
- Positronium bound to atoms is an active field of research but at present only theory has been presented since the measurement of Schrader *et al*

Reviews

- S. Berko and N. Pendelton *Ann. Rev. Nucl. Part. Sci* 30 543 (1980)
- A. Rich *Rev Mod Phys* 35 127 (1981)
- M. Charlton and J. W. Humberston *'Positron Physics'* Cambridge University Press (2000)
- P. Coleman *'Positron Beams and their applications'* World Scientific (2000)
- G. Laricchia, S. Armitage, A. Kover, D. J. Murtagh *Ad. At. Mol. Opt. Phys.* **56** 1 (2008)
- D. B. Cassidy *Euro J. Phys. D.* 71 **72** (2018)