Interaction of Positronium with Atoms and Molecules

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Outline

1. Positronium (Ps)
2. Formation of Ps and Monoenergetic Ps beam experiment
3. Ps experiments using silica aerogel
4. Momentum transfer cross section and thermalization of Ps
5. Pick-off quenching
6. Spin conversion through electron exchange
7. Spin conversion through spin-orbit interaction
8. Chemical quenching
Wave function of Ps

\[ 2^{S+1} \Phi_M (\vec{r}_p, \vec{r}_e) = | S \rangle M \]

Space Part

\[ \phi(\vec{r}_p, \vec{r}_e) = \frac{e^{-r/2a_0} \ e^{i\vec{K} \cdot \vec{R}}}{\sqrt{8\pi a_0^3} \ \sqrt{V}} \]

\[ \vec{r} = \vec{r}_e - \vec{r}_p, \quad \vec{R} = \frac{\vec{r}_e + \vec{r}_p}{2} \]

\[ a_0 : \text{Bohr radius,} \]

\[ \vec{K} : \text{translational wave vector} \]
Geometrical similarity and difference between H and Ps
Spin part

$$\chi = |SM>$$

$$|11> = |\uparrow\uparrow>$$

$$|1-1> = |\downarrow\downarrow>$$

$$|10> = \frac{1}{\sqrt{2}}(|\uparrow\downarrow> + |\downarrow\uparrow>)$$

$$|00> = \frac{1}{\sqrt{2}}(|\uparrow\downarrow> - |\downarrow\uparrow>)$$
ortho-Ps (o-Ps) and para-Ps (p-Ps)

\[ \chi = |SM > \]

- S=1 (triplet: \( |1 \pm 1 > \) and \( |10 > \)) : o-Ps
  self-annihilates into 3 \( \gamma \)
  lifetime 142ns

- S=0 (singlet: \( |00 > \)) : p-Ps
  self-annihilates into 2 \( \gamma \)
  lifetime 125ps
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Ps beam apparatus

Fig. 2. A schematic diagram of the Ps beam at UCL.

\[ e^+ + M \rightarrow Ps + M^+ \]

Formation of Ps

\[ e^+ + M \rightarrow Ps + M^+ \]

\[ E_{Ps} = E_+ - I_M + 6.8 \text{eV} / n^2 \]

- \( E_{Ps} \) Kinetic energy of Ps
- \( E_+ \) Kinetic energy of incident positron
- \( I_M \) Ionization energy of M
- \( 6.8 \text{eV} / n^2 \) Ionization Energy of Ps in the state with quantum number \( n \)
Total cross section $\sigma_T$

$$I = I_0 e^{-\sigma_T n L} = I_0 e^{-\sigma_T \frac{p}{k_B T} L} \rightarrow \sigma_T = \frac{k_B T}{p L} \ln \left( \frac{I_0}{I} \right)$$

$I_0$  Incident beam intensity  
$I$  Transmitted beam intensity  
$n$  Number density of the target gas  
$p$  Pressure  
$T$  Temperature  
$k_B$  Boltzmann constant  
$L$  Effective interaction length
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Silica aerogel – Ps formation medium and micro-chamber for Ps-Molecule Interactions
Details around source-specimen
Condition for Ps formation in gases

\[ E_+ > I_M - 6.8 \text{eV} \]

• In a situation where e\(^+\) and Ps can collide with molecules many times:
  \[ E_+ < E_{\text{ex}} \]
  Otherwise, excitation of the molecules is likely.

\[ I_M - 6.8 \text{eV} < E_+ < E_{\text{ex}} : \text{Ore gap} \]

• Ps formation fraction \( F \):
  \[ \frac{E_{\text{ex}} - (I_M - 6.8 \text{eV})}{I_M} < F < \frac{6.8 \text{eV}}{I_M} \]
Ps formation in insulator solids

• Ore process in solids

\[ E_G - E_B < E_+ < E_{ex} : \text{Ore gap in solid} \]

\[ E_G : \text{band gap} \]

\[ E_B : \text{binding energy of Ps in the solid} \]

• Spur (blob) process

Spur: a region containing excited (free) electrons and ions, created around the point where the positron deposit part of its energy during slowing down.

The positron can be bound with one of the free electrons in the terminal spur to form Ps.
Ps emission from insulator surface

• Ps is quickly thermalized regardless of the formation process.
• It is emitted after thermalization if work function
  \[ \phi_{Ps} = \phi_- + \phi_+ - E_G + E_B - 6.8\text{eV} \]
  is negative
• The emission energy is
  \[ E_{Ps}^{\text{Bulk}} = -\phi_{Ps} \]

\( \phi_- \) electron work function
\( \phi_+ \) positron work function
Emission of Ps formed on the insulator surface

• Ps formed on the surface is emitted immediately with energy

\[ E_{Ps}^{surface} = -\phi_- - \phi_+ + E_G + 6.8 \text{eV} \]

cf.

\[ E_{Ps}^{Bulk} = -\phi_- - \phi_+ + E_G - E_B + 6.8 \text{eV} \]
Time-of-flight spectra for Ps emission from SiO$_2$ surface.

Two components from SiO$_2$.

(Nagashima et al. PRB)
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Momentum transfer cross section

\[ \sigma_m = \int (1 - \cos \theta) \frac{d\sigma_{el}}{d\Omega} \, d\Omega \]

\( \sigma_{el} \) elastic cross section

As \( E_{Ps} \to 0 \), \( \sigma_T = \sigma_{el} = \sigma_m \)
Spin Averaged **Positron** Annihilation Cross Section (Dirac)

**2 \( \gamma \)** annihilation:

\[
\sigma_{2\gamma} = \pi r_0^2 c / v \\
\lambda_{2\gamma} = \rho_{av} \sigma_{2\gamma} v = \pi r_0^2 c \rho_{av}
\]

\( \rho_{av} \) = spin averaged electron density at the positron

\[
\lambda^+_{\text{gas}} = \pi r_0^2 c \bar{\rho} = \pi r_0^2 c n Z_{\text{eff}} \quad \rightarrow \quad Z_{\text{eff}} = \frac{\lambda^+_{\text{gas}}}{\pi r_0^2 c n}
\]

**3 \( \gamma \)** annihilation:

\[
\sigma_{3\gamma} = \frac{4}{3} (\pi^2 - 9) \alpha r_0^2 c / v
\]

\[
\lambda_{3\gamma} = \rho_{av} \sigma_{3\gamma} v = \frac{4}{3} (\pi^2 - 9) \alpha r_0^2 c \rho_{av} \approx \frac{\lambda_{2\gamma}}{370}
\]
Positronium (Ps)

electron density at $e^+$ in Ps: $\rho_{\text{Ps}} = |\phi(r, r)|^2 = \frac{1}{8\pi a_0^3}$

p-Ps: annihilates into $2\gamma$

$$\lambda_{p-\text{Ps}} = 4\pi r_0^2 c \rho_{\text{Ps}} = \frac{\alpha^5 mc^2}{2\hbar} = (125 \text{ps})^{-1}$$

o-Ps: annihilates into $3\gamma$

$$\lambda_{o-\text{Ps}} = \frac{4}{3} \frac{4}{3} (\pi^2 - 9) \alpha r_0^2 c \rho_{\text{Ps}} = \frac{2(\pi^2 - 9)\alpha^6 mc^2}{9\pi \hbar}$$

$$\approx \frac{\lambda_{p-\text{Ps}}}{1160} \approx (142 \text{ns})^{-1}$$
Ps in a magnetic field

- Magnetic field $B$

$$| - > = \frac{-y}{\sqrt{1+y^2}} | 10 > + \frac{1}{\sqrt{1+y^2}} | 00 >$$

$$| + > = \frac{1}{\sqrt{1+y^2}} | 10 > + \frac{y}{\sqrt{1+y^2}} | 00 >$$

$$| 11 >$$

$$| 1-1 >$$

$$y = \frac{x}{\sqrt{1+x^2} + 1}, \quad x = \frac{4\mu_B B}{\Delta E}$$
Thermalization and $\sigma_m$

energy loss per collision

$$\Delta E = \frac{2m_{Ps}}{M} (E(t) - \frac{3}{2} k_B T)$$

$$\frac{dE_{av}(t)}{dt} = -\sigma_m n v_{Ps} \times \Delta E = -\frac{2\sigma_m n}{M} \sqrt{2m_{Ps} E_{av}(t) (E_{av}(t) - \frac{3}{2} k_B T)}$$

$$E(t) = \frac{1 + \alpha \cdot \exp(-\beta t)}{1 - \alpha \cdot \exp(-\beta t)} \cdot k_B T$$

$$\alpha = \frac{\sqrt{E(0)} - \sqrt{3k_B T / 2}}{\sqrt{E(0) + \sqrt{3k_B T / 2}}}$$

$$\beta = \frac{2\sigma_m n}{M} \sqrt{3m_{Ps} k_B T}$$

Thermalization due to silica grain surface collision rate

$$\sigma_m n v_{Ps} \rightarrow \frac{v_{Ps}}{L}$$
Thermalization with ACAR

Vacuum
(silica aerogel alone)

He

Ps thermalization

Vacuum
(silica aerogel alone)

He

![Graph showing the thermalization of Ps in vacuum and He.](image-url)
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Quenching of o-Ps

\[ \lambda_{\text{total}} = \lambda_{\text{o-Ps}} + \lambda_{\text{quench}} \]

\[ \lambda_{\text{quench}} = \lambda_{\text{o-Ps}} + \lambda_{\text{pick-off}} + \lambda_{\text{ex}} + \lambda_{\text{chem}} + \lambda_{\text{spin-orbit}} + \cdots \]

- Quenching of o-Ps
  - Electron pick-off without converting into p-Ps
  - Spin-conversion into p-Ps due to electron exchange
  - Spin-conversion into p-Ps due to spin-orbit Interaction
  - Chemical quenching
  - (o-Ps) – (o-Ps) quenching
  - …….
Detection of o-Ps Quenching

• Positron lifetime technique:
  – Shortening of the o-Ps lifetime
  – Applicable to all kinds of quenching

• Momentum distribution:
  (Angular Correlation of Annihilation $\gamma$ s, or Doppler Broadening of Annihilation $\gamma$
  – Insensitive to pick-off:
    broad component similar to $e^+$ annihilation
  – Applicable to conversion to p-Ps $\rightarrow$ narrow component
    (thermal center-of-mass momentum of p-Ps)

• Age-Momentum correlation technique
  – Time resolved momentum distribution
Pick-off quenching and $^1Z_{\text{eff}}$

$$\lambda_{\text{pick-off}} = 4\pi r_0^2 c \, ^1\overline{\rho}$$

($^1\overline{\rho}$ : temporally and spatially coarse-grained density of the outer molecular electrons whose spin is singlet to that of the Ps positron )

$$= 4\pi r_0^2 c n \, ^1Z_{\text{eff}} \quad \rightarrow \quad ^1Z_{\text{eff}} = \frac{\lambda_{\text{pick-off}}}{4\pi r_0^2 c n}$$

$$= 4\pi r_0^2 c \frac{\Delta t}{\tau} \, ^1\rho_{\text{on-molecule}}$$
$^1Z_{\text{eff}}$ in time-independent approach

$$\Psi(\vec{r}_1, s_1; \vec{r}_2, s_2; \ldots; \vec{r}_p, s_p)$$

$$\Phi_i(\vec{r}_1, s_1; \vec{r}_2, s_2; \ldots; \vec{r}_i; \ldots; \vec{r}_p) = \langle ^1\chi_0 (s_p, s_i) | \Psi \rangle$$

$$^1Z_{\text{eff}} = \sum_i \sum_{s(\neq s_p, s_i)} d\vec{r}_1^3 d\vec{r}_2^3 \ldots d\vec{r}_p^3$$

$$\times \delta(\vec{r}_i - \vec{r}_p) | \Phi_i(\vec{r}_1, s_1; \vec{r}_2, s_2; \ldots; \vec{r}_i; \ldots; \vec{r}_p) |^2$$
$Z_{\text{eff}}$
Geometric cross sections

\[ \frac{\lambda_q}{4\pi\sigma_0^2 cn} \]
$^{1}Z_{\text{eff}}$ in inert gases (Exp. and Theory)
Low vapor pressure gases

• CH$_3$F, CH$_3$Cl, CH$_3$Br, CH$_3$I
• (CH$_4$ for reference)
Gas handling system

C1, C2, C3: chamber
C4: Cylinder
M1, M2: ceramic capacitance manometer
G1: pirani gauge + cold cathode ionization gauge
TMP: Turbomolecular pump
☒: valve
Analysis of positron lifetime data
Extrapolation to no silica grain limit
$^{1}Z_{\text{eff}}$ for inert gases and halogenized methane
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Spin-conversion into p-Ps due to electron exchange with a molecule having unpaired electron

\[
\lambda_{ex} = n V \sigma_{el-ex-conv} \\
(= 4 \pi r_0^2 c n^1 Z_{eff})
\]
Ps-O$_2$ interaction


Figure 2. Angular correlation of annihilation radiation from silica aerogel in vacuum (a) and in 1 atm N$_2$ (b) and various pressures of O$_2$ ((c), 0.1 atm; (d), 0.2 atm; (e), 0.4 atm; (f), 0.8 atm).
AMOC systems
(Age-Momentum Correlation)

Example of AMOC data (metal)
o-Ps Quenching due to electron exchange with $O_2$, which is triplet

Doppler broadening spectrum
Pickoff and spin conversion in $\text{O}_2$

- $\lambda_{\text{SiO}_2} = 0.37 \pm 0.04 \mu s^{-1}$

- $\lambda_{\text{ox}} + \lambda_{\text{spin}} = 25.1 \pm 0.2 \mu s^{-1}$

- \[
\frac{I_{\text{broad}}}{I_{\text{narrow}}} = \frac{I_{\text{SiO}_2} + I_{\text{ox}}}{I_{\text{spin}}} = \frac{\lambda_{\text{SiO}_2} + \lambda_{\text{ox}}}{\lambda_{\text{spin}}}
\]

- $\lambda_{\text{spin}} = 24.7 \pm 0.2 \mu s^{-1}$  \quad $\lambda_{\text{ox}} = 0.4 \pm 0.3 \mu s^{-1}$

- $^1Z_{\text{eff}} = 06 \pm 0.4$  \quad $\sigma_{\text{spin}} = (1.16 \pm 0.01) \times 10^{-19} \text{ cm}^2$
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Spin-orbit quenching

\[ \text{o-Ps} + \text{Xe} \rightarrow \text{p-Ps} + \text{Xe} \rightarrow 2\gamma + \text{Xe} \]

Proposed to explain “Xe puzzle of Ps formation”

(Mitroy and Novikov: PRL 90, 183202 (2003))

Confirmed by using Ps lifetime measurements in a magnetic field

(Saito and Hyodo: PRL 97, 243502 (2006))
Positron lifetime spectrum in Xe

so-called “Xe puzzle”

• Intensity of long-lived Ps is very low for Xe.

• Fast components are observed.

“Xe (and Kr) puzzle” in Ps formation probability
Mitroy’s solution for “Xe Problem”

• Spin-orbit interaction induces Ps spin transition → quenching (o-Ps → p-Ps → 2γ).

• Initial Ps energy is relatively high (>1eV)
  → strong spin-orbit interaction
  → extra fast component due to quenching

• After o-Ps is thermalized (energy ~0.1eV)
  → weak and constant spin-orbit interaction
  → week but well-defined long-lived component from surviving Ps
Prediction of Mitroy and Novikov.
Spin-Orbit Interaction

\[
V_{SO} = \alpha^2 \frac{1}{r_p} \frac{dV_p}{dr_p} \vec{l}_p \cdot \vec{s}_p + \alpha^2 \frac{1}{r_e} \frac{dV_e}{dr_e} \vec{l}_e \cdot \vec{s}_e
\]

\[
\Psi(S, M, \tilde{k}) = \sum_{LM} i^L \phi(r) Y_{00}(\vec{r}) \Phi_L(k, R) Y_{LM}(\vec{R}) \times Y_{LM}^*(\vec{k}) \chi(SM)
\]

Conservation Rules

Total angular momentum: \( \vec{J} = \vec{L} + \vec{S} \)

Parity: \( \Pi = (-1)^L \)
Allowed transitions in spin-orbit interaction

\[ T_{L',L}^{S,S'} : \text{T matrix for} \]

\((L, S)\): spin \(S\) state Ps in \(L\) partial wave

\[ \rightarrow (L', S') \]
Selection rules for spin-orbit scattering

p-Ps $\leftrightarrow$ p-Ps: $T_{LL}^{00} \neq 0$ only for $L' = L$, since $\bar{J}' = \bar{J} = \bar{L}$ must be conserved.

o-Ps $\leftrightarrow$ p-Ps

$T_{00}^{10} = 0$ since $\bar{J}$ is not conserved.

$T_{L'L}^{01}(L \geq 1)$ can be finite since $\bar{J} = \bar{J}' = \bar{L}$ may be satisfied.

$(T_{L \pm 1L}^{S'S} = 0$ since parity is not conserved.)
Selection rules for SO scattering

\[ o-Ps \iff o-Ps: \quad T_{LL}^{11} \neq 0 \quad \text{if} \quad J_z' = J_z, \]

but no effect on the experimental data.

In a magnetic field

\[ |11\rangle, \quad |1-1\rangle \iff |+\rangle, \quad |-> \quad \text{is allowed.} \]

Effect of \[ |11\rangle, \quad |1-1\rangle \iff |-> \quad \text{is appreciable because the lifetime of} \quad |-> \quad \text{is much shorter than} \quad |10\rangle. \]
spin-transition due to Xe among positronium states in a magnetic field

- When the magnetic field is increased, the lifetime of $|+\rangle$ becomes shorter, and the transitions of $|11\rangle$ and $|1-1\rangle$ to $|+\rangle$ reduce the lifetime of the former.
Digital Positron Lifetime Measurement System

Diagram:
- PMT3
- BaF$_2$ 28° × 10
- γ-ray source
- PMT1
- BaF$_2$ 28° × 10
- PMT2
- FET Voltage Follower
- digital oscilloscope Wavepro 960
- trigger
- GPIB
- Personal computer

Diagrams:
- Discs
- Coincidence
H3378, -2900V
BaF₂ 28 φ × 10mm

Voltage (output of ADC)

Time (ns)

4GS/s
250ps/point
in vacuum

Normalized counts

Channel number (1ch=1ns)

(B=1T)/I(B=0T)

(B=1T)

(B=0T)

Normalized counts

Channel number (1ch=1ns)
in Xe 1atm

Channel number (1ch=1ns)

$I(B=1T)/I(B=0T)$
### o-Ps lifetime in gases

<table>
<thead>
<tr>
<th></th>
<th>pressure /atm</th>
<th>B=0T</th>
<th>B=1.0T</th>
</tr>
</thead>
<tbody>
<tr>
<td>vacuum</td>
<td></td>
<td>132.9 ± 0.5 ns</td>
<td>131.4 ± 0.6 ns</td>
</tr>
<tr>
<td>Xe</td>
<td>1.5</td>
<td>112.1 ± 0.5 ns</td>
<td>102.1 ± 0.6 ns</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>118.4 ± 0.5 ns</td>
<td>109.0 ± 0.6 ns</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>123.6 ± 0.6 ns</td>
<td>119.1 ± 0.9 ns</td>
</tr>
<tr>
<td>Kr</td>
<td>2.5</td>
<td>119.5 ± 0.3 ns</td>
<td>115.7 ± 0.4 ns</td>
</tr>
<tr>
<td>Ar</td>
<td>2.5</td>
<td>123.9 ± 0.7 ns</td>
<td>124.6 ± 1.0 ns</td>
</tr>
</tbody>
</table>

*spin-transition due to spin-orbit interaction.*
\[ \lambda_q = (4\pi r_0^2 c)n_A \, {}^1Z_{\text{eff}} \]

\[ {}^1Z_{\text{eff}} \text{ determined from} \]

<table>
<thead>
<tr>
<th></th>
<th>( {}^1Z_{\text{eff}} ) (pick-off)</th>
<th>( {}^1Z_{\text{eff}} ) (spin-orbit)</th>
<th>( {}^1Z_{\text{eff}} ) (total)</th>
<th>( {}^1Z_{\text{eff}} ) * (total)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe</td>
<td>0.48 ± 0.10</td>
<td>0.77 ± 0.10</td>
<td>1.25 ± 0.04</td>
<td>1.25</td>
</tr>
<tr>
<td>Kr</td>
<td>0.34 ± 0.10</td>
<td>0.12 ± 0.10</td>
<td>0.48 ± 0.10</td>
<td>0.478</td>
</tr>
<tr>
<td>Ar</td>
<td>0.35 ± 0.10</td>
<td>−0.05 ± 0.10</td>
<td>0.30 ± 0.03</td>
<td>0.314</td>
</tr>
</tbody>
</table>

* M. Charlton, 
48, 737 (1985).
\( {^1Z_{\text{eff}}(\text{total})} \) is decomposed into \( {^1Z_{\text{eff}}(\text{spin-orbit})} \) and \( {^1Z_{\text{eff}}(\text{pickoff})} \).
atomic number dependence

Normalized $^{1}\text{Zeff}$ (spin-orbit)

Normalized $^{1}\text{Zeff}$ (spin-orbit)

atomic number (Z)
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Chemical Quenching  
(formation of resonances)

\[ \lambda_{\text{chem}} \quad \left( = 4\pi r_0^2 cn^1 Z_{\text{eff}} \right) \]

\[ = nV\sigma_{\text{res}} \]

\[ \approx 4\pi r_0^2 c^{1-}\bar{\rho}_{\text{on-molecule}} \quad \text{(for large } n) \]

NO_2: \[ \lambda_{\text{chem}} \] saturates at about 0.1 amagat

\[ ^1 Z_{\text{eff}} \approx \frac{1-\bar{\rho}_{\text{on-molecule}}}{n} \approx \frac{1/10^{-30}}{0.1\times3\times10^{25}} \approx 3\times10^6 \]
Chemical Quenching of o-Ps in NO$_2$